PHASE-RETRIEVAL METHOD

AND

APPLICATIONS

Volume I

Fundamentals and X-Ray Crystallography

Pui Sum Yuen

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Volume I

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Preface

The magnitude of the wave diffracted from an object can be measured, but not the phase. If the phase is known, the structure of the object can be determined. By applying general and reasonable constraints on the object, the phase can be obtained from the magnitude. The solution is unique or very probably unique. The phase is not lost; it is just hidden in the magnitude.

Phase-retrieval approach has numerous applications. The structure of all molecules in a human body is essential to understanding biological processes, diseases and developing suitable medicine. In fact, many disciplines involve wave or particle scattered from an object, phase-retrieval approach will be useful.

I have completed three articles about basic principles of phase-retrieval method, and four articles about applications to optical scattering, acoustical scattering, thermal image, seismology, earthquake, tsunami and nuclear physics. Instead of publishing them in journals, I choose to publish a book so that the publication will not be delayed. I think the method and the applications are very important. The sooner they are published the better. If they are published one day earlier, many more lives will be saved. These articles are not peer-reviewed. Because of my very limited knowledge, there are bounded to be many errors. I would be very grateful if readers of this book can inform me of the errors (use the Forum in this website).

As a researcher, I hope my discovery will benefit all mankind. In fact, it is The Lord’s grace through an unworthy person.

> I declare that now I give the intellectual property of all my discoveries in this book to all mankind including those born in future. The intellectual properties of other people are not affected.

I hope no one will use my discoveries in this book to apply for any patent.

Pui Sum Yuen
December 19, 2007

Preface to the 2011 Edition

Since the book first published in 2007, it has many revisions. Articles are deleted and added. It is now divided into two volumes.

Volume I. Fundamentals and X-ray Crystallography. A systematic development of fundamentals of phase-retrieval method is presented. It provides the theoretical foundation and gives the conditions that iterative algorithms with oversampling will give a unique or probably unique solution. Methods in X-ray crystallography, with and without using in-between｜Fourier coefficients｜, are presented. These are applied to structure determination of crystals, nonperiodic objects, fibres and planar periodic membranes.

Volume II. Applications: Optics and Miscellaneous. It presents imaging of object irradiated with quasi-monochromatic coherent, incoherent, or partially coherent waves. The direct and inverse problems are dealt with. Unique decomposition of quasi-monochromatic partially coherent waves into coherent and incoherent components is presented. Imaging with partially coherent wave can then be dealt with in terms of the coherent and incoherent components. The direct and inverse problems of imaging of object emitting polychromatic coherent, incoherent or partially coherent waves are presented. For imaging of object emitting polychromatic partially coherent wave, coherent and incoherent components may be employed. Because of the small size of the nuclei, phase-retrieval method is meaningful. An article is presented for determining nuclear structure and force by means of phase retrieval in scattering.

For readers to inform me of the errors of this book, or suggestions, comments and criticisms, please use e-mail address: puisumyuen@netvigator.com . Thanks.

Pui Sum Yuen
November 2011

Contents

Centrosymmetric real nonpeiodic object, crystal, fibre and membrane **1**

Centrosymmetric real

nonperiodic object, crystal, fibre and membrane

1

An analytical solution for the phase problem of multidimensional centrosymmetric real object

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Abstract

It is important and useful to determine electron densities of centrosymmetric molecules and crystals. They can be determined from magnitude of the diffraction intensity by solving the multidimensional phase problem. An analytical and unique solution is presented. The multidimensional problem is reduced to one-dimensional problem. The method applies to nonperiodic (aperiodic) and periodic objects, small molecule, fibre, membrane and macromolecule. The in-between｜Fourier coefficients｜(or mid- Bragg intensities) are obtained by interpolation in multidimensions or experimental measurement. The objects are subjected to powerful constraints. The centrosymmetic fibre, fixed fibre molecule and planar periodic membrane are important as they are periodic structures determined without using interpolation. The electron densities obtained are exact. These and the freely rotating gas-molecule show whether atoms and molecules have finite size or sharp boundary, or the electron density is non-negative. The method is very suitable for nonperiodic and periodic macromolecules. The electron densities obtained for freely rotating molecule and fibre can be treated as fundamental data for model fitting. In this method, concept of atom and other knowledge of physics and chemistry are not used. Electron density is determined solely from diffraction intensity. Algorithms applicable to experimental electron diffraction data of molecules and X-ray crystallographic data need to be developed. The authors may re-determine the spherically averaged electron densities of the molecules and the electron densities of the centrosymmetric crystal structures in the literature. The atomic coordinates can then be obtained. The results can be compared with those in the literature, in particular, the electron densities. The oversampling ratio for one, two and three dimensions is shown to be exactly 2. This is useful for works in iterative algorithms.

Keywords: Phase retrieval; inverse scattering; Hilbert transform; discrete Hilbert transform; Shannon's sampling; minimum phase; electron density distribution; one-dimensional periodic object; two-dimensional periodic object; diffraction rods; Bragg rods; in-between | Fourier coefficients | ; mid-Bragg intensities.

1. Introduction

To investigate the structure of an object, we can use wave incident on the object. The diffraction pattern is given by the Fourier transform of the object. Experimentally we can only measure the magnitude of the Fourier transform, but not the phase. Therefore the structure of the object cannot be derived from the magnitude alone. Information from other sources is needed. In X-ray crystallography, we use the concept of atoms, the chemical structure of the object and the established data of bond lengths, bond angles, interatomic distances, obtained from all scientific studies, both experimental and theoretical, including quantum mechanical calculations and atomic scattering factors. The atomic coordinates and the electron density of the crystal are then determined (Coppens, 1997). The electron density result may not be as accurate as the atomic coordinates. If the phase of the Fourier transform can be determined from the diffraction intensity alone, the electron density and hence the atomic coordinates can be determined. The missing of the phase greatly reduces the power of X-ray diffraction method.

The question is whether the phase is really lost or just hidden in the magnitude. Can we recover the phase from the magnitude? This is the phase problem or phase retrieval problem or inverse scattering problem. In one dimension, the phase obtained from the magnitude, and hence the structure, may not be unique (Walther, 1963; Makowski, 1981). In multidimensions, for general positive object, the structure obtained may not be unique, as illustrated in Bruck & Sodin (1979) for discrete objects, and Huiser &

van Toorn (1980). However the solution is, in general, unique, in particular, if the size of the object is known (Bruck & Sodin, 1979; Hayes, 1982; Hayes & McClellan, 1982; Sanz & Huang, 1983; Barakat & Newsam, 1984; Bates, 1984). Klibanov (2006) presents a uniqueness theorem for the solution for a two-dimensional complex object, a phase screen. For centrosymmetric or antisymmetric real nonperiodic object of finite size, Hosemann & Bagchi (1962), p. 532 show that the structure determined is unique. Lawton (1981) shows the uniqueness for radial functions. Uniqueness means that phase is hidden in the magnitude.

Ramachandran (1969) applies Hilbert transform to the phase problem in crystallography. His result involves unknown derivatives. Kaufmann (1985) investigates possibility of using Hilbert transform to obtain the phase from the magnitude, and presents some difficulties, in particular for diffraction at single crystals. Mishnev (1993), (1996) present discrete Hilbert transform for X-ray crystallography, employing in-between structure factors. He suggests that a good phase set may be obtained, through the autocorrelation function (the Patterson function for a single unit cell). Zanotti et al. (1996) use these discrete Hilbert transforms for a model independent iterative procedure for phase extension and improvement. Phases of all or a fraction of the structure factors are assumed to be known.

Nieto-Vesperinas (1980) presents an important result that two- or three-dimensional Hilbert transforms do not exist. Napier & Bates (1974) investigate the two-dimensional phase problem by extending the one-dimensional complex zeros technique. Nakajima & Asakura (1985) present a two-dimensional phase retrieval method using the logarithmic Hilbert transform in one-dimension. For a review in phase retrieval using properties of entire functions, see Nakajima (1995). Deighton et al. (1985) present a two-dimensional phase retrieval method. They treat one-dimensional strips, and use the requirement that the phases given by two orthogonal strips are equal at their point of intersection. The method can be applied to three-dimensional case. Multidimensional phase problems are discussed in Millane (1996). Phase retrieval is discussed in the book by Hurt (1989). Nugent (2010) reviews coherent methods in the X-ray sciences. The phase problem of real or complex object, in particular, the multidimensional case, remains to be investigated.

Fienup has done much work in iterative algorithm (see, for examples Fienup (1982) and references in this article and Yuen (2011b, 2011c)). Millane & Stroud (1997) present an algorithm for reconstructing symmetric images from their undersampled Fourier intensities. Oversampling is presented in Sayre (1952), Bates (1982, 1984) and Millane (1996). Miao et al. (1998) presents an important demonstration of achieving phase retrieval by applying oversampling in computer phasing experiments. For iterative algorithms and oversampling, see Yuen (2011b, 2011c). Direct method uses atomicity with real and non-negative electron density (Karle, 1978). Methods for solving crystal structures is discussed in the books by Woolfson & Fan (1995) and Giacovazzo (1998). Phase retrieval in macromolecular crystallography is presented in Millane et al. (1986). Phase retrieval in crystallography and optics is reviewed in Millane (1990).

Theory of fibre diffraction is presented in Makowski (1982) and Stubbs (2001). Langmuir-Blodgett film is discussed in the book of Petty (1996). For two-dimensional solids, see Brinkman et al. (1982). Marks (1999) uses support constraint and direct methods to study three-dimensional surface structures. Wang et al. (2006) present phase determination for diffraction of centrosymmetric membrane by anomalous dispersion and Patterson function. Macromolecular crystallography is discussed in the book by Rossmann & Arnold (2001). Experimental phasing is reviewed in McCoy & Read (2010). Current state and prospects of macromolecular crystallography is reviewed in Dauter (2006). Macromolecular crystallography at synchrotron radiation is reviewed in Duke & Johnson (2010). Zhang et al. (2000) investigates protein structure by wide angle solution X-ray scattering. Diffraction pattern and imaging from a beam of laser-aligned protein molecules is presented in Spence et al. (2005). Structure determination of noncrystalline specimens by means of oversampling is reviewed in

Miao et al. (2004) and Miao et al. (2008).

The phase retrieval problem can be reduced to a mathematical problem: What are the constraints of the object, under which the phase of the Fourier transform of an object can be obtained from the magnitude (see, for example, Klibanov et al., 1995, p.2)? This article and Yuen (2011b, 2011c) form a systematic and comprehensive treatment of the phase problem. This article presents an analytical and unique solution for centrosymmetric real object. The constraints used are very general and reasonable: finite size, real, centrosymmetric and bounded. The method determines electron density from diffraction intensity and applies to the large number of centrosymmetric molecules and crystals.

Much work has been done for the one-dimensional case (for example, Walther, 1963; Hofstetter, 1964; Burge at al., 1976; Makowski, 1981). Klibanov et al. (1995) provides a review. We present solution for one-dimensional case in Sect. 2.1. This solution for one-dimensional case is then used to obtain an analytical solution for multidimensional case (Sect. 2.2).

Sect. 2.2.1 deals with the multidimensional centrosymmetric nonperiodic and periodic object

Sect. 2.2.2 multidimensional antisymmetric nonperiodic object

Sect. 2.2.3 centrosymmetic fibre, or fixed fibre molecule (a one-dimensional periodic object)

Sect. 2.2.4. real membrane

Sect. 2.2.5 centrosymmetric real macromolecule. The method is very suitable for determination of periodic macromolecular structure, as shown in Appendix.

The structure determined is up to a multiplicative factor of -1. The location of the object is not determined. For periodic object, as in Millane (1996), we deal with the content of the basic unit cell as an object, and define $f_0(\mathbf{x}) = f(\mathbf{x})$ for $\mathbf{x}$ ε the basic unit cell, or = 0 otherwise.

2. Method

2.1. One-dimensional real object

The diffraction pattern is given by the Fourier transform of $f(x)$,

$$F(\omega) = \int_{-\infty}^{\infty} f(x)\exp(-i\omega x)dx \tag{1}$$

(Duffieux, 1983, p. 101-102). $F(\omega) = |F(\omega)|\exp[i\phi(\omega)]$.

Experimentally, we can only measure $|F(\omega)|$; the phase $\phi(\omega), -\infty < \omega < \infty$, is missed. The present article uses constraints of $f(x)$ so that $\phi(\omega)$ can be recovered from $|F(\omega)|, -\infty < \omega < \infty$. The general and reasonable constraints on an object include:

(i) finite size from $-L/2$ to $L/2$. This constraint is reasonable because either an object has finite size or experimentally we can only deal with finite part of the object. The unit cell of a crystal has finite size.

(ii) $f(x)$ is real

(iii) centrosymmetric, $f(x)$ is even, or antisymmetric, $f(x)$ is odd

(iv) $f(x)$ is bounded

(v) $f(x)$ is non-negative, $f(x) \geq 0$

(vi) $f(x)$ is a continuous periodic object with $f_0(-L/2) = f_0(L/2)$. L is the unit cell constant.

(vii) other symmetry elements for multidimensional object

2.1.1. Centrosymmetric object, $f(x) = f(-x)$

(A) One-dimensional centrosymmetric nonperiodic object

Constraints used:

(i) size of object is from $-L/2$ to $L/2$. Value of L will be determined

(ii), (iii) and (iv).

We have $|F(\omega)|$ which is continuous. The sign of $F(\omega)$ changes at a zero of odd order and remains unchanged at a zero of even order. To obtain the structure $f(x)$, we need only locate the zeros of $|F(\omega)|$, and finding out whether they are of even order or odd. (The values of the orders are not needed). This is similar to Sayre (1952). Choose a sign of $F(0)$. The sign of $F(\omega)$ is determined. The structure of the object, including the value of L, is then determined. It is centrosymmetric and unique. This agrees with the result in Hosemann & Bagchi (1962), p. 532.

For discrete $|F(\omega)|$, we have to extrapolate to the zeros. This is not very accurate. May be it is not a zero at all. This may ruin the result. If size of object is known, use this size. If size is unknown, use a sufficiently large L to contain the object. Then add its replica successively to give a periodic object and use case (B) in below. If needed, the | Fourier coefficients | and in-between | Fourier coefficients | (Millane et al. (1986), p. 60) can be obtained by interpolation. Values obtained by interpolation are more accurate than those zeros obtained by extrapolating $|F(\omega)|$ to zero.

(B) One-dimensional centrosymmetric periodic object (for example, crystal)

Choose the interval $[-L/2, L/2]$ as the basic unit cell. Let $F_0(\omega)$ be the Fourier transform of $f_0(x)$. Constraints used:

(i) $f_0(x)$ has finite size with size = the known unit cell constant,

(ii), (iii) and (iv).

Bilateral Laplace transform of $f_0(x)$,

$$F_{0II}(s) = \int_{-\infty}^{\infty} f_0(x)\exp(-sx)dx \qquad (2)$$

$$(F_{0II}(s))^* = F_{0II}(s^*) \tag{3}$$

where * denotes complex conjugate.

The size of the object $f_0(x)$ is from $-L/2$ to $L/2$. $f_0(x)$ and $F_{0II}(s)$ satisfy the Paley-Wiener theorem. $F_{0II}(s)$ is an entire function of exponential type, order 1 and type $L/2$, but not of type less than $L/2$ (Boas, 1954, 6.8.1, 6.8.7.). $F_{II}(s)$ has an infinite number of zeros (Titchmarsh, 1968, p. 287, Example 23). Let γ_1, γ_2,… be the zeros of $F_{0II}(s)$ not at the origin. Since $f_0(x)$ is even, $F_{0II}(s) = F_{0II}(-s)$. If γ_n is a zero of $F_{0II}(s)$, then $-\gamma_n$ is also a zero. Since the zeros occur in conjugate pairs, γ_n^* and $-\gamma_n^*$ are also zeros. Therefore the zeros are symmetric with respect to the real axis and the imaginary axis (Nussenzveig (1967), p. 568, Bates, 1969, p. 425). From Hadamard's factorization theorem,

$$F_{0II}(s) = As^{\alpha}\left[\prod_{n=1}^{\infty}\left(1-\frac{s}{\gamma_n}\right)\exp(s/\gamma_n)\right] \tag{4}$$

where A is a real constant, α is an even integer ≥ 0 for the multiplicity of the origin as a zero of $F_{0II}(s)$.

Let $u(x) = f_0(x - L/2)$ (5)

$u(x)$ is casual.

Follow Yuen (2011b), Sect. 2.1 and obtain a minimum-phase function $H_M(\omega)$.

$$H_M(\omega) = |H_M(\omega)|\exp[i\phi_M(\omega)]$$

$$|F_0(\omega)| = |H_M(\omega)| \tag{6}$$

Use discrete Hilbert transform for $|H_M(\omega)|$ (Papoulis, 1977, (7-132); Marks, 1991, p. 49).

$$\phi_M(l\omega_{1/2}) = -\frac{2}{\pi}\sum\frac{\log|F_0(m\omega_{1/2})|}{l-m} \tag{7}$$

where M stands for minimum-phase function (Papoulis, 1977, Sect. 7-2; Papoulis, 1962, Sect. 10-3)

$l = 0,\pm 1,\pm 2,\ldots$, $\omega_{1/2} = 2\pi/(2L) = \pi/L$, and summation over m , $l-m$ odd.

Fourier coefficients of $f(x)$ occur at $m\omega_0$, where $m = 0,\pm 1,\pm 2,\ldots$, $\omega_0 = 2\pi/L$. $\omega_{1/2} = \omega_0/2$. Experimentally the intensity measured is proportional to the $\left|\text{ Fourier coefficient }\right|^2$. Fourier coefficient at $m\omega_0$, $= (1/L)F_0(m\omega_0)$. Between every two adjacent Fourier coefficients at $m\omega_0$ and $(m+1)\omega_0$, the intensity at $[m+(1/2)]\omega_0$ cannot be measured. We can obtain $|F_0((m+(1/2))\omega_0)|$, $m = 0,\pm 1,\pm 2,\ldots$, by interpolation. As explained in the Discussion, the result obtained by interpolation is fairly accurate. With $\phi_M(l\omega_{1/2})$ obtained from (7), we get $H_M(l\omega_{1/2}) = |H_M(l\omega_{1/2})|\phi_M(l\omega_{1/2})$. Then, for $-\infty < \omega < \infty$, Shannon's sampling theory gives

$$H_M(\omega) = \sum_{l=-\infty}^{\infty} H_M(l\omega_{1/2}) \frac{\sin(L\omega - l\pi)}{L\omega - l\pi} \tag{8}$$

By analytic continuation, $H_{II}(s) = H_M(s/i)$.

As explained in the Discussion, $H_{II}(s)$ is smooth. This facilitates the process to locate the zeros. All the zeros have $\operatorname{Re} s \leq 0$. The order of each zero can be determined.

Let γ_n be a zero in the left-hand plane ($\operatorname{Re}\gamma_n < 0$). It has even order for centrosymmetric object (Bates, 1969, p. 425). The zeros occur in conjugate pairs. If we multiply $H_{II}(s)$ by

$$\frac{\gamma_n\left[\left(-\gamma_n^*\right)-s\right]}{(\gamma_n - s)\left(-\gamma_n^*\right)}\exp(-s/\gamma_n)\exp\left[s/\left(-\gamma_n^*\right)\right].$$

$$\frac{\gamma_n^*[(-\gamma_n)-s]}{(\gamma_n^*-s)(-\gamma_n)}\exp(-s/\gamma_n^*)\exp[s/(-\gamma_n)]$$

The resulting function has the same magnitude at $i\omega$, but the phase is changed (Toll, 1956, p. 1767; Hofstetter, 1964). The zeros γ_n, γ_n^* are flipped to $-\gamma_n^*$ and $-\gamma_n$ in the right-hand plane respectively.

Flip half of the zeros in the left-hand plane $\operatorname{Re} s < 0$ to the right-hand plane. $F_0(\omega)$ is obtained from (4). From $F_0(m\omega_0)$, $m = 0,\pm1,\pm2,\ldots$, and the Fourier series, $f_0(x)$ is obtained.

In fact, we can follow (A). Use (8) and (6). We need only locate the zeros of $|F_0(\omega)|$, and finding out whether they are of even order or odd. (The values of the orders are not needed). Choose a sign of $F_0(0)$. The sign of $F_0(\omega)$ is determined. The structure of the object, including the value of L, is then determined. It is centrosymmetric and unique. Analytic continuation to the complex plane and flipping of zeros are not needed.

Since the in-between | Fourier coefficients| are obtained by interpolation. They are approximate. Some of the zeros of $H_{II}(s)$ in the left-hand plane may have odd order. We may flip these zeros alternatively to the right hand plane according to their distances from the origin. For this arrangement of the zeros, $F_0(\omega)$ is obtained from (4). From $F_0(m\omega_0)$, $m = 0,\pm1,\pm2,\ldots$, and the Fourier series, $f_0(x)$ is obtained. The known size is a tight support, a strong constraint. Adjust the in-between | Fourier coefficients| so that $f_0(x)$ satisfies the four constraints. The structure is unique for this set of | Fourier coefficients| and in-between | Fourier coefficients| .

We may use a negativity index $= \left|\int_{-L/2}^{L/2} f_0(x)dx\right|\left[\int_{-L/2}^{L/2} f_0(x)dx\right]^{-1} \geq 0$

where $f_0(x) < 0$ in the first factor, and ≥ 0 in the second factor. For non-negative object, this index =0. If applicable, the interpolated in-between | Fourier coefficients| are adjusted to satisfy constraints:

(v) $f_0(x)$ is non-negative, $f_0(x) \geq 0$

(vi) $f(x)$ is a continuous periodic object with $f_0(-L/2) = f_0(L/2)$

For nonperiodic object, mathematically adding its replica successively gives a periodic object. If the period is chosen such that the | Fourier coefficients| and in-between | Fourier coefficients| are known, interpolation is not needed. Discrete Hilbert transform (7), and Shannon's sampling (8) can then be used. Use (6), $|F_0(\omega)| = |H_M(\omega)|$. Use this continuous $|F_0(\omega)|$ and follow (A). The structure of the object, including the value of L, is determined. It is centrosymmetric and unique.

2.1.2. One-dimensional antisymmetric object, $f(x) = -f(-x)$

For nonperiodic object, the procedure used is the same as in (A) except that the sign of $F(\omega)$ for one value of ω has to be chosen. The structure obtained is unique.

For periodic object, $F_{0II}(s) = -F_{0II}(-s)$. As in the centrosymmetric case, the zeros of $F_{0II}(s)$ are symmetric with respect to the real axis and the imaginary axis. The only difference is that α in (4) is an odd integer> 0. The constraints used are same as those for the centrosymmetric case except (iii) $f(x)$ is antisymmetric. The procedure used is the same as in (B). $\omega = 0$ is a zero of $|F(\omega)|$ of odd order. The structure obtained is unique for each set of | Fourier coefficients| and in-between | Fourier coefficients| .

2.2. Multidimensional real object

2.2.1. General centrosymmetric object

(C) Multidimensional centrosymmetric nonperiodic object,

$$f(x_1, x_2) = f(-x_1, -x_2)$$

Constraints used:

(i) finite size. $f(x_1, x_2) = 0$ for $|x_1| > L/2$, $x_2 < H(x_1)$ or $x_2 > K(x_1)$.

Values of L, $H(x_1)$ and $K(x_1)$ will be determined.

(ii), (iii) and (iv).

$$F(\omega_1, \omega_2) = \int_{-L/2}^{L/2} \int_{H(x_1)}^{K(x_1)} f(x_1, x_2) \exp[-i(\omega_1 x_1 + \omega_2 x_2)] dx_1 dx_2$$

$$= \frac{1}{2} \int_{-L/2}^{L/2} \left\{ \int_{H(x_1)}^{K(x_1)} f(x_1, x_2) \exp(-i\omega_2 x_2) dx_2 + \int_{-K(x_1)}^{-H(x_1)} f(-x_1, x_2) \exp(-i\omega_2 x_2) dx_2 \right\} \cdot \exp(-i\omega_1 x_1) dx_1$$

$$= \int_{-L/2}^{L/2} \left\{ \int_{H(x_1)}^{K(x_1)} f(x_1, x_2) \cos(\omega_2 x_2) dx_2 \right\} \exp(-i\omega_1 x_1) dx_1 \tag{9}$$

Since $|f(x_1, x_2)|$ is bounded, by the Fubini theorem, the integral is independent of the order of integration. As suggested in Nieto-Vesperinas (1980), p. 383, and used by Klibanov (2006), p. 820, for a fixed $\omega_2 = \omega_2'$, the term in curly brackets, $q(x_1, \omega_2')$ is a one-dimensional centrosymmetric real object, a function of x_1, with size from $x_1 = -L/2$ to $x_1 = L/2$. This is a one-dimensional problem. As shown in (A), a unique $F(\omega_1, \omega_2')$ is obtained from $|F(\omega_1, \omega_2')|$ for all ω_1. For all ω_2, we obtain $F(\omega_1, \omega_2)$. Its inverse Fourier transform gives the unique $f(x_1, x_2)$ which is centrosymmetric. L, $H(x_1)$ and $K(x_1)$ are determined.

Replicas of the nonperiodic object can be added successively to give a periodic object. (D) in below can then be applied.

For three-dimensional object,

$$F(\omega_1,\omega_2,\omega_3)=\int_{-L/2}^{L/2}\int_{H(x_1)}^{K(x_1)}\int_{P(x_1,x_2)}^{Q(x_1,x_2)} f(x_1,x_2,x_3)\exp[-i(\omega_1 x_1+\omega_2 x_2+\omega_3 x_3)]\cdot$$

$$dx_1 dx_2 dx_3$$

$$=\frac{1}{2}\int_{-L/2}^{L/2}\left\{\int_{H(x_1)}^{K(x_1)}\left[\int_{P(x_1,x_2)}^{Q(x_1,x_2)} f(x_1,x_2,x_3)\exp[-i(\omega_2 x_2+\omega_3 x_3)]dx_3\right]dx_2+\right.$$

$$\left.\int_{-K(x_1)}^{-H(x_1)}\left[\int_{P(-x_1,x_2)}^{Q(-x_1,x_2)} f(-x_1,x_2,x_3)\exp[-i(\omega_2 x_2+\omega_3 x_3)]dx_3\right]dx_2\right\}\exp(-i\omega_1 x_1)dx_1$$

$$=\int_{-L/2}^{L/2}\left\{\int_{H(x_1)}^{K(x_1)}\left[\int_{P(x_1,x_2)}^{Q(x_1,x_2)} f(x_1,x_2,x_3)\cos(\omega_2 x_2+\omega_3 x_3)dx_3\right]dx_2\right\}\exp(-i\omega_1 x_1)dx_1$$

(10)

For fixed ω_2', ω_3', a unique $F(\omega_1,\omega_2',\omega_3')$ is obtained from

$|F(\omega_1,\omega_2',\omega_3')|$ for all ω_1, $-\infty<\omega_1<\infty$. For all ω_2, ω_3, we obtain $F(\omega_1,\omega_2,\omega_3)$. Its inverse Fourier transform gives the unique $f(x_1,x_2,x_3)$ including L, $H(x_1)$, $K(x_1)$, $P(x_1,x_2)$ and $Q(x_1,x_2)$. The uniqueness agrees with Hosemann & Bagchi (1962), p. 532. If the object is non-negative, continuous or has other symmetry elements, these will be shown in the result.

A 2-dimensional circularly symmetric object $g(r)=f(x_1,x_2)$,

where $r^2=x_1^2+x_2^2$.

(9) becomes

$$F(\omega_1,\omega_2)=\int_{-R}^{R}\left\{2\int_{0}^{K(x_1)}f(x_1,x_2)\cos(\omega_2x_2)dx_2\right\}\exp(-i\omega_1x_1)dx_1 \qquad (11)$$

where $K(x_1)$ is a semicircle. Fixed $\omega_2=0$. Follow (A) and obtain the unique $F(\omega_1,0)$ from $|F(\omega_1,0)|$ for all ω_1.

The object has circular symmetry.

Let $U(\omega)=F(\omega_1,\omega_2)$

where $\omega^2=\omega_1{}^2+\omega_2{}^2$.

Hence $U(\omega)=F(\omega,0)$ is obtained.

The unique structure,

$$g(r)=\frac{1}{2\pi}\int_{0}^{\infty}U(\omega)J_0(r\omega)\omega d\omega \qquad (12)$$

where $J_0(r\omega)$ is the 0th order Bessel function of the first kind. Value of R is determined.

A three-dimensional spherically symmetric object $g(r)=f(x_1,x_2,x_3)$,

where $r^2=x_1{}^2+x_2{}^2+x_3{}^2$.

(10) becomes

$$F(\omega_1,\omega_2,\omega_3)=\int_{-R}^{R}\left\{\iint f(x_1,x_2,x_3)\cos(\omega_2x_2+\omega_3x_3)dx_2dx_3\right\}\exp(-i\omega_1x_1)dx_1$$

(13)

where the double integral inside the curly brackets is over a circular section of the sphere of radius R. Fix $\omega_2=\omega_3=0$. Follow (A) and obtain the unique

$F(\omega_1, 0, 0)$ from $|F(\omega_1, 0, 0)|$ for all ω_1.

The object has spherical symmetry.
Let $U(\omega) = F(\omega_1, \omega_2, \omega_3)$

where $\omega^2 = \omega_1{}^2 + \omega_2{}^2 + \omega_3{}^2$.

Hence $U(\omega) = F(\omega, 0, 0)$ is obtained.
$U(\omega)$ is the form factor, or for an atom, atomic scattering factor. The unique structure,

$$g(r) = \frac{1}{2\pi^2} \int_0^{\infty} U(\omega)\omega \frac{\sin \omega r}{r} d\omega \tag{14}$$

Value of R is determined. The uniqueness agrees with Lawton (1981), Answer 2. This applies to electron diffraction of gas molecules.

(D) Multidimensional centrosymmetric periodic object

Choose the cell, $-a/2 \le x_1 \le a/2, -b/2 \le x_2 \le b/2$ as the basic unit cell. Constraints used:

(i) $f_0(x_1, x_2)$ has finite size with size = the known unit cell constants, a, b, α

(ii), (iii), and (iv).

Let $\omega_{10} = 2\pi / a$, $\omega_{20} = 2\pi / b$. $|F_0(l\omega_{10}, m\omega_{20})|$ are measured experimentally.

Follow (9),

$$F_0(l\omega_{10}, m\omega_{20}) = \int_{-a/2}^{a/2} \left\{ \int_{-b/2}^{b/2} f_0(x_1, x_2) \cos(m\omega_{20} x_2) dx_2 \right\} \exp(-il\omega_{10} x_1) dx_1 \tag{15}$$

For a fixed m', obtain $|F_0((l + 1/2)\omega_{10}, m'\omega_{20})|$, $l = 0, \pm 1, \pm 2., \ldots$, by interpolation in multidimensions (Bates, 1984, p. 216; Kincaid & Cheney, 1996, Sect.

6.10; Cheney & Light, 2000; Anita, 2002, Sect. 4.7, 4.8, Hau-Riege et al., 2004) or by experimental measurement (Yuen, 2011d, for a crystal). Follow the method in (C) and (B). Adjust the interpolated values such that the term in curly brackets, $q(x_1, m'\omega_{20})$ is a real, bounded and even function of x_1 with size from $x_1 = -a/2$ to $x_1 = a/2$. The known size is a tight support, a strong constraint. Find the unique $F_0(l\omega_{10}, m'\omega_{20})$ for all l. Repeat this for all m, get all $F_0(l\omega_{10}, m\omega_{20})$. With the Fourier coefficients obtained from $F_0(l\omega_{10}, m\omega_{20})$, the Fourier series gives a structure $f(x_1, x_2)$ (cf. Millane, 1996, Sect. 3). The interpolated values are further adjusted such that $f(x_1, x_2)$ is centrosymmetric. For this set of | Fourier coefficients | and in-between | Fourier coefficients | , the structure obtained is unique.

For three-dimensional case, we have $|F_0(l\omega_{10}, m\omega_{20}, n\omega_{30})|$. Fix m', n' and obtain $|F_0((l+1/2)\omega_{10}, m'\omega_{20}, n'\omega_{30})|$, $l = 0, \pm1, \pm2., \ldots$, by interpolation in multidimensions or experimental measurement. Find the unique $F_0(l\omega_{10}, m'\omega_{20}, n'\omega_{30})$, $l = 0, \pm1, \pm2., \ldots$. Repeat this for all m, n, get all $F_0(l\omega_{10}, m\omega_{20}, n\omega_{30})$. The Fourier series gives the unique structure.

In X-ray crystallography, the electron density satisfies:

(v) $f_0(x_1, x_2, x_3)$ is non-negative

(vi) $f(x_1, x_2, x_3)$ is continuous

with $f_0(-a/2, x_2, x_3) = f_0(a/2, x_2, x_3)$ for all x_2, x_3

$f_0(x_1, -b/2, x_3) = f_0(x_1, b/2, x_3)$ for all x_1, x_3

$f_0(x_1, x_2, -c/2) = f_0(x_1, x_2, c/2)$ for all x_1, x_2

(vii) other symmetry elements, if applicable

The interpolated in-between | Fourier coefficients | are adjusted to satisfy these constraints. As the | Fourier coefficients| are used in obtaining the structure, the R -index = 0.

Note that for one, two or three dimensions, we use interpolated in-between | Fourier coefficients| in only one index. The oversampling ratio is exactly 2. It is mentioned in the Discussion that, for more accurate values, choose the longest unit cell edge along which the interpolated in-between | Fourier coefficients| are employed.

2.2.2. Multidimensional antisymmetric nonperiodic object, $f(x_1,x_2)=-f(-x_1,-x_2)$

The constraints used are similar to those for the centrosymmetric case (C) except that for (iii). $f(x_1,x_2)$ is antisymmetric.
Similar to (9),

$$F(\omega_1,\omega_2)=-i\int_{-L/2}^{L/2}\left\{\int_{H(x_1)}^{K(x_1)} f(x_1,x_2)\sin(\omega_2 x_2)dx_2\right\}\exp(-i\omega_1 x_1)dx_1$$

For a fixed $\omega_2{}'$, the term in curly brackets, $q(x_1,\omega_2{}')$ is a one-dimensional antisymmetric real object of x_1, with size from $x_1=-L/2$ to $x_1=L/2$. This is a one-dimensional problem. As shown in Sect. 2.1.2, a unique $F(\omega_1,\omega_2{}')$ is obtained from $|F(\omega_1,\omega_2{}')|$ for all ω_1. For all ω_2, we obtain $F(\omega_1,\omega_2)$. Its inverse Fourier transform gives the unique $f(x_1,x_2)$ which is antisymmetric. L, $H(x_1)$ and $K(x_1)$ are determined.

2.2.3. Real fibre

(E) Centrosymmetric fibre

Fibre has circular symmetry in the x_1x_2 plane with radius $R(x_3)$, and is periodic in

x_3 with period c. Choose the cell $({x_1}^2+{x_2}^2)^{1/2} \le R(x_3), -c/2 \le x_3 \le c/2$, as the basic unit cell. Constraints used:

(i) For each x_3, the object $f_0(x_1,x_2,x_3)$ has finite radius $R(x_3)$ (value will be determined) in the x_1x_2 plane. Value of c is known.

(ii) $f_0(x_1,x_2,x_3)$ is real

(iii) For each x_3, the object has circular symmetry in the x_1x_2 plane.

$f_0(x_1,x_2,x_3)$ is centrosymmetric

(iv) $f_0(x_1,x_2,x_3)$ is bounded

The weak diffraction intensity is collected into strong diffraction planes which can be measured accurately. Let $\omega_0 = 2\pi/c$. For all l, $l = 0,\pm1,\pm2.,\ldots,$

$|F_0(\omega_1,\omega_2,l\omega_0)|$ are measured experimentally. It has circular symmetry in the $\omega_1\omega_2$ plane.

$$F_0(\omega_1,\omega_2,l\omega_0) = \iint\left\{\int_{-c/2}^{c/2} f_0(x_1,x_2,x_3)\exp(-il\omega_0x_3)dx_3\right\}\cdot$$

$$\exp[-i(\omega_1x_1+\omega_2x_2)]dx_1dx_2 \tag{16}$$

where the triple integral is over the basic unit cell. For a fixed l', the term in curly brackets, $q(x_1,x_2,l'\omega_0)$, is a two-dimensional centrosymmetric real object of x_1 and x_2. $q(x_1,x_2,l'\omega_0)$ is circularly symmetric in the x_1x_2 plane with radius=the largest $R(x_3)$. This becomes a two-dimensional problem. Apply the method in (C). Use $|F_0(\omega_1,\omega_2,l'\omega_0)|$ for all ω_1, ω_2. Obtain the unique $F_0(\omega_1,\omega_2,l'\omega_0)$ and $q(x_1,x_2,l'\omega_0)$. Repeat this for all l. Get all $q(x_1,x_2,l\omega_0)$.

Now $f(x_1, x_2, x_3)$ is periodic in x_3 with period c. For fixed x_1 and x_2, express $f(x_1, x_2, x_3)$ as a Fourier series of x_3.
The Fourier coefficient

$$= \frac{1}{c}\int_{-c/2}^{c/2} f_0(x_1, x_2, x_3)\exp\left(-il\omega_0 x_3\right)dx_3 = \frac{1}{c}q(x_1, x_2, l\omega_0)$$

Hence $f(x_1, x_2, x_3) = \frac{1}{c}\sum_{l=-\infty}^{\infty} q(x_1, x_2, l\omega_0)\exp\left(il\omega_0 x_3\right)$ (17)

For all x_1 and x_2, the unique structure $f(x_1, x_2, x_3)$ including $R(x_3)$ is determined.
If (vi) the fibre $f(x_1, x_2, x_3)$ is continuous with

$$f_0(x_1, x_2, -c/2) = f_0(x_1, x_2, c/2) \text{ for all } x_1,\ x_2,$$

this will be shown in the structure $f(x_1, x_2, x_3)$ obtained.

This applies to helix polymer with random orientation about the helix axes. The body may be an aggregate of two or more parallel (or antiparallel) helical molecules and the aggregate has random orientation about the helix axis.

(F) Fixed centrosymmetric fibre molecule (a one-dimensional periodic object)

Choose the cell

$H(x_2, x_3) \le x_1 \le K(x_2, x_3), L(x_3) \le x_2 \le M(x_3), -c/2 \le x_3 \le c/2$, as the basic unit cell. Constraints used:

(i) $f_0(x_1, x_2, x_3)$ has finite size. Values of $H(x_2, x_3)$, $K(x_2, x_3)$, $L(x_3)$ and $M(x_3)$ will be determined.

(ii), (iii) and (iv).

Let $\omega_0 = 2\pi/c$. For all l, $\left|F_0(\omega_1, \omega_2, l\omega_0)\right|$ are measured experimentally. As in (16), apply the method in (C). Obtain the unique $q(x_1, x_2, l\omega_0)$ for all l. The unique structure $f(x_1, x_2, x_3)$ is given by (17). $H(x_2, x_3)$, $K(x_2, x_3)$, $L(x_3)$ and $M(x_3)$ are determined.

2.2.4. Real membrane

The membrane may be an open surface or a closed surface. The thickness of the membrane may or may not be uniform.

For centrosymmetric nonperiodic real membrane, use (C). The solution is unique.

(G) Planar centrosymmetric periodic membrane (a two-dimensional periodic object)

The membrane is periodic in x_1 and x_2 with periods a and b respectively. Choose the cell $-a/2 \le x_1 \le a/2$, $-b/2 \le x_2 \le b/2$, $H(x_1, x_2) \le x_3 \le K(x_1, x_2)$ as the basic unit cell. Constraints used:

(i) $f_0(x_1, x_2, x_3)$ has finite size with size = the known unit cell constants, a, b, α. Values of $H(x_1, x_2)$ and $K(x_1, x_2)$ will be determined.

(ii), (iii) and (iv).

The weak diffraction intensity is collected into strong diffraction rods (Bragg rods) which can be measured accurately. Let $\omega_{10} = 2\pi/a$, $\omega_{20} = 2\pi/b$. For all l and m, $\left|F_0(l\omega_{10}, m\omega_{20}, \omega_3)\right|$ are measured experimentally.

$$F_0(l\omega_{10}, m\omega_{20}, \omega_3)$$

$$= \int \left\{ \int_{-a/2}^{a/2} \int_{-b/2}^{b/2} f_0(x_1, x_2, x_3) \exp[-i(l\omega_{10}x_1 + m\omega_{20}x_2)] dx_1 dx_2 \right\} \exp(-i\omega_3 x_3) dx_3$$

where the triple integral is over the basic unit cell.

For fixed l' and m', the term in curly brackets, $q(l'\omega_{10}, m'\omega_{20}, x_3)$, is a one-dimensional centrosymmetric real object of x_3, with size from x_3 = minimum of $H(x_1, x_2)$ to x_3 = maximum of $K(x_1, x_2)$. This is a one-dimensional problem. From $|F_0(l'\omega_{10}, m'\omega_{20}, \omega_3)|$ for all ω_3, use (A) and obtain the unique $F_0(l'\omega_{10}, m'\omega_{20}, \omega_3)$ and $q(l'\omega_{10}, m'\omega_{20}, x_3)$. Repeat this for all l and m. Get all $q(l\omega_{10}, m\omega_{20}, x_3)$.

Now $f(x_1, x_2, x_3)$ is periodic in x_1 and x_2. For fixed x_3, express $f(x_1, x_2, x_3)$ as a Fourier series of x_1 and x_2. The Fourier coefficient

$$= \frac{1}{ab} \int_{-a/2}^{a/2} \int_{-b/2}^{b/2} f_0(x_1, x_2, x_3) \exp[-i(l\omega_{10}x_1 + m\omega_{20}x_2)] dx_1 dx_2$$

$$= \frac{1}{ab} q(l\omega_{10}, m\omega_{20}, x_3)$$

Hence

$$f(x_1, x_2, x_3) = \frac{1}{ab} \sum_{l=-\infty}^{\infty} \sum_{m=-\infty}^{\infty} q(l\omega_{10}, m\omega_{20}, x_3) \exp[i(l\omega_{10}x_1 + m\omega_{20}x_2)]$$

For all x_3, the unique structure $f(x_1, x_2, x_3)$ including $H(x_1, x_2)$ and $K(x_1, x_2)$ is determined.

Note that (E), (F) and (G) are cases of determining unique periodic structures without using interpolation. The structures obtained are exact. If the object is non-negative, continuous or has other symmetry elements, these will be shown in the result.

For Sect. 2.2.2, (F) and (G), unique solution can be obtained for antisymmetric object, or one which is even in some of the variables x_1, x_2, x_3 and odd in the remaining variables, and for antisymmetric fibre.

2.2.5. Centrosymmetric real macromolecule

For centrosymmetric nonperiodic and periodic real macromolecule ((C), (D), (E), (F), (G) and Sect. 2.2.2), the structure determined is unique:

For (C), (E), (F) and (G), no interpolation is used. The structure obtained is exact.

For periodic object ((D)), the in-between | Fourier coefficients | for one index are obtained by interpolation in multidimensions. For crystal of a macromolecule, the unit cell is large. Hence the reciprocal lattice points are closely spaced. The interpolated values and the structure determined can be more accurate (see the Appendix and Hau-Riege et al., 2004, p. 300). This method is very suitable for the determination of the structures of nonperiodic and periodic macromolecules. For periodic macromolecule (D), the structure determined is unique for each set of | Fourier coefficients | and in-between | Fourier coefficients | .

3. Discussion

For centrosymmetric nonperiodic or periodic real object, the four constraints (i), (ii), (iii) and (iv) are sufficient to give a unique solution of the phase problem. In daily life, we only see finite real object. We seldom see complex object. As the phase can only be 0 or π radians, the correct phase can be obtained even if there is experimental uncertainty or computational approximation (Berg & Goffeney, 1997, p. 622).

For nonperiodic object, the shape and dimensions of the object are determined. In many cases, these are the results that we are interested in. $f(\mathbf{x})$ may taper to zero continuously at the boundary or has sharp boundary. Continuity and differentiability of $f(\mathbf{x})$ are not required. The object can have sharp corners. This agrees with our experience that in our daily life, we see objects with sharp corners.

For periodic object, in (B) and (D), we obtain $|F((n+(1/2))\omega_0)|$, $n = 0, \pm 1, \pm 2, \ldots$, by interpolation in multidimensions. For one-dimensional case, the ruggedness of $F(\omega)$ is due to the value of $f_0(x)$ for large value of x. Bandlimited function is smooth (Marks, 1991, (2.40); Papoulis, 1984, p. 332). Define

$$(\Delta x)^2 = \int_{-\infty}^{\infty} x^2 |f(x)|^2 dx \text{ and } (\Delta\omega)^2 = \int_{-\infty}^{\infty} \omega^2 |F(\omega)|^2 d\omega .$$

$$(\Delta x)^2 = \frac{1}{2\pi} \int_{-\infty}^{\infty} \left[\left(\frac{d|F(\omega)|}{d\omega} \right)^2 + |F(\omega)|^2 \left(\frac{d\phi}{d\omega} \right)^2 \right] d\omega ,$$

(Papoulis, 1962, (4-39)). $f_0(x) = 0$ for x outside the basic unit cell. Δx is not large. Hence the ripple in $|F(\omega)|$ or ϕ is small. $|F(\omega)|$ is smooth. For example, the Fourier transform of rectangular and triangular pulses are very smooth. Bracewell (1986), Ch. 21 shows many illustrations. Since $|F(\omega)|$ is smooth, the interpolated values would be fairly accurate. Δx and $\Delta\omega$ are related by the uncertainty principle, $\Delta x \Delta\omega \geq \sqrt{\pi/2}$.

If the unit cell constant, L is large, $\omega_o = 2\pi/L$ is small. The Fourier coefficients are closely spaced. Since Δx is larger, $\Delta\omega$ may be reduced. The question is whether the in-between | Fourier coefficients | obtained by interpolation is more accurate. The Appendix shows that the interpolated values would be more accurate. In the limit as $L \to \infty$, the discrete spectrum of Fourier coefficients approaches a continuous function. The interpolated values would be exact, no longer approximate. Hence the structure of the object determined would be exact. For crystal of large unit cell, the unit cell contains a large number of atoms, the accurate values of interpolation leads to more accurate value of electron density. This method is very suitable for crystals with large unit cell, in strong contrast to the Patterson function where large number of peaks overlap. Choose the longest unit cell edge along which the interpolated in-between | Fourier coefficients | are employed. The ω_0 will then be smallest and the interpolated values more accurate.

The in-between | Fourier coefficients | are obtained by interpolation as used by Bates (1984), Millane (1993) and Hau-Riege et al. (2004) in oversampling. The uncertainty introduced by interpolation is similar to experimental uncertainty. If the object satisfies:
(v) $f_0(x_1, x_2, x_3)$ is non-negative
(vi) $f(x_1, x_2, x_3)$ is continuous

with $f_0(-a/2, x_2, x_3) = f_0(a/2, x_2, x_3)$ for all x_2, x_3

$f_0(x_1, -b/2, x_3) = f_0(x_1, b/2, x_3)$ for all x_1, x_3

$f_0(x_1, x_2, -c/2) = f_0(x_1, x_2, c/2)$ for all x_1, x_2

(vii) other symmetry elements, if applicable,
the interpolated in-between | Fourier coefficients | are adjusted to satisfy the tight support of known unit cell constants, the centrosymmetric constraint and these constraints. The more the number of other symmetry elements, the constraint (vii) is more effective. Thus the in-between | Fourier coefficients | cannot have arbitrary values; they are subjected to all the constraints, including these five powerful constraints. It remains to investigate the restriction of these constraints upon the values of the in-between | Fourier coefficients | (Palmer, 1994). Method of projection may be considered (Youla, 1987; Levi & Stark, 1987; Elser, 2003).

Millane (1996), Sect. 3 shows that the oversampling ratio is 4 for three dimensions. Miao et al. (1998), Sect. 2 show that the oversampling ratio > 2. In the present article, for one-, two- or three-dimensions ((B) and (D)), the oversampling ratio is exactly 2. Hence, for two-dimensional object periodic along one direction and three-dimensional object periodic along one direction (fibre, (E) and (F)), or two directions (membrane (G)), no interpolation is needed. The magnitude of the Fourier transform of the object for all ω corresponding to the remaining directions can be measured experimentally; no interpolation is used. These are important cases of determining periodic structures without using interpolation. The electron densities obtained are unique and exact. The power of the periodicity of the structure is fully utilized so that the measured intensity is greatly enhanced, while in-between | Fourier coefficients | are not needed. The result will show whether the object has sharp boundary or not. This is important for system of atoms.

The periodicity of the structure greatly enhances the diffraction intensity which can be measured accurately, and introduces the strong constraint (i), the very regular shape of a unit cell and the accurately measured unit cell constants ($a, b, c, \alpha, \beta, \gamma$).

For freely rotating real molecule and fibre, a common method is to fit models of atoms to the experimental diffraction intensity. Chemical formula of the molecule, bond lengths, bond angles, and molecular dynamics may be used (Wang & Stubbs, 1993). Now, we may go one step further. For example, the unique structure $g(r)$ in (14), obtained from electron diffraction intensity is a direct determination of the spherically averaged electron density of gas-phase molecule. It can be treated as fundamental data. Models of atoms are fitted such that its spherical averaging gives this $g(r)$. Here chemical and physical knowledge of atoms are employed. The atomic coordinates are then obtained. This fitting is more direct, has clearer physical meaning, easier to visualize and perform, and saves much computation.

We only use the fact that the diffraction pattern is the Fourier transform of the object. The nature of the constituent of the object is not needed, in particular, whether the object is non-negative or not. For freely rotating real molecule (D) or centrosymmetric fibre (E), $f(x_1, x_2, x_3)$ is the spherically or cylindrically averaged electron density. If the electron density is negative in a region, this negative region may be lost in the averaging such that the resulting $f(x_1, x_2, x_3)$ is non-negative. Such averaging does not occur in the fixed centrosymmetric real fibre molecule (F) and the planar centrosymmetric periodic real membrane (G). If the constituent is negative in any region, this will be exhibited. In X-ray crystallography, electron density is assumed non-negative. This can be verified.

The method presented does not assume the concept of atoms. Atomic scattering factor and other knowledge of physics and chemistry are not employed. Electron density is determined solely from diffraction intensity. It is a direct picture of the electron density of the atom. In the case of X-ray crystallography, the goal of this method is to determine the electron density. It fully utilizes the power of X-ray diffraction, and the electron density of the crystal is ‘observed’ directly. The atomic positions will come out in the results. The observed electron density can be compared with quantum mechanical result. This is a direct verification of the quantum theory. Atomic scattering factor can be calculated and compared with the literature.

If algorithms applicable to experimental electron diffraction data of molecules and X-ray crystallographic data are developed, the authors may re-determine or refine the spherically averaged electron densities of the molecules and the electron densities of the centrosymmetric crystal structures in the literature (Yuen, 2011a). Experimental measurement of the non-Bragg intensities of a crystal may be used (Yuen, 2011d). The atomic coordinates can then be obtained. The results can be compared with those in the literature, in particular, the electron densities. Phase retrieval problem occurs in many disciplines (see, for example, Bates et al., 1991, Table 6.2). The above method can be applied.

If the electron density and structure of the biological molecules are determined, it is helpful in understanding biochemical processes, including diseases. The electron density determines the active site of the molecule. Suitable medicine may then be found. It is important to determine the structure of a macromolecule in solution because its configuration may be different from that in crystal. More work is needed in the area of measuring accurately the scattering intensity from a biological cell or molecule. For example, lenslets microscope and lenslets telescope in Yuen (2011e) may be used. Short exposure of weak radiation can be used to avoid damage to the cell or molecule. During each short exposure, some diffraction rods are measured.

In quantum mechanics, the electron density of an object extends to infinity (wave character). The above method solves the phase problem by employing the constraint of finite size (particle character). This may be due to experimental limitation or the object does have finite size. It is very interesting to determine whether atoms, molecules and nuclei have sharp boundary. This can be determined from a number of experimental results of many objects, in particular, from the freely-rotating gas molecules, the centrosymmetric real fibre (E) or fixed fibre molecule (F), and the planar centrosymmetric periodic real membrane (G). If atoms, molecules and nuclei do have finite size, in the diffraction process the wave-particle duality of the incident radiation may couple with the wave-particle duality of the object. The incident radiation exhibits wave character while the object exhibits particle character, finite size. Then it is interesting to investigate the conditions under which the incident radiation and the object exhibit wave or particle character, so that wave-wave, wave-particle, or particle-particle interactions will actually occur.

It is useful and important to obtain an electron density equation for atoms, molecules and crystals. This can be obtained by analytically solving the difficult problem of Schrödinger equation for many-particle systems or by employing the density-functional theory (Kohn, 1999). Alternatively, we can obtain an electron density equation from the electron density of many atoms, molecules and crystals determined from the diffraction intensity. Then we can develop a theory to explain this equation. We can have more understanding of the physics involved. The electron density determined is regarded as fundamental data. All acceptable theories, including quantum mechanics, should reproduce these data.

4. Conclusions

As a conclusion, the main result of this article is to show that in scattering and diffraction of centrosymmetric real object, the phase is not lost. It is just hidden in the magnitude. An analytical and unique solution of the multidimensional phase problem can be obtained. This article and the two articles in sequel, Yuen (2011b, 2011c) form a systematic and comprehensive treatment of the important phase-retrieval problem for real and complex objects. They show that mathematically, a solution is possible. This is the first step. They form a starting point. The next step is to apply the method to real data and many researches can be done. In particular, the authors may use all the experimental electron diffraction data of gas-phase molecules, and X-ray diffraction data of centrosymmetric crystals in the literature and re-determine or refine the electron densities and atomic co-ordinates. One of the purposes of these articles is to stimulate interest in the method presented and the many subsequent researches. In order to develop algorithms applicable to real data, researchers with expertise in numerical computation may develop algorithms which can be used by researchers in electron diffraction of gas-phase molecules and crystallographers. Researchers with expertise in mathematical analysis may also find an expression for the solution so that flipping of zeros is not needed. These articles also show that the oversampling ratio is exactly 2. This will be useful for works in iterative algorithms.

Appendix

Let $f_0(x)$ be an atom with size from $-d/2$ to $d/2$ and Fourier transform $F_0(\omega)$. For two identical atoms from $-d$ to 0 and 0 to d,

$$f_1(x) = f_0(x+\frac{d}{2}) + f_0(x-\frac{d}{2})$$

$$F_1(\omega) = 2F_0(\omega)\cos(\omega d/2)$$

Now extend $f_1(x)$ to a periodic function with period $= 2d$.

$\omega_{01} = 2\pi/(2d) = \pi/d$.

The Fourier coefficients occur at $m\omega_{01}$, $m = 0,\pm1,\pm2.,\dots$.

$F_1(m\omega_{01}) = 2F_0(m\pi/d)\cos(m\pi/2)$

For n pairs of atoms from $-nd$ to nd ,

$$f_n(x) = f_0(x + (n - \frac{1}{2})d) + \dots + f_0(x - (n - \frac{1}{2})d)$$

$$F_n(\omega) = 2F_0(\omega)\left[\cos(\omega d/2) + \dots + \cos(\omega(n - \frac{1}{2})d)\right]$$

Now extend $f_n(x)$ to a periodic function with period $= 2nd$.

$\omega_{0n} = 2\pi/(2nd) = \pi/(nd)$.

The Fourier coefficients occur at $m\omega_{0n}$, $m = 0,\pm1,\pm2.,\dots$

$$F_n(m\omega_{0n})$$
$$= 2F_0(m\pi/(nd))\{\cos[(m\pi/n)(1/2)] + \dots + \cos[(m\pi/n)(n-1/2)]\}$$

For $m = 0$, $F_n(0) = 2nF_0(0)$

$|F_n(\omega)|$ will be larger in a region about the origin and gradually decreases to zero as $\omega \to \pm\infty$. Δx is larger. $\Delta\omega$ is smaller. The uncertainty principle, $\Delta x \Delta\omega \geq \sqrt{\pi/2}$, is satisfied. For large n , ω_{0n} is small. Hence the in-between $|$ Fourier coefficients $|$ obtained by interpolation are more accurate. The method is very suitable for crystals with large unit cell.

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2

Ab initio determination of electron density of gas-phase molecule from the magnitude in electron diffraction

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Abstract

Phase-retrieval method may be used to determine the spherically symmetric electron density of gas-phase molecules from electron diffraction intensity directly. The spherically symmetric electron densities of the molecules obtained can be regarded as fundamental data. It can be used for fitting of model of molecules. The atomic coordinates can then be obtained. The result shows whether atoms and molecules have finite extent or sharp boundary. This is a direct verification of quantum chemistry.

Keywords: Phase retrieval; unique solution.

1. Introduction

Electron diffraction is an important tool in the determination of gas-phase molecular structures. It is discussed in the books of Vilkov et al. (1983), Hargittai & Hargittai (1988) and the reviews Hargittai (1992a), (1992b), Karle (1994). Hargittai (2000) provides a review of molecular structure of gas-phase metal halides. Spiridonov et al. (2001) review the determination of molecular structure in terms of potential energy functions from electron diffraction. Zeqail (2006) presents a review of 4D ultrafast electron diffraction, crystallography, and microscopy. Gundersen et al., 2007 present a method for improved sector correction. Vishnevskiy (2007a), (2007b) deal with the initial processing of the gas electron diffraction data. Habershon & Zewail (2006) present a genetic algorithm for determining molecular structures and conformation directly from electron diffraction. Structure of many gas-phase molecules have been determined by electron diffraction (see, for example, Morino et al., 1968; Rankin, 1982;

Vilkov et al., 1988; Mastryukov et al., 1999; Blockhuys et al., 2001; Johansen et al., 2001; Downs et al., 2002; Hagen et al., 2002; Kuze et al., 2002; Della Védova et al., 2004; Oberhammer et. al., 2004; Aarset et al., 2005; Hedberg, 2005; Hinchley et al., 2005; Kuchitsu, 2005; Naumov et al., 2005; Egawa et al., 2006; Girichev et al., 2006; Giricheva et al., 2006; Lu et al., 2006; Petrov et al., 2006; Tang et al., 2006; Bodi et al., 2007; Dorofeeva et al., 2007; Masters et al., 2007; Montejo et al., 2007; Samdal et al., 2007; Shen et al., 2007; Shlykov et al., 2007; Takeshima et al., 2007; Takeuchi et al., 2007; Tutolomondo et al., 2007;Ezhov, et al., 2008; Hnyk et al., 2008; Hölbling et al., 2008; Keys et al., 2008; Noble-Eddy et al., 2008; Shishkov et al., 2008; Strenalyuk et al., 2008; Traetteberg et al., 2008; Vest et al., 2008; Vishnevskiy et al., 2008; Vogt et al., 2008; Wann et al., 2008; Zakharov et al., 2008). For the MOGADOC database of gas-phase molecular structures determined by electron diffraction, see Vogt & Vogt (2004). van der Waal (1999) reviews the structure analysis of large argon clusters from gas-phase electron diffraction data.

In determination of gas-phase molecular structures by electron diffraction, a common method is to fit models of atoms to the experimental diffraction intensity. Chemical formula of the molecule, bond lengths and bond angles may be used. The radial distribution is related to the probability density distribution of internuclear distances. If the number of atoms is large, the number of internuclear distances is very large. There are many overlapping peaks. The determination of the molecular structure may be difficult. The majority of the structure determination is for molecules containing less than, typically about 80 non-hydrogen atoms (Habershon & Zewail, 2006, p. 353). The main difficulty is that only the magnitude, not the phase, of the diffracted wave is measured. If the phase can be measured or retrieved by other means, the spherically averaged electron density can be obtained directly from the measured intensity of the diffracted wave, this will simplify greatly the model fitting.

An analytical solution for the phase problem of centrosymmetric real object (for example, a gas-phase molecule) is presented in Yuen (2011a). This can be applied to the electron-diffraction intensity. The unique spherically averaged electron density is determined.

2. Phase-retrieval method

Constraints used:

(i) $f(x_1, x_2, x_3)$ has finite size

(ii) $f(x_1, x_2, x_3)$ is real

(iii) $f(x_1, x_2, x_3)$ is centrosymmetric

(iv) $f(x_1, x_2, x_3)$ is bounded

For a freely rotating molecule, the electron density is the spherically averaged electron density $g(r)$. $g(r) = f(x_1, x_2, x_3)$

where $r^2 = x_1^{\ 2} + x_2^{\ 2} + x_3^{\ 2}$.

Let $F(\omega_1, \omega_2, \omega_3)$ be the Fourier transform of $f(x_1, x_2, x_3)$.

The magnitude of $F(\omega_1, \omega_2, \omega_3)$ is the square root of the intensity obtained in electron-diffraction.

From Yuen (2011a), (13),

$$F(\omega_1, \omega_2, \omega_3) = \int_{-R}^{R} \left\{ \iint f(x_1, x_2, x_3) \cos(\omega_2 x_2 + \omega_3 x_3) dx_2 dx_3 \right\} \exp(-i\omega_1 x_1) dx_1 \tag{1}$$

where the double integral inside the curly brackets is over a circular section of the sphere of radius R. Fix $\omega_2 = \omega_3 = 0$.

$$F(\omega_1, 0, 0) = \int_{-R}^{R} \left\{ \iint f(x_1, x_2, x_3) dx_2 dx_3 \right\} \exp(-i\omega_1 x_1) dx_1 \tag{2}$$

Denote the term in curly brackets in (2) by $q(x_1)$.

$$F(\omega_1, 0, 0) = \int_{-R}^{R} q(x_1) \exp(-i\omega_1 x_1) dx_1$$

$q(x_1)$ is centrosymmetric, non-negative, bounded and has finite size with size from

$-R$ to R

From Yuen (2011a), (A), unique value of the phase of $F(\omega_1,0,0)$ and the value of R, can be obtained from the magnitude of $F(\omega_1,0,0)$ for $-\infty < \omega_1 < \infty$, by means of the phase-retrieval method.

The object has spherical symmetry.

Let $U(\omega) = F(\omega_1, \omega_2, \omega_3)$

where $\omega^2 = \omega_1{}^2 + \omega_2{}^2 + \omega_3{}^2$.

Hence $U(\omega) = F(\omega,0,0)$ is obtained.

$U(\omega)$ is the form factor, or for an atom, atomic scattering factor. The unique spherically averaged electron density is then given by

$$g(r) = \frac{1}{2\pi^2}\int_0^\infty U(\omega)\omega\frac{\sin\omega r}{r}d\omega \qquad (3)$$

This is a direct determination of the spherically averaged electron density. From this electron density, the atomic coordinates are then obtained from knowledge of the chemical formula of the molecule, and values of bond lengths and bond angles.

This method may be applied to all the experimental electron diffraction data of molecules in the literature. All the spherically averaged electron densities of the molecules and the atomic coordinates can be re-determined by the authors. The results may be compared with those in the literature.

3. Discussions

For freely rotating real molecule, a common method is to fit models of atoms to the experimental diffraction intensity. Chemical formula of the molecule, bond lengths and bond angles may be used. Now, we may go one step further. The unique structure $g(r)$ in (3), obtained from electron diffraction intensity is a direct determination of the spherically averaged electron density of gas-phase molecule. It can be treated as fundamental data. Models of atoms are fitted such that its spherical averaging gives this $g(r)$. Here chemical and physical knowledge of atoms are employed. The atomic coordinates are then obtained. This fitting is more direct, has clearer physical meaning, easier to visualize and perform, and saves much computation. The phase-retrieval result may also be useful in structure analysis of large clusters (van de Waal, 1999).

Quantum chemistry says that electron density extends to infinity. If the spherically averaged electron densities obtained have finite size, quantum chemistry needs to be re-examined. This is a direct verification of quantum chemistry. The electron density may taper to zero. It is more interesting if it has sharp boundary.

It is a common belief that the electron density in atoms and molecules is non-negative. This ‘positivity constraint’ is used in crystal structure analysis. It would be more convincing if this can be verified experimentally. The phase-retrieval method for centrosymmetric real object presented in Yuen (2011a) does not require any knowledge of physics and chemistry of the atoms and molecules, including the non-negative condition. If atoms and molecules have regions of negative electron density with large magnitude, the negative region will not be lost in the spherical averaging. It will exhibit in the result. If the negative region has small magnitude so that it does not appear in the spherical average, we can use the fixed centrosymmetric real fibre molecule or the planar centrosymmetric periodic real membrane (Yuen, 2011a, (F), (G)).

In astronomy, the structure of 2-dimensional circularly symmetric objects can also be determined by phase-retrieval method (Yuen (2011a), (C)).

The treatment in this article can be compared with that in Yuen (2011b).

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3

Ab initio determination of electron densities of centrosymmetric non-negative crystals from diffraction intensities in X-ray crystallography. Iterative algorithms with oversampling

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Abstract

For centrosymmetric real crystal, the phase of the diffracted wave is not lost. It is just hidden in the magnitudes of the diffracted waves. It can be retrieved from the magnitudes and unique result is obtained. Hence the electron density can be obtained directly from the diffraction intensities in X-ray crystallography. The oversampling ratio is exactly 2. Values of in-between ∣Fourier coefficients∣ for one index l, m or n is sufficient for determination of the unique electron density. Values of the in-between ∣Fourier coefficients∣ for the three indices l, m and n are not independent. They must be consistent so that the electron densities obtained for each index are identical. This consistent condition must be satisfied. The process of phase retrieval is time-consuming. However, it provides the theoretical foundation and gives the conditions that iterative algorithms with oversampling will give a unique solution. The in-between Fourier coefficients calculated from crystal structure reported in the literature are reasonably good. For crystal structure reported in the literature (R-index ≠ 0), iterative algorithms with oversampling can be used to refine the electron density. The iterative algorithms will converge to a unique *ab initio* non- negative electron density with R-index = 0. This is an improvement of the electron density reported in the literature. The in-between ∣Fourier coefficients∣ are also improved because of the consistent condition. For crystal structure which has not been solved, its electron density with R-index = 0 can also be determined by means of iterative algorithms with oversampling. The *ab initio* electron density determined may be unique. It is determined by the experimental lattice constants, diffraction intensities and initial in-between ∣Fourier coefficients∣ (or mid-Bragg intensities); no other chemical or physical information (for example, atomicity, chemical formula, bond length, bond angle, etc) is used. From the refined electron density with R-index = 0, atomic

coordinates are obtained which is an improvement of the atomic coordinates obtained by conventional methods in X-ray crystallography. If some of the light atoms do not appear, or some bond lengths and bond angles are different from those obtained by other means, we can use chemical or physical informations, including the atomic coordinates and electron density determined in conventional X-ray crystallography to improve the in-between｜Fourier coefficients｜ and/or the｜Fourier coefficients｜ within their uncertainties to obtain all the atoms and reasonable bond lengths and bond angles.

Keywords: Phase retrieval; unique solution; consistent condition; iterative algorithms; oversampling; in-between｜Fourier coefficients｜; mid-Bragg intensities; refinement of electron density; improvement of｜Fourier coefficients｜; racemate; macromolecule; protein; polypeptide; DNA.

1. Introduction

This is a sequel of Yuen (2011a).

Determination of the structures of macromolecules, for example, proteins, nuclei acids, protein-nuclei acid complexes, viruses, is very important, particularly in medicine and biology. Macromolecular crystallography is discussed in the books by Blundell & Johnson (1976), McRee (1999), Drenth (2007) and Read & Sussman (2007). Reviews of macromolecular crystallography is presented in Dauter (2003) and Fan (2005). Methods for solving crystal structures are presented in the book by Woolfson & Fan (1995). Liu et al. (1999) combine direct methods with isomorphous replacement or anomaolous scattering data. Delarue, (2000) presents resolution of the phase-ambiguity problem by Monte Carlo methods. Delarue & Orland (2000) discusses a statistical thermodynamics approach for phase combination and phase refinement.

The phase of the structure factors of a centrosymmetric crystal can only be 0 or π. Hence it is easier to solve the phase problem, and the result is more accurate.

In general, only L-amino acids are found in natural proteins. Hence protein crystals are non-centrosymmetric. Mackay (1989) suggests that centrosymmetric racemic crystals can be synthesized.

Structures of centrosymmetic crystals may be determined by molecular replacement methods. The structure of racemic rubredoxin is determined in Zawadzke & Berg

(1993). The racemic DL-monellin is determined in Hung et al. (1999). The structure of enantiomorphic DNA is presented in Doi et al. (1993). Rypniewski et al. (2006) determines the structure of an RNA racemate.

Pentelute et al. (2008) has determined the structure of a centrosymmetric racemic crystal of snow flea antifreeze protein. The structure of a quasi-racemate crystal is determined by multiple wavelength anomalous dispersion (MAD) phasing. The crystal structure of L-sfAFP is determined by molecular replacement.

Structures of centrosymmetic crystals may be determined by direct methods. Doi et al. (1994) presents the structure of a polypeptide racemate, Leu-enkephalin. The structure of racemic trichogin A IV is determined in Toniolo et al. (1994). The structure of D,L-Alpha-1 is determined in Patterson et al. (1999). Mandal et al. (2009a) and Mandal et al. (2009b) present the structures of racemic proteins. Datta et al. (1997) presents the structure of a tripeptide. Karle et al. (1997) presents the structure of a glycine rich peptide. Structures of homooligopeptides are presented in Tanaka et al. (1999). De Simone et al. (2000) presents the structure of a *Dcp*-containing peptide. Kaul et al. (2000) presents structure of a centrosymmetric tetrapeptide. Lombardi et al. (2000) presents the structure of Afc-containing peptides. Tanaka et al. (2001) presents the structure of a tripeptide. The structures of racemates 1/2, 3/4, and 5/6 of tripeptides are presented in Saha et al. (2008). Stamm et al. (2003) determines the structures of two racemic benzamides. Structures of two racemic monoperoxo complexes of vanadium(v) are determined in Tatiersky et al. (2004).

For reviews of racemic crystallography, see Berg & Zawadzke (1994), Lamzin et al. (1995), Berg & Goffeney (1997) and Matthews (2009).

In the molecular replacement methods, multiple wavelength anomalous dispersion (MAD) phasing and direct methods, it is assumed that the phase of the diffracted wave is lost. Atomicity, bond lengths, bond angles and non-negativity of electron density are used so that the atomic co-ordinates can be determined. The electron density so obtained is less accurate. As the electron density determines the active site of biological molecules, their interactions and the biological activities, it is important to determine the electron density accurately.

For centrosymmetric real crystal, it is found that the phase is not lost. It is just hidden in the magnitudes of the diffracted wave (Yuen (2011a), (D)). It can be retrieved from the magnitude (Sect. 2.1). Unique result is obtained. Hence the electron density can be obtained directly from the diffraction intensities in X-ray crystallography. This can be applied to protein, DNA, and other crystals. The oversampling ratio is exactly 2. Values of in-between | Fourier coefficients| for one index l, m or n is sufficient for determination of the unique electron density. Values of the in-between | Fourier coefficients| for the three indices l, m and n are not independent. They must be consistent so that the electron densities obtained for each index are identical. This consistent condition must be satisfied.

For crystal structure reported in the literature, the primary result of conventional X-ray crystallography is determination of atomic coordinates. The electron density is not so accurate. As the Fourier transform involves integration over the unit cell, it is a group property. The in-between Fourier coefficients calculated from crystal structure reported in the literature is more reliable than individual value of the electron density. They are reasonably good. Magnitudes of these in-between Fourier coefficients can be used for phase retrieval (Sect 2.2).

Much work has been done by many researchers with iterative algorithms and oversampling, and has gained much empirical success. For references in iterative algorithms and oversampling, see Yuen (2011a, 2011b, 2011c, 2011d, 2011e, 2011f).

As shown in the phase-retrieval method in Yuen (2011a, 2011c, 2011d), just using the magnitudes of the in-between Fourier coefficients (the commonly called oversampling) is not sufficient to give a unique solution to the phase problem. For a nonperiodic object, even if we know the magnitude of its Fourier transform, but not its phase, the structure of the object cannot be determined. Hence it is necessary to find the conditions that iterative algorithms with oversampling will provide a unique solution. It is stated in the review of Thibault & Elser (2010), p. 244 that 'It is probably fair to say that for every successful reconstruction, tens of diffraction patterns have failed to yield any results whatsoever'. Yuen (2011a, 2011c, 2011d) show that certain general and reasonable constraints have to be imposed on the object. The process of phase retrieval in Sect. 2.1 is time-consuming. However, it provides the theoretical foundation and gives the conditions that iterative algorithms with oversampling will give a unique or probably unique solution.

The degree of oversampling required is discussed in p. 240 of the review Thibault & Elser (2010). It is stated that 'no further information can be gained beyond a certain degree of sampling of the intensity'. Zayed (1993), p. 22 states that oversampling greater than the Nyquist rate can reduce interpolation noise level due to noisy data, the sampled values are dependent and the signal reconstruction is not affected by losing an arbitrarily large but finite number of samples values (Marks (1991), Sect. 4.1.1, 5,1,1).

For crystal structure reported in the literature (R-index $\neq 0$), iterative algorithms with oversampling can be used to refine the electron density (Sect. 2.3). The iterative algorithms will converge to a unique *ab initio* non-negative electron density with R-index = 0. This is an improvement of the electron density reported in the literature. The in-between $|$ Fourier coefficients $|$ are also improved.

For crystal structure which has not been solved, its electron density with R-index = 0 can also be determined by means of iterative algorithms with oversampling (Sect 2.4). The *ab initio* electron density obtained may be unique.

The electron density obtained has R-index = 0. This means that the uncertainty of the experimentally measured $|$ Fourier coefficients $|$ is not taken into account. If some of the light atoms do not appear, or some bond lengths and bond angles are different from those obtained by other means, we can use chemical or physical informations, including the atomic coordinates and electron density determined in conventional X-ray crystallography to improve the in-between $|$ Fourier coefficients $|$ and/or the $|$ Fourier coefficients $|$ within their uncertainties to obtain all the atoms and reasonable bond lengths and bond angles (Sect. 2.5).

This article can be applied for determination or refinement of electron density of proteins, polypeptides and DNA.

2.1. Phase-retrieval method. Consistent condition

For a crystal with electron density $f(x_1, x_2, x_3)$, we deal with the content of the basic unit cell of the crystal as an object, and define $f_0(x_1,x_2,x_3) = f(x_1,x_2,x_3)$ for (x_1,x_2,x_3) ε the basic unit cell, or = 0 otherwise. Choose the cell, $-a/2 \le x_1 \le a/2$, $-b/2 \le x_2 \le b/2$, $-c/2 \le x_2 \le c/2$ as the basic unit cell. The phase of the diffraction points will be retrieved from the magnitude by imposing some general and reasonable constraints upon the object. For centrosymmetric non-negative crystal, the constraints used are:

(i) $f_0(x_1,x_2,x_3)$ has finite size with size = the known unit cell constants, $a,b,c,\alpha,\beta,\gamma$

(ii) $f_0(x_1,x_2,x_3) \ge 0$

(iii) $f_0(x_1,x_2,x_3)$ is centrosymmetric

(iv) $f_0(x_1,x_2,x_3)$ is bounded

Let $F_0(\omega_1,\omega_2,\omega_3)$ be the Fourier transform of $f_0(x_1,x_2,x_3)$.

Let $\omega_{10} = 2\pi/a$, $\omega_{20} = 2\pi/b$, $\omega_{30} = 2\pi/c$. $\left|F_0(l\omega_{10},m\omega_{20},n\omega_{30})\right|$ are measured experimentally.

$$F_0(l\omega_{10},m\omega_{20},n\omega_{30}) =$$

$$\int_{-a/2}^{a/2}\int_{-b/2}^{b/2}\int_{-c/2}^{c/2} f_0(x_1,x_2,x_3)\exp[-i(l\omega_{10}x_1 + m\omega_{20}x_2 + n\omega_{30}x_3)]dx_1dx_2dx_3 =$$

$$\int_{-a/2}^{a/2}\left\{\int_{-b/2}^{b/2}\left[\int_{-c/2}^{c/2} f_0(x_1,x_2,x_3)\cos(m\omega_{20}x_2 + n\omega_{30}x_3)dx_3\right]dx_2\right\}\exp(-il\omega_{10}x_1)dx_1 \tag{1}$$

For fixed $m = m'$, and $n = n'$,

$$F_0(l\omega_{10},m'\omega_{20},n'\omega_{30}) = \int_{-a/2}^{a/2} q(x_1,m'\omega_{20},n'\omega_{30})\exp(-il\omega_{10}x_1)dx_1$$

Obtain $\left|F_0((l+1/2)\omega_{10},m'\omega_{20},n'\omega_{30})\right|$, $l = 0,\pm1,\pm2.,\ldots$, by interpolation in multidimensions (Bates (1984), p. 216, Kincaid & Cheney (1996), Sect. 6.10, Cheney

& Light (2000), Anita (2002), Sect. 4.7, 4.8, Hau-Riege et al. (2004)) or by experimental measurement (Yuen (2011g)). For fixed m', n', the term in curly brackets in (1), $q(x_1, m'\omega_{20}, n'\omega_{30})$ is a one-dimensional centrosymmetric real object, a function of x_1, with size from $x_1 = -a/2$ to $x_1 = a/2$. This is a one-dimensional problem. As shown in Yuen (2011a), (B), a unique $F_0(l\omega_{10}, m'\omega_{20}, n'\omega_{30})$ is obtained from $|F_0(l\omega_{10}, m'\omega_{20}, n'\omega_{30})|$ for all l. Repeat this for all m, n, get all $F_0(l\omega_{10}, m\omega_{20}, n\omega_{30})$. The Fourier series gives a unique electron density $f(x_1, x_2, x_3)$.

The interpolated in-between | Fourier coefficients | are adjusted so that the electron density satisfies constraints:

(v) $f(x_1, x_2, x_3)$ is continuous with

$$f_0(-a/2, x_2, x_3) = f_0(a/2, x_2, x_3) \quad \text{for all } x_2, x_3$$

$$f_0(x_1, -b/2, x_3) = f_0(x_1, b/2, x_3) \quad \text{for all } x_1, x_3$$

$$f_0(x_1, x_2, -c/2) = f_0(x_1, x_2, c/2) \quad \text{for all } x_1, x_2$$

(vi) the crystal may have other symmetry elements.

The | Fourier coefficients | are obtained from the experimentally measured intensity. As the | Fourier coefficients | are used in obtaining the electron density, the R-index = 0.

We have used in-between | Fourier coefficients | in only one index l. The oversampling ratio is exactly 2. The in-between | Fourier coefficients | have uncertainties. From the electron density obtained, we can calculate the in-between | Fourier coefficients | for indices m, n. In general, these do not agree with the values obtained by interpolation. Use interpolated in-between | Fourier coefficients | indices m and then n. Repeat the phase-retrieval process and obtain two more electron densities.

Values of in-between ∣Fourier coefficients∣ for one index l, m or n (oversampling ratio = 2) is sufficient for determination of the unique electron density. Values of the in-between ∣Fourier coefficients∣ for the three indices l, m and n are not independent. They must be consistent so that the electron densities obtained for each index are identical. This consistent condition must be satisfied.

Obtain the average of these three electron densities. Adjust the in-between ∣Fourier coefficients∣ within their uncertainties so that the electron densities obtained for each index are identical and it satisfies the six constraints with R -index = 0.

2.2. Refinement of electron density of crystal structure reported in the literature (R -index $\neq 0$). The calculated in-between Fourier coefficients are reasonably good

For crystal structure reported in the literature, the primary result of conventional X-ray crystallography is determination of atomic coordinates. The electron density is not so accurate. The phase-retrieval method in Sect. 2.1 is to obtain the electron density directly. It can be used to refine the electron density of crystal structure reported in the literature. Use the reported electron density in the basic unit cell. From its Fourier transform, calculate the in-between Fourier coefficients. As the Fourier transform involves integration over the unit cell, it is a group property. The calculated in-between Fourier coefficients are more reliable than individual value of the electron density. They are reasonable good. Use magnitudes of these in-between Fourier coefficients and apply Sect. 2.1, an electron density with R -index = 0 is obtained. It is an improvement of that reported in the literature.

The ∣Fourier coefficients∣ and in-between ∣Fourier coefficients∣ calculated from an electron density, satisfy the consistent condition in Sect. 2.1. In general, the experimentally measured ∣Fourier coefficients∣ and the calculated in-between ∣Fourier coefficients∣ , do not satisfy the consistent condition in Sect. 2.1.

2.3. Iterative algorithms with oversampling. Refinement of electron density of crystal structure reported in the literature (R -index $\neq 0$)

As the process in Sect. 2.1 is time-consuming, iterative algorithms with oversampling may be employed. Much work has been done by many researchers with iterative algorithms and oversampling, and has gained much empirical success. The phase-retrieval method in Sect. 2.1 provides the theoretical foundation and gives the conditions that iterative algorithms with oversampling ratio =2 will lead to the unique electron density. Hence we have an important conclusion of this article:

For a crystal structure reported in the literature, calculate the Fourier coefficients and in-between Fourier coefficients from the electron density in the basic unit cell. Use their phases (0 or π) as initial phases.

Use

1. the experimentally determined unit cell constants and ∣Fourier coefficients∣,
2. initial in-between ∣Fourier coefficients∣ obtained from the electron density reported in the literature with R-index $\neq 0$,
3. the initial phases,
4. ∣Fourier coefficients∣ and initial in-between ∣Fourier coefficients∣ for all (l,m,n), and all ($l+\frac{1}{2}$, and/or $m+\frac{1}{2}$, and/or $n+\frac{1}{2}$). The oversampling ratio is $2^3=8$.
5. the six constraints in Sect. 2.1, plus constraint
   (vii) the sharp-boundary constraint. The boundary values at the six boundary surfaces have the same order of magnitude as the value $f_0(x_1,x_2,x_3)$ inside a unit cell.
6. Although, as shown in Sect. 2.2, the initial in-between ∣Fourier coefficients∣ calculated from the electron density reported in the literature are reasonably good, they still have uncertainties much larger than those of the experimental ∣Fourier coefficients∣, and that they are not independent. The ∣Fourier coefficients∣ and the in-between ∣Fourier coefficients∣ need to satisfy the consistent condition in Sect. 2.1. Hence, in the final iterations, the in-between ∣Fourier coefficients∣ are relaxed or adjusted, within their uncertainties, to satisfy this consistent condition. Therefore we have obtained in-between ∣Fourier coefficients∣ more accurate than

those calculated from the electron density reported in the literature.

The iterative algorithms will converge to a non-negative electron density with R -index = 0. The *ab initio* electron density is unique for this set of | Fourier coefficients | and initial in-between | Fourier coefficients | . This is an improvement of the electron density reported in the literature.

Crystal structures determined by conventional methods in X-ray crystallography provide accurate atomic ordinates. The electron density is less accurate. Since the structure has been refined by conventional methods in X-ray crystallography, the electron density should not be very different from the true electron density. Using the electron density reported in the literature as the initial electron density, it is reasonable to expect that the iterations will converge to a unique and improved electron density. As explained in Sect. 2.2, the initial in-between | Fourier coefficients | obtained from the reported electron density is more reliable than individual value of the electron density. Using these initial in-between | Fourier coefficients | , the electron density obtained by iterative algorithms with oversampling may show some light atoms which are missed in the electron density reported in the literature. From the refined electron density with R -index = 0, atomic coordinates are obtained which is an improvement of the atomic coordinates reported.

2.4. Iterative algorithms with oversampling. Determination of electron density of crystal structure which has not been solved

For a crystal structure which has not been solved, obtain the initial in-between | Fourier coefficients | by interpolation in multidimensions or by experimental measurement. Use the phase-retrieval method in Sect. 2.1, and a small $\omega_{\max}$ to obtain a gross structure (Yuen (2011c), Sect. 2.3). Calculate the | Fourier coefficients | and in-between | Fourier coefficients | . Use their phases as initial phases. Process similar to Sect. 2.3 can then be applied. The uncertainties of the in-between | Fourier coefficients | may be larger than those of the | Fourier coefficients | . The iterative algorithms will converge to a non-negative electron density with R -index = 0. The in-between | Fourier coefficients | are also improved because of the consistent condition.

The process of obtaining a gross structure may be time-consuming. We may use random initial phases (0 or π) which are commonly employed. The phase-retrieval method in Sect. 2.1 shows that oversampling ratio =2 leads to a unique electron density with R-index = 0. To compromise that proper initial phases are not used, more oversampling can be employed. We use | Fourier coefficients | and initial in-between | Fourier coefficients| for all (l,m,n), and all ($l+\frac{1}{2}$, and/or $m+\frac{1}{2}$, and/or $n+\frac{1}{2}$). The oversampling ratio is $2^3=8$. Iteration algorithms with oversampling in Sect. 2.3 will then be applied. The iterations will converge to an electron density with R-index = 0. The *ab initio* electron density may be unique. This is because of the known unit cell constants which is a tight support, and a very strong and powerful constraint, in particular for three-dimensional crystal; Millane (1996), p. 726 states that one might suspect that in going from two dimensions to three, the phase problem may go from being unique to being "more than unique". The converged electron density may be unique. This convergence can be investigated empirically and theoretically. As experience accumulates, we will learn how much oversampling is needed for the fastest and most efficient convergence of the iterations.

From the refined electron density with R-index = 0, atomic coordinates are obtained which is an improvement of the atomic coordinates obtained by conventional methods in X-ray crystallography.

2.5. Recovery of missing atoms and improvement of bond lengths and bond angles

The electron density obtained in Sect. 2.3 or 2.4 has R-index = 0. This means that the uncertainty of the experimentally measured intensity (or | Fourier coefficients|) is not taken into account. The experimentally measured | Fourier coefficients| do have uncertainties. The uncertainties of the in-between | Fourier coefficients| may be larger than those of the | Fourier coefficients| . If some of the light atoms (for example, hydrogen atom) do not appear, or some bond lengths and bond angles are different from those obtained by other means (for example, neutron diffraction, spectroscopy), and if we cannot find any good reason, such discrepancies may be due to uncertainties of the in-between |Fourier coefficients| and/or the |Fourier coefficients| . Hence we can use chemical or physical informations, including the atomic coordinates and electron density determined in conventional X-ray crystallography to improve first the in-between |Fourier coefficients| , then the |Fourier coefficients| . They are adjusted within their uncertainties to obtain all the atoms and reasonable bond lengths and bond angles.

3. Discussions and conclusion

The phase-retrieval method in Sect. 2.1 provides the theoretical foundation and gives the conditions that iterative algorithms with oversampling will give a unique solution.

Hence, an important conclusion is obtained:

For refinement of electron density of crystal structure reported in the literature R-index $\neq 0$), if we follow the process in Sect. 2.3, **the iterative algorithms will converge to a non-negative electron density with R-index = 0. The *ab initio* electron density is unique for this set of |Fourier coefficients| and initial in-between |Fourier coefficients| . This is an improvement of the electron density reported in the literature.** The electron density obtained by iterative algorithms with oversampling may show some light atoms which are missed in the electron density reported in the literature.

The in-between Fourier coefficients calculated from crystal structure reported in the literature are reasonably good. We urge that crystallographers will apply iterative algorithms with oversampling (Sect. 2.3) to refine electron densities of structures of the many centrosymmetric non-negative crystals reported in the literature, in particular, those crystal structures solved by themselves. Improved in-between |Fourier coefficients| and improved electron density with R-index = 0 can be obtained. The *ab initio* electron density is determined by the experimental lattice constants, diffraction intensities and in-between |Fourier coefficients| ; no other chemical or physical information (for example, atomicity, chemical formula, bond length, bond angle, etc) is used. Moreover, the in-between | Fourier coefficients | are also refined because of the consistent condition This is different from conventional X-ray crystallography, where atomicity, atomic scattering factor and chemical formula are used in structure determination. From the electron density with R -index = 0, the atoms, their species and coordinates can be determined. The atomic coordinates obtained is an improvement of the atomic coordinates obtained by conventional methods in X-ray crystallography. The chemical formula can then be obtained (Wu et al. (2004), p. 328, Cionca (2007), p. 4). This serves as an experimental method of obtaining the atomic species and the chemical formula, and an experimental verification that atomicity holds in crystals.

As we have obtained an electron density with R -index = 0, we have made full use of the | Fourier coefficients | . Hence it is meaningful to have Sect. 2.5: Recovery of missing atoms and improvement of bond lengths and bond angles. If any light atom is missed, or some bond lengths and bond angles are different from those obtained by other means (for example, neutron diffraction, spectroscopy), such discrepancies may be due to uncertainties of the in-between | Fourier coefficients | and/or the | Fourier coefficients | . These discrepancies are corrected by adjusting first the in-between | Fourier coefficients | , then the | Fourier coefficients | , within their uncertainities; the in-between | Fourier coefficients | and/or the | Fourier coefficients are improved.

For determining the electron density of crystals, the structures of which have not been solved by conventional methods in X-ray crystallography, random initial phases (0 or π) may be adopted (Sect. 2.4). To compromise that proper initial phases are not used, more oversampling is employed by using all the initial in-between | Fourier coefficients | . The oversampling ratio is $2^3 = 8$. The iterations may converge to a unique *ab initio* electron density because of the known unit cell constants, a very strong and powerful constraint. This convergence can be investigated empirically and theoretically. The atomic species and the chemical formula need not be known. They will be shown in the electron density obtained.

Yuen (2011a), Appendix shows that for crystals of large unit cells, the in-between | Fourier coefficients | obtained by interpolation is more accurate. In contrary to conventional methods in X-ray crystallography, the phase-retrieval method and the iterative algorithms with oversampling is very suitable for crystals of macromolecules, for example, proteins.

Oversampling ratio =2 is sufficient for determination of the unique electron density. Values of the in-between | Fourier coefficients | are not independent. They must be consistent so that the electron densities obtained for each index are identical. In the final iterations, the in-between | Fourier coefficients | are relaxed or adjusted, within their uncertainties, to satisfy this consistent condition. The in-between | Fourier coefficients | are improved.

Crystallographers who have reported preliminary X-ray diffraction studies of centrosymmetric non-negative crystals in the literature may apply Sect. 2.4 to determine the electron density of their crystals.

According to Yuen (2011a), the solution of the phase problem is unique for centrosymmetic, real, bounded and finite-size object; non-negativity is not needed. From the non-negative electron density obtained, if we stop using the constraint that the electron density is non-negative, and use the iterations and if we get the same result, this shows that the electron density is indeed non-negative.

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Ab initio determination of electron densities of centrosymmetric non-negative nonperiodic objects from diffraction intensities. Iterative algorithms with oversampling

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Abstract

For centrosymmetric non-negative nonperiodic object, the phase of the diffracted wave is not lost. It is just hidden in the magnitudes of the diffracted waves. It can be retrieved uniquely from the magnitudes. Hence the electron density can be obtained directly from the diffraction intensities. The oversampling ratio is exactly 2. We have a very rare result: Sets of experimental results cannot be independent; they needed to be adjusted according to theory. The process of phase retrieval is time-consuming. However, it provides the theoretical foundation and gives the conditions that iterative algorithms with oversampling will give a unique solution. For electron density of object reported in the literature, iterative algorithms with oversampling can be used to refine the electron density. The iterative algorithms will converge to a unique *ab initio* non-negative electron density with R-index = 0. This is an improvement of the electron density reported in the literature. For electron density of object which has not been solved, its electron density with R-index = 0 can also be determined by means of iterative algorithms with oversampling. The *ab initio* electron density obtained may not be unique. In all cases, the experimental ｜Fourier coefficients｜ and in-between ｜Fourier coefficients｜ are improved by means of the consistent condition.

Keywords: Phase retrieval; unique solution; consistent condition; rare result; superresolution; iterative algorithms; oversampling; refinement of electron density.

1. Introduction

This is a sequel to (Yuen 2011a, 2011b).

Much work has been done by many researchers in the determination of the electron density of a nonperiodic object.

Millimeter-wave spectroscopy is used in determination of molecular structure (Tinti et al. (2006)). Infrared spectroscopy is used in Borba et al. (2008). Electronic spectroscopy is used in Sunahori et al. (2007). Single-particle cryo-electron microscopy is presented in Elmlund et al. (2008). Image reconstruction is presented in Chung et al. (2010). Tomographic image alignment is presented in Chen et al. (2009). Density functional theory is used in Glossman-Mitnik (2007). Solution structures of proteins are presented in Gonçalves et al. (2010) and Salvay et al. (2010). An overview of proteins, lipids, and water in the gas phase is presented in van der Spoel et al. (2011). X-ray microscopy is reviewed in Kaulich et al. (2011). Potential for biomolecular imaging with femtosecond X-ray pulses is discussed in Neutze, et al. (2000). X-ray imaging is discussed in Shapiro (2008), Chapman (2009). Diffraction imaging of single particles and biomolecules is discussed in Huldt et al. (2003). Phasing diffraction data from a stream of hydrated proteins is presented in Wu & Spence (2005). Pełka (2008) and Pełka et al. (2010) discuss applications of synchrotron radiation and free electron lasers in biology and medicine. Bioimaging is reviewed in Pełka (2009). Kodama & Nakasako (2011) presents biological macromolecules enveloped by water in coherent x-ray diffraction microscopy. Larabell & Nugent (2010) reviews imaging cellular architecture with X-rays. Reviews of single-molecule imaging are presented in Hajdu (2000) and Sako & Uyemura (2002).

For centrosymmetric molecule, see Mondal et al. (1999). Vibration spectroscopic study is presented in Edwards & Lewis (1998). Infrared spectroscopy is employed in Okamoto (1999). Density functional theory and polarization-sensitive resonance coherent anti-Stokes Raman scattering spectroscopy are employed in Schlücker et al. (2001). Molecular orbital, semiempirical finite-field and time-dependent Hartree-Fock methods are employed in Yang & Jiang (2001a, 2001b).

For reviews about molecular structure, see Idrissi (2005) and Polavarapu (2007).

Electron and X-ray methods of ultrafast structural dynamics is reviewed in Gaffney & Chapman (2007) and Chergui & Zewail (2009). Pump-probe studies of photoexcited nanoparticles is discussed in Plech et al. (2007).

For centrosymmetric real nonperiodic object, it is found that the phase is not lost. It is just hidden in the magnitudes of the diffracted wave (Yuen (2011a), (C)). It can be retrieved uniquely from the magnitude (Sect. 2.1). Hence the electron density can be obtained directly from the diffraction intensities.

Much work has been done by many researchers with iterative algorithms and oversampling, and has gained much empirical success. The process of phase retrieval in Sect. 2.1 is time-consuming. However, it provides the theoretical foundation and gives the conditions that iterative algorithms with oversampling will give a unique solution.

The oversampling ratio is exactly 2. Values of the in-between | Fourier coefficients | for the three indices l, m and n are not independent. They must be consistent so that the electron densities obtained for each index are identical. This consistent condition must be satisfied. All the | Fourier coefficients | and in-between | Fourier coefficients | need to be adjusted within their experimental uncertainties so that the electron densities obtained for each index are identical. We have a very rare result: Sets of experimental results cannot be independent; they needed to be adjusted according to theory.

For iterative algorithms and oversampling, see Miao et al. (2002), Miao et al. (2003), Jurek et al. (2004), Shen et al. (2004), Faigel et al. (2005), Miao et al. (2005), Takahashi et al (2007), Barty et al. (2008), Bogan et al. (2008), Fill et al. (2008), Kamimura et al. (2008), Lima et al. (2009), Nishino et al. (2009), Ravasio et al. (2009), Baghaei et al. (2010), Dai et al. (2010), Jiang et al. (2010), Raines et al. (2010), Sandberg et al. (2010), Takahashi et al. (2010a, 2010b), Xu et al. (2011) and references in Yuen (2011a, 2011c, 2011d, 2011e, 2011g).

For electron density of object reported in the literature, iterative algorithms with oversampling can be used to refine the electron density (Sect. 2.2). The iterative algorithms will converge to a unique *ab initio* non-negative electron density with R -index = 0. This is an improvement of the electron density reported in the literature.

For electron density of object which has not been solved, its electron density with R -index = 0 can also be determined by means of iterative algorithms with oversampling (Sect 2.3). The *ab initio* electron density obtained may not be unique.

In all cases, the experimental | Fourier coefficients | and in-between | Fourier coefficients | are improved by means of the consistent condition.

2.1. Phase-retrieval method

The phase of the diffracted wave can be retrieved from the magnitude by imposing some general and reasonable constraints upon the object. For centrosymmetric non-negative object, the constraints used are:

(i) $f(x_1, x_2, x_3)$ has finite size
(ii) $f(x_1, x_2, x_3) \geq 0$
(iii) $f(x_1, x_2, x_3)$ is centrosymmetric
(iv) $f(x_1, x_2, x_3)$ is bounded

The Fourier transform of $f(x_1, x_2, x_3)$,

$$F(\omega_1, \omega_2, \omega_3) = \int_{-L/2}^{L/2} \left\{ \iint f(x_1, x_2, x_3) \exp[-i(\omega_1 x_1 + \omega_2 x_2 + \omega_3 x_3)] dx_2 dx_3 \right\} dx_1$$

where the triple integral is over the volume of the object.

From Yuen (2011a), (C), a unique solution of the phase problem can be obtained by means of the phase-retrieval method.

If the object is continuous or has other symmetry elements, these will be shown in the result.

2.2. Iterative algorithms with oversampling. Refinement of electron density of object reported in the literature

The phase-retrieval method (Sect. 2.1) is time-consuming. However, it provides the theoretical foundation and gives the conditions that iterative algorithms with oversampling will give a unique solution.

The shape and dimensions of the centrosymmetric convex hull (envelope) of the object is determined (Yuen (2011f), Sect. 2.4). Use a unit cell centered at the origin to contain the convex hull of the object. Then mathematically add replica of the unit cell successively to give a crystal. The | Fourier coefficients | are given by the magnitude of the Fourier transform of the object. Yuen (2011b) can then be applied.

The basic unit cell centered at the origin, $f_0(x_1, x_2, x_3)$ is dealt with. The constraints of the object and the basic unit cell $f_0(x_1, x_2, x_3)$ are:

(i) the known shape and dimensions of the centrosymmetric convex hull of the object. If the object is convex, this is a tight support. If the object is concave, this is a loose support.

(ii) $f_0(x_1, x_2, x_3)$ has finite size with size = the known unit cell constants, $a, b, c, \alpha, \beta, \gamma$

(iii) $f_0(x_1, x_2, x_3) \geq 0$

(iv) $f_0(x_1, x_2, x_3)$ is centrosymmetric

(v) $f_0(x_1, x_2, x_3)$ is bounded

(vi) the object is continuous

(vii) the object may have other symmetry elements.

Consider an object the electron density of which has been reported in the literature.

Unlike X-ray crystallography, the Fourier coefficients | and in-between | Fourier coefficients | of the object are measured in the same manner. They have the same experimental uncertainties. As the oversampling ratio = 2, values of in-between | Fourier coefficients | for one index l, m or n is sufficient for determination of the unique electron density. Values of the in-between | Fourier coefficients | for the three indices l, m and n are not independent. They must be consistent so that the electron densities obtained for each index are identical. This consistent condition must be satisfied (Yuen (2011b), Sect. 2.1). All the | Fourier coefficients | and in-between | Fourier coefficients | need to be adjusted within their experimental uncertainties so that the electron densities obtained for each index are identical. We have a very rare result: Sets of experimental results cannot be independent; they needed to be adjusted according to theory. This is also the beauty of the phase-retrieval method. The adjustments can be done by means of iterative algorithms with oversampling.

Use the electron density reported. Calculate the Fourier coefficients and in-between Fourier coefficients from the electron density in the basic unit cell. Use their phases (0 or π) as initial phases. R -index $\neq 0$.

Follow Yuen (2011b), Sect. 2.3. Use

1. the experimentally determined unit cell constants, | Fourier coefficients | , and in-between | Fourier coefficients | ,
2. the initial phases,
3. oversampling ratio $= 2^3 = 8$; use | Fourier coefficients | and in-between | Fourier coefficients | for all (l, m, n),

   and all ($l + \frac{1}{2}$, and/or $m + \frac{1}{2}$, and/or $n + \frac{1}{2}$).
4. the seven constraints,
5. in the final iterations, the | Fourier coefficients | in-between | Fourier coefficients | are relaxed or adjusted, within their uncertainties, to satisfy the consistent condition. Therefore we have **improved the experimental | Fourier coefficients | and in- between | Fourier coefficients |** reported in the literature.

The iterative algorithms will converge to a unique *ab initio* non-negative electron density with R -index = 0.

The electron density of an object reported in the literature should not be very different from the true electron density. When we use the initial phases from this electron density, it is reasonable to expect that the iterations will converge to a unique and improved electron density.

2.3. Iterative algorithms with oversampling. Determination of electron density of object which has not been obtained by other methods

Follow Sect. 2.2. We use random initial phases (0 or π) which are commonly employed. The phase-retrieval method in Yuen (2011b), Sect. 2.1 shows that oversampling ratio =2 leads to a unique electron density. To compromise that proper initial phases are not used, more oversampling is employed. We use | Fourier coefficients | and in-between | Fourier coefficients | for all (l, m, n), and all ($l + \frac{1}{2}$, and/or $m+\frac{1}{2}$, and/or $n+\frac{1}{2}$). The oversampling ratio is $2^3 = 8$. The iterative algorithms may converge to an *ab initio* non-negative electron density with R-index = 0. This convergence can be investigated empirically and theoretically. The experimental | Fourier coefficients | and in-between | Fourier coefficients | are improved by means of the consistent condition. The electron density obtained may not be unique.

3. Discussions and conclusion

The phase-retrieval method in Sect. 2.1 provides the theoretical foundation and gives the conditions that iterative algorithms with oversampling will give a unique solution.

The oversampling ratio is exactly 2. Values of the in-between | Fourier coefficients | for the three indices l, m and n are not independent. They must be consistent so that the electron densities obtained for each index are identical. This consistent condition must be satisfied. All the | Fourier coefficients | and in-between | Fourier coefficients | need to be adjusted within their experimental uncertainties so that the electron densities obtained for each index are identical. We have a very rare result: Sets of experimental results cannot be independent; they needed to be adjusted according to theory. According to the resolution needed, we only use all $\boldsymbol{\omega}$, $|\boldsymbol{\omega}| \leq \omega_{\max}$ (Yuen (2011c), Sect. 2.3). Following Maiden et al. (2011), this redundancy of data can also be used for superresolution. That is, the | Fourier coefficients | and in-between | Fourier coefficients | for all $\boldsymbol{\omega}$, $|\boldsymbol{\omega}| \leq \omega_{\max}$, are regarded as exact. The tail for $\boldsymbol{\omega}$, $|\boldsymbol{\omega}| > \omega_{\max}$ is then fitted in by extrapolation. This is superresolution by extrapolation in Yuen (2011c), Sect. 2.3.

Iterative algorithms with oversampling can be applied for determination of the electron density of a nonperiodic object. The shape and dimensions of the convex hull (envelope) of the object and large oversampling ratio $= 2^3 = 8$ are used to help the iterations to converge to a non-negative electron density.

We urge that researchers will apply iterative algorithm with oversampling (Sect. 2.2) to refine the electron density of the many electron densities of nonperiodic objects reported in the literature, in particular, those structures solved by themselves. Improved electron density with R-index = 0 can be obtained. The *ab initio* electron density obtained is unique. It is determined by the experimental diffraction intensities alone; no other chemical or physical information (for example, atomicity, chemical formula, bond length, bond angle, etc) is used. It can be regarded as a fundamental property of the object. It is just a mathematical transformation of the diffraction intensities to an internal property of the object.

For electron density of nonperiodic object which has not been obtained by other methods, iterative algorithm with oversampling can also be used. Random initial phases (0 or π) may be adopted. To compromise that proper initial phases are not used, more oversampling is employed by using all the in-between | Fourier coefficients | . The oversampling ratio is $2^3 = 8$. The iterative algorithms may converge to an *ab initio* non-negative electron density with R-index = 0. This convergence can be investigated empirically and theoretically. The electron density obtained may not be unique. From the electron density, the atoms, their species and co-ordinates can be determined. The chemical formula can then be obtained. This serves as an experimental method of obtaining the atomic species and the chemical formula.

In all cases, the experimental | Fourier coefficients | and in-between | Fourier coefficients | are improved by means of the consistent condition.

According to Yuen (2011a), the solution of the phase problem is unique for centrosymmetic, real, bounded and finite-size object; non-negativity is not needed. From the non-negative electron density obtained, if we stop using the constraint that the electron density is non-negative, and use the iterations and if we get the same result, this shows that the electron density is indeed non-negative.

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5

Ab initio determination of electron density of gas-phase molecule from the magnitude in electron diffraction. Iterative algorithms with oversampling

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Abstract

The phase-retrieval method provides the theoretical foundation and gives the conditions that iterative algorithms with oversampling will give a unique solution. For electron density of a gaseous molecule reported in the literature, iterative algorithms with oversampling can be used to refine the electron density. The iterative algorithms will converge to an *ab initio* unique non-negative electron density with R -index = 0. This is an improvement of the electron density reported in the literature. For electron density of a gaseous molecule which has not been solved, its electron density with R -index = 0 can also be determined by means of iterative algorithms with oversampling. The *ab initio* electron density obtained may not be unique.

Keywords: Phase retrieval; unique solution; iterative algorithms; oversampling; refinement of electron density.

1. Introduction

This is a sequel to (Yuen 2011a, 2011b).

Much work has been done by many researchers in the determination of the electron density of gas-phase molecules from the magnitude in electron diffraction. For references, see Yuen (2011b). Reckenthaeler et al. (2009) presents electron diffraction from aligned molecules. High-energy X-ray diffraction is presented in Idziak et al. (2007).

A common method is to fit models of atoms to the experimental diffraction intensity. Chemical formula of the molecule, bond lengths and bond angles may be used. Sect.

2.1 presents a phase-retrieval method. Unique phase of the diffracted wave can be retrieved from the magnitude of the diffracted wave (Yuen (2011b), Sect. 2).

Much work has been done by many researchers with iterative algorithms and oversampling, and has gained much empirical success. The phase-retrieval method provides the theoretical foundation and gives the conditions that iterative algorithms with oversampling will give a unique solution.

For references in iterative algorithms and oversampling, see Yuen (2011a, 2011d, 2011e, 2011f, 2011g, 2011i).

For electron density of a gaseous molecule reported in the literature, iterative algorithms with oversampling can be used to refine the electron density (Sect. 2.2). The iterative algorithms will converge to an *ab initio* unique non-negative electron density with R-index = 0. This is an improvement of the electron density reported in the literature.

For electron density of a gaseous molecule which has not been solved, its electron density with R-index = 0 can also be determined by means of iterative algorithms with oversampling (Sect 2.3). The *ab initio* electron density obtained may not be unique.

2.1. Phase-retrieval method.

For a spherically symmetric real and bounded object of radius R,

$$F(\omega_1,0,0)=\int_{-R}^{R}\left\{\iint f(x_1,x_2,x_3)dx_2dx_3\right\}\exp(-i\omega_1x_1)dx_1 \tag{1}$$

where the double integral inside the curly brackets is over a circular section of the sphere of radius R.

From (2011b), Sect. 2, unique value of the phase of $F(\omega_1,0,0)$ and the value of R can be obtained from the magnitude of $F(\omega_1,0,0)$ for $-\infty<\omega_1<\infty$, by means of the phase-retrieval method.

2.2. Iterative algorithms with oversampling. Refinement of electron density of a

gaseous molecule reported in the literature (R -index $\neq 0$)

An alternative method is to use iterative algorithms with oversampling for which the phase-retrieval method has provided the theoretical basis. From Yuen (2011h), (F), the radius of the sphere R is determined. Use a basic unit cell centered at the origin to contain the sphere. Then mathematically add replica of the unit cell successively to give a crystal. For an electron density of a gaseous molecule reported in the literature, calculate the Fourier coefficients and in-between Fourier coefficients from the electron density in the basic unit cell. Use their phases as initial phases. Yuen (2011c), Sect. 2.3 can then be applied.

The constraints of the object and the basic unit cell are:

(i) the known radius R of the sphere. This is a tight support.

(ii) $f_0(x_1, x_2, x_3)$ has finite size with size = the known unit cell constants, $a, b, c, \alpha, \beta, \gamma$

(iii) $f_0(x_1, x_2, x_3) \geq 0$

(iv) $f_0(x_1, x_2, x_3)$ is centrosymmetric

(v) $f_0(x_1, x_2, x_3)$ is bounded

(vi) the object is continuous

According to phase-retrieval theory, values of the | Fourier coefficients | and in-between | Fourier coefficients | for the three indices l, m and n are not independent. They must be consistent so that the electron densities obtained for each index are identical (Yuen (2011c), Sect. 2.1). This consistent condition is satisfied because of the spherical symmetry of the electron density.

Use

1. the known unit cell constants, the experimentally determined | Fourier coefficients | , and in-between | Fourier coefficients | ,
2. the initial phases,
3. oversampling ratio $= 2^3 = 8$; use | Fourier coefficients | and in-between | Fourier coefficients | for all (l, m, n),

and all ($l + \frac{1}{2}$, and/or $m + \frac{1}{2}$, and/or $n + \frac{1}{2}$),

4. the six constraints.

The iterative algorithms will converge to a unique *ab initio* non-negative electron density with R-index = 0.

The electron density of a gaseous molecule reported in the literature is reasonably good. We use the initial phases from this electron density. It is reasonable to expect that the iterations will converge to a unique electron density. This is an improvement of the electron density reported in the literature.

2.3. Iterative algorithms with oversampling. Determination of electron density of a gaseous molecule which has not been solved

Follow Sect. 2.2. We use random initial phases (0 or π) which are commonly employed. The phase-retrieval method in Yuen (2011c), Sect. 2.1 shows that oversampling=2 leads to a unique electron density. To compromise that proper initial phases are not used, we use the known radius R of the sphere and large oversampling ratio. We use | Fourier coefficients | and in-between | Fourier coefficients | for all (l, m, n), and all ($l+\frac{1}{2}$, and/or $m+\frac{1}{2}$, and/or $n+\frac{1}{2}$). The oversampling ratio is $2^3 = 8$. The iterative algorithms may converge to a non-negative electron density with R-index = 0. This convergence can be investigated empirically and theoretically. The *ab initio* electron density obtained may not be unique.

3. Discussions and conclusion

The phase-retrieval method in Sect. 2.1 provides the theoretical foundation and gives the conditions that iterative algorithms with oversampling will give a unique solution.

We urge that researchers in electron diffraction will apply iterative algorithm with oversampling (Sect. 2.2) to refine the electron density of the many gas-phase molecules reported in the literature, in particular, those structures solved by themselves. Improved electron density with R-index = 0 can be obtained. No interpolation has been used. The *ab initio* electron density obtained is unique and exact. It is determined by the experimental diffraction intensities alone; no other chemical or physical information (for example, atomicity, chemical formula, bond length, bond angle, etc) is used. It can be regarded as a fundamental property of the molecule. It is just a mathematical transformation of the diffraction intensities to an internal property of the molecule. Theories of molecular structure and properties (for example, quantum theory) should agree with this internal property. It forms a base for interpretation of experimental data. By fitting to this electron density, atomic coordinates are obtained. The improved atomic coordinates can be compared with those reported in the literature or obtained from spectroscopy. The electron density obtained shows experimentally whether the molecule has sharp boundary or not.

For electron density of a gaseous molecule which has not been obtained by other methods, iterative algorithm with oversampling can also be used. Random initial phases (0 or π) may be adopted. The iterative algorithms may converge to a non-negative electron density with R -index = 0. The *ab initio* electron density obtained may not be unique.

According to Yuen (2011a), the solution of the phase problem is unique for centrosymmetic, real, bounded and finite-size object; non-negativity is not needed. From the non-negative electron density obtained, if we stop using the constraint that the electron density is non-negative, and use the iterations and if we get the same result. This shows that the electron density is indeed non-negative.

For the case of two-dimensional circularly symmetric object, we can use treatment similar to this article. It will be useful in astronomy.

For two-dimensional circularly symmetric object, or three-dimensional spherically symmetric object, it would be helpful if we can develop or use iterative algorithms with oversampling specially designed for such objects.

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6

Ab initio determination of electron densities of projections of a fibre to a plane normal to the fibre axis, a plane containing the fibre axis, an axis normal to the fibre axis, and the fibre axis. Phase-retrieval method and iterative algorithms with oversampling

November 29, 2011

Abstract

This article presents determination of the *ab initio* electron density of projection of a fibre (freely rotating) to a plane normal to the fibre axis, by means of phase-retrieval method. Unique result is obtained. The phase-retrieval method provides the theoretical foundation and serves as a theoretical basis for iterative algorithms with oversampling so that the iterative algorithms very probably converge to a unique non-negative electron density with R-index = 0. Projections to a plane containing the fibre axis, to an axis normal to the fibre axis, and to the fibre axis are also dealt with.

Keywords: Fibre diffraction; projections; two-dimensional object; diffraction planes; circular symmetry; phase retrieval; discrete Hilbert transform; Shannon's sampling; unique solution; exact solution.

1. Introduction

This is a sequel to (Yuen 2011a, 2011b).

For fibre diffraction, see Makowski et al. (1980), Folkhard et al. (1981), Symmons et al. (1995), Yamashita et al. (1995), Paris et al. (2001), and Fujiwara et al. (2009). Reviews are presented in Millane (1988, 1990), and Davidson (1996).

This article presents determination of the *ab initio* electron density of projection of a fibre (freely rotating) to a plane normal to the fibre axis, by means of phase-retrieval method (Sect. 2.1). The phase is not lost. It is just hidden in the magnitude of the diffracted wave. It can be retrieved from the magnitude. The solution is unique.

The phase-retrieval method in Sect. 2.1 is time-consuming. However, it provides the theoretical foundation and serves as a theoretical basis for iterative algorithms with oversampling so that the iterative algorithms very probably converge to a unique non-negative electron density with R -index = 0 (Sect. 2.2).

Determination of *ab initio* electron density of projection to a plane containing the fibre axis, by means of phase-retrieval method, is presented in Sect. 2.3. Iterative algorithms with oversampling are presented in Sect. 2.4.

Determination of *ab initio* electron density of projection to an axis normal to the fibre axis, by means of phase-retrieval method, is presented in Sect. 2.5. Iterative algorithms with oversampling are presented in Sect. 2.6.

Determination of *ab initio* electron density of projection to the fibre axis, by means of phase-retrieval method, is presented in Sect. 2.7. Iterative algorithms with oversampling are presented in Sect. 2.8.

2.1. Projection to a plane normal to the fibre axis. Phase-retrieval method

This is Yuen (2011a), (E). Fibre (freely rotating) has circular symmetry in the x_1x_2 plane with radius $R(x_3)$, and is periodic in x_3 with period c. Let $f(x_1, x_2, x_3)$ be the electron density distribution of the fibre. The diffracted wave is given by the Fourier transform of $f(x_1, x_2, x_3)$. Because of the one-dimensional periodic structure, the diffracted intensity is greatly enhanced to form strong diffraction planes which can be measured accurately.

Consider one period of the fibre.

$$\iiint f_0(x_1, x_2, x_3)\exp[-i(\omega_1 x_1 + \omega_2 x_2 + \omega_3 x_3)]dx_1 dx_2 dx_3 \tag{1}$$

The triple integral is over the volume of one period of the membrane.

$f_0(x_1, x_2, x_3)$ satisfies:

(i) finite size. $f_0(x_1, x_2, x_3) = 0$ for $x_3 < -c/2$ or $x_3 > c/2$. For each x_3, the object $f_0(x_1, x_2, x_3)$ has finite radius $R(x_3)$ in the x_1x_2 plane. Value of c is known.

(ii) $f_0(x_1, x_2, x_3)$ is non-negative

(iii) For each x_3, the object has circular symmetry in the x_1x_2 plane.

(iv) $f_0(x_1, x_2, x_3)$ is bounded

For $\omega_3 = 0$, (1) becomes

$$\int\int\left\{\int_{-c/2}^{c/2} f_0(x_1, x_2, x_3)dx_3\right\}\exp[-i(\omega_1 x_1 + \omega_2 x_2)]dx_1 dx_2 \tag{2}$$

Denote the term in curly brackets by $f_p(x_1, x_2)$. It is the projection of the electron density of one period of the fibre to a plane normal to the fibre axis. It is a two-dimensional object and has circular symmetry in the x_1x_2 plane. Denote (2) by

$$F_p(\omega_1, \omega_2) = \int\int f_p(x_1, x_2)\exp[-i(\omega_1 x_1 + \omega_2 x_2)]dx_1 dx_2$$

$|F_p(\omega_1, \omega_2)|$ has circular symmetry, and is obtained experimentally. The problem is to determine $f_p(x_1, x_2)$. Phase-retrieval method in Yuen (2011a), (C) is used.

Let $g(r) = f_p(x_1, x_2)$,

where $r^2 = x_1{}^2 + x_2{}^2$,

and $U(\omega) = F_p(\omega_1, \omega_2)$

where $\omega^2 = \omega_1{}^2 + \omega_2{}^2$.

Constraints used:

(i) a circle of finite size. $g(r) = 0$, for $r > R_{\max}$, where $R_{\max}$ is the maximum radius of the fibre (value will be determined).

(ii) $g(r)$ is non-negative

(iii) $g(r)$ has circular symmetry

(iv) $g(r)$ is bounded

Use Yuen (2011a), (C), $U(\omega)$ is determined.

The structure of the circle, $g(r) = \frac{1}{2\pi}\int_0^\infty U(\omega)J_0(r\omega)\omega d\omega$

where $J_0(r\omega)$ is the 0th order Bessel function of the first kind. Value of $R_{\max}$ is determined. The result is unique and exact.

2.2. Projection to a plane normal to the fibre axis. Iterative algorithms with oversampling

The phase-retrieval method in Sect. 2.1 is time-consuming. However, it provides the theoretical foundation and serves as a theoretical basis for iterative algorithms with oversampling. From Yuen (2011c), (H), the maximum radius of the fibre $R_{\max}$ is determined. Use a square basic unit cell centered at the origin to contain the circle. Then mathematically add replica of the unit cell successively to give a two-dimensional crystal. Yuen (2011b), Sect. 2.3 or 2.4 can then be applied.

If we use

1. known value of the maximum radius $R_{\max}$ of the circle. This is a tight support.
2. the known unit cell constants, the experimentally determined | Fourier coefficients| for $(l, m, 0)$, and in-between | Fourier coefficients| for ($l + \frac{1}{2}$, and/or $m + \frac{1}{2}$, 0),
3. For an electron density of projection of a fibre to a plane normal to the fibre axis, reported in the literature, calculate the Fourier coefficients and in-between Fourier coefficients from the electron density in the basic unit cell. Use their phases as initial phases.

   Otherwise, use random initial phases (0 or π),

4. oversampling ratio $= 2^2 = 4$; use | Fourier coefficients | for all $(l, m, 0)$,

   and in-between | Fourier coefficients | for all ($l + \frac{1}{2}$, and/or $m + \frac{1}{2}$, 0).

5. constraints (i) to (iv) in Sect. 2.1, plus

   (v) $g(r)$ is continuous,

the iterative algorithms very probably converge to a non-negative electron density with R -index = 0. The electron density is very probably unique.

According to phase-retrieval theory, values of the | Fourier coefficients | and in-between | Fourier coefficients | for the two indices l and m ($n = 0$) are not independent. They must be consistent so that the electron densities obtained for each index are identical (Yuen (2011b), Sect. 2.1). This consistent condition is satisfied because of the circular symmetry of the electron density.

2.3. Projection to a plane containing the fibre axis. Phase-retrieval method

Consider the projection of the fibre to the $x_2 x_3$ plane, x_3 the fibre axis. Similar to Sect. 2.1. For $\omega_1 = 0$, (1) becomes

$$\int\int_{-c/2}^{c/2} \left\{ \int f_0(x_1, x_2, x_3) dx_1 \right\} \exp[-i(\omega_2 x_2 + \omega_3 x_3)] dx_2 dx_3$$

Denote the term in curly brackets by $f_h(x_2, x_3)$. It is the projection of the electron density of one period of the fibre to a plane containing the fibre axis. Assume that it is centrosymmetric. $f_h(x_2, x_3)$ satisfies constraints:

(i) finite size. $f_h(x_2, x_3) = 0$ for $x_3 < -c/2$ or $x_3 > c/2$, and bounded by the fibre surface

(ii) $f_h(x_2, x_3)$ is non-negative

(iii) $f_h(x_2, x_3)$ is centrosymmetric

(iv) $f_h(x_2, x_3)$ is bounded

| Fourier coefficients | for all $(0, m, n)$, and in-between | Fourier coefficients | for

all $(0,\ m$ or $m+\frac{1}{2},\ n)$ are determined experimentally.

Phase-retrieval method in Yuen (2011a), (D) can be applied. The result is unique and exact.

2.4. Projection to a plane containing the fibre axis. Iterative algorithms with oversampling

Sect. 2.3 provides the theoretical foundation and serves as a theoretical basis for iterative algorithms with oversampling. From Yuen (2011c), (H), the maximum radius of the fibre $R_{\max}$ is determined. Use a basic unit cell centered at the origin to contain the object $f_h(x_2,x_3)$. One of the unit cell edges is c. Then mathematically add replica of the unit cell successively to give a two-dimensional crystal. Yuen (2011b), Sect. 2.3 or 2.4 can then be applied.

If we use

1. known value of the maximum radius $R_{\max}$ of the fibre. This is a loose support.
2. the known unit cell constants, the experimentally determined | Fourier coefficients | for all $(0,m,n)$, and in-between | Fourier coefficients | for all $(0,\ m$ or $m+\frac{1}{2},\ n)$,

   in-between | Fourier coefficients | for all $(0,\ m$ or $m+\frac{1}{2},\ n+\frac{1}{2})$ obtained by extrapolation in multidimensions,
3. For the electron density of projection of a fibre to a plane containing the fibre axis, reported in the literature, calculate the Fourier coefficients and in-between Fourier coefficients from the electron density in the basic unit cell. Use their phases as initial phases.

   Otherwise, use random initial phases (0 or π),
4. oversampling ratio $=2^2=4$; use | Fourier coefficients | for all $(0,m,n)$, and in-between | Fourier coefficients | for all $(0,\ m+\frac{1}{2}$, and/or $n+\frac{1}{2})$.

5. constraints (i) to (iv) in Sect. 2.3, plus
   (v) $f_h(x_2, x_3)$ is continuous,
6. in the final iterations, the interpolated in-between | Fourier coefficients | are relaxed or adjusted, within their uncertainties, to satisfy the consistent condition (Yuen (2011b), Sect. 2.1); the interpolated in-between | Fourier coefficients | are improved.

the iterative algorithms very probably converge to a non-negative electron density with R -index = 0. The *ab initio* electron density is very probably unique for this set of | Fourier coefficients | and initial in-between | Fourier coefficients | .

2.5. Projection to an axis normal to the fibre axis. Phase-retrieval method

For $\omega_2 = \omega_3 = 0$, (1) becomes

$$\int\left\{\int\int_{-c/2}^{c/2} f_0(x_1, x_2, x_3)dx_2dx_3\right\}\exp[-i\omega_1 x_1]dx_1$$

The integrals for x_1 and x_2 are from $-R_{\max}$ to $R_{\max}$.

Denote the term in curly brackets by $f_n(x_1)$. It is the projection of the electron density of one period of the fibre to an axis normal to the fibre axis. It is a one-dimensional centrosymmetric object.

Apply Yuen (2011a), (A) for the one-dimensional centrosymmetric nonperiodic object $f_n(x_1)$. $F_n(\omega_1)$ is the Fourier transform of $f_n(x_1)$. We have $|F_n(\omega_1)|$ which is continuous. The sign of $F_n(\omega_1)$ changes at a zero of odd order and remains unchanged at a zero of even order. To obtain the structure $f_n(x_1)$, we need only locate the zeros of $|F_n(\omega_1)|$, and finding out whether they are of even order or odd. (The values of the orders are not needed). Choose a sign of $F_n(0)$, positive for a non-negative membrane. The sign of $F_n(\omega_1)$ is determined. The structure of the object, including the length of the object (2 $R_{\max}$ of the fibre), is then determined.

In practice, $|F_n(\omega_1)|$ for discrete values of ω_1 are measured. We have to extrapolate to the zeros. This is not very accurate. May be it is not a zero at all. This may ruin the result. Follow Yuen (2011c), (H) to determine $R_{\max}$ of the fibre. Use a period $\geq 2R_{\max}$, to contain the object $f_n(x_1)$. Then add its replica successively to give a periodic object and use Yuen (2011a), (B) for one-dimensional centrosymmetric periodic object. Constraints used:

(i) size of $f_n(x_1)$ is from $-R_{\max}$ to $R_{\max}$. Value of $R_{\max}$ will be determined
(ii) $f_n(x_1)$ is non-negative
(iii) $f_n(x_1)$ is centrosymmetric,
(iv) $f_n(x_1)$ is bounded

Discrete Hilbert transform and Shannon's sampling (Yuen (2011a), (7) and (8)) can then be used. $|F_n(\omega_1)|$ is obtained. Use this continuous $|F_n(\omega_1)|$ and follow the procedure in above. $f_n(x_1)$, including value of $R_{\max}$ of the fibre, is determined. The result is unique and exact.

2.6. Projection to an axis normal to the fibre axis. Iterative algorithms with oversampling

Sect. 2.5 provides the theoretical foundation and serves as a theoretical basis for iterative algorithms with oversampling. From Yuen (2011c), (H), the maximum radius of the fibre $R_{\max}$ is determined. Use a basic unit cell centered at the origin to contain $f_n(x_1)$. Then mathematically add replica of the unit cell successively to give a one-dimensional crystal. Yuen (2011b), Sect. 2.3 or 2.4 can then be applied.

If we use

1. known value of the maximum radius $R_{\max}$. This is a tight support.
2. the known unit cell constants, the experimentally determined | Fourier coefficients | for $(l,0,0)$, and in-between | Fourier coefficients | for

$(l+\frac{1}{2},0,\ 0)$,

3. For an electron density of projection of a fibre to an axis normal to the fibre axis, reported in the literature, calculate the Fourier coefficients and in-between Fourier coefficients from the electron density in the basic unit cell. Use their phases as initial phases.

   Otherwise, use random initial phases (0 or π),

4. oversampling ratio =2; use | Fourier coefficients | for all $(l,0,0)$, and in-between | Fourier coefficients | for all $(l+\frac{1}{2},0,0)$.

6. constraints (i) to (iv) in Sect. 2.5, plus

   (v) $f_n(x_1)$ is continuous,

the iterative algorithms may or may not converge to a unique non-negative electron density with R -index = 0.

2.7. Projection to the fibre axis. Phase-retrieval method

For $\omega_1 = \omega_2 = 0$, (1) becomes

$$\int_{-c/2}^{c/2}\left\{\iint f_0(x_1,x_2,x_3)dx_1dx_2\right\}\exp[-i\omega_3x_3]dx_3$$

The integrals for x_1 and x_2 are from $-R_{\max}$ to $R_{\max}$.

Denote the term in curly brackets by $f_c(x_3)$. It is the projection of the electron density of one period of the fibre to the fibre axis. It is an one-dimensional object. Assume that it is centrosymmetric. For the whole fibre, this is an one-dimensional periodic object. The | Fourier coefficients | for $(0,0,n)$ are obtained experimentally. The in-between | Fourier coefficients | for $(0,0,n+\frac{1}{2})$ are obtained by interpolation in multidimensions. Phase-retrieval method in Yuen (2011a), (B) is used with constraints:

(i) size of $f_c(x_3)$ is from $-c/2$ to $c/2$. Value of c is known.

(ii) $f_c(x_3)$ is non-negative

(iii) $f_c(x_3)$ is centrosymmetric,

(iv) $f_c(x_3)$ is bounded

The unique $f_c(x_3)$ is determined, for this set of | Fourier coefficients | and in-between | Fourier coefficients | .

2.8. Projection to the fibre axis. Iterative algorithms with oversampling

Sect. 2.7 provides the theoretical foundation and serves as a theoretical basis for iterative algorithms with oversampling. Yuen (2011b), Sect. 2.3 or 2.4 can be applied.

If we use

1. the known unit cell constant c, the experimentally determined | Fourier coefficients | for $(0,0,n)$, and the in-between | Fourier coefficients | for $(0,0,n+\frac{1}{2})$ obtained by interpolation in multidimensions,
2. For an electron density of projection of a fibre to an axis normal to the fibre axis, reported in the literature, calculate the Fourier coefficients and in-between Fourier coefficients from the electron density in the basic unit cell. Use their phases as initial phases.
   Otherwise, use random initial phases (0 or π),
3. oversampling ratio =2; use | Fourier coefficients | for all $(0,0,n)$, and in-between | Fourier coefficients | for all $(0,0,n+\frac{1}{2})$.
4. constraints (i) to (iv) in Sect. 2.7, plus
   (v) $f_c(x_3)$ is continuous,
5. in the final iterations, the interpolated in-between | Fourier coefficients | are relaxed or adjusted, within their uncertainties, to satisfy the consistent condition (Yuen (2011b), Sect. 2.1); the interpolated in-between | Fourier coefficients | are improved,

the iterative algorithms may or may not converge to a unique non-negative electron density with R-index = 0, for this set of｜Fourier coefficients｜ and in-between｜Fourier coefficients｜.

3. Discussions and conclusion

For determination of *ab initio* electron density of projection to a plane normal to the fibre axis, the phase-retrieval method in Sect. 2.1 provides the theoretical foundation and gives the conditions that iterative algorithms with oversampling will very probably give a unique solution (Sect. 2.2).

Because of the one-dimensional periodic structure, the diffracted intensity is greatly enhanced to form strong diffraction planes and can be measured accurately. No interpolation is needed. The *ab initio* electron density obtained is unique and exact. It is determined by the experimental diffraction intensities alone; no other chemical or physical information (for example, atomicity, chemical formula, bond length, bond angle, etc) is used.

According to Yuen (2011a), the solution of the phase problem is unique for centrosymmetic, real, bounded and finite-size object; non-negativity is not needed. From the non-negative electron density obtained, we stop using the constraint that the electron density is non-negative, and use the iterations. If we get the same result, this shows that the electron density is indeed non-negative.

It would be helpful if we can develop or use iterative algorithms with oversampling specially designed for two-dimensional circularly symmetric object.

Determination of *ab initio* electron density of projection to a plane containing the fibre axis, by means of phase-retrieval method, is presented in Sect. 2.3. Unique solution is determined. Iterative algorithms with oversampling are presented in Sect. 2.4. The result is very probably unique.

Determination of *ab initio* electron density of projection to an axis normal to the fibre axis, by means of phase-retrieval method, is presented in Sect. 2.5. Unique solution with value of the maximum radius of the fibre is determined. Iterative algorithms with oversampling are presented in Sect. 2.6. They may or may not converge to a unique non-negative electron density.

Determination of *ab initio* electron density of projection to the fibre axis, by means of phase-retrieval method, is presented in Sect. 2.7. Unique solution is obtained. Iterative algorithms with oversampling are presented in Sect. 2.8. They may or may not converge to a unique non-negative electron density.

For the general case of non-centrosymmetric or complex object, the solution of the one-dimensional phase-retrieval problem may not be unique (Yuen (2011a), Sect. 1, Sect. 2.1.1, (B)). Even though it is unique for a centrosymmetric real object, the iterative algorithms may or may not converge to a unique non-negative electron density (Sect. 2.6, 2.8). For centrosymmetric real or the general case of non-centrosymmetric or complex object, to help the iterative algorithms to converge to a unique non-negative electron density, very strong container constraint and solvent constraint are used, as in microcavity arrays (Yuen (2011d)).

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7

Ab initio determination of electron densities of centrosymmetric non-negative fibres from diffraction intensities. Iterative algorithms with oversampling

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Abstract

For centrosymmetric non-negative freely rotating or fixed fibre, the phase is not lost. It is just hidden in the magnitudes of the diffracted wave. It can be retrieved from the magnitude. Unique result is obtained. The phase-retrieval method provides the theoretical foundation and serves as a theoretical basis for iterative algorithms with oversampling so that the iterative algorithms very probably converge to a unique non-negative electron density with R -index = 0.

Keywords: Fiber diffraction; one-dimensional periodic object; phase retrieval; unique solution.

1. Introduction

This is a sequel to (Yuen 2011a, 2011b).

For fiber diffraction, see Hulmes et al. (1977), Hulmes et al. (1980), Millane (1990, 1992), Millane & Stroud (1995), Stroud & Millane (1995a, 1995b, 1996), and Premilat & Albiser (1999) . Maximum entropy method is used in calculating the electron density in Welsh et al. (1998), Marvin et al. (2006) and Straus et al. (2008). Direct method in fibre diffraction is presented in Dorset (1997), Dorset & McCourt (1997) and Dorset (2003). Morris & Serpell (2010) presents a review.

For centrosymmetric non-negative freely rotating or fixed fibre, it is found that the phase is not lost. It is just hidden in the magnitudes of the diffracted wave (Yuen (2011a), (E), (F)). It can be retrieved from the magnitude (Sect. 2.1, 3.1). Unique result is obtained. Hence the electron density can be obtained directly from the diffraction intensities.

The phase-retrieval method in Sect. 2.1, 3.1 is time-consuming. However, it provides the theoretical foundation and serves as a theoretical basis for iterative algorithms with oversampling so that the iterative algorithms very probably converge to a unique non-negative electron density with R -index = 0 (Sect. 2.2, 3.2).

2.1. Fibre (freely rotating). Phase-retrieval method.

This is Yuen (2011a), (E). Fibre has circular symmetry in the x_1x_2 plane with radius $R(x_3)$, and is periodic in x_3 with period c. Consider one period of the fibre. Constraints used:

(i) finite size. $f_0(x_1, x_2, x_3) = 0$ for $x_3 < -c/2$ or $x_3 > c/2$. For each x_3, the object $f_0(x_1, x_2, x_3)$ has finite radius $R(x_3)$ (value will be determined) in the x_1x_2 plane. Value of c is known.

(ii) $f_0(x_1, x_2, x_3)$ is non-negative

(iii) For each x_3, the object has circular symmetry in the x_1x_2 plane.

$f_0(x_1, x_2, x_3)$ is centrosymmetric

(iv) $f_0(x_1, x_2, x_3)$ is bounded

Because of the one-dimensional periodic structure, the diffracted intensity is greatly enhanced to form strong diffraction planes which can be measured accurately. Let $\omega_0 = 2\pi/c$. For all l, $l = 0,\pm1,\pm2.,...$, $|F_0(\omega_1, \omega_2, l\omega_0)|$ are measured experimentally. It has circular symmetry in the $\omega_1\omega_2$ plane.

$$F_0(\omega_1, \omega_2, l\omega_0)$$

$$= \iint \left\{ \int_{-c/2}^{c/2} f_0(x_1, x_2, x_3) \exp(-il\omega_0 x_3) dx_3 \right\} \exp[-i(\omega_1 x_1 + \omega_2 x_2)] dx_1 dx_2 \qquad (1)$$

The triple integral is over one period of the freely rotating fibre. Denote the term in curly brackets by $q(x_1, x_2, l\omega_0)$. $q(x_1, x_2, l\omega_0)$ is circularly symmetric in the x_1x_2 plane with radius = the largest $R(x_3)$. From Yuen (2011a), (C) and (E), get

$q(x_1, x_2, l\omega_0)$.

$$f(x_1, x_2, x_3) = \frac{1}{c}\sum_{l=-\infty}^{\infty} q(x_1, x_2, l\omega_0)\exp\left(il\omega_0 x_3\right) \tag{2}$$

The unique structure $f(x_1, x_2, x_3)$ including $R(x_3)$ is determined.
If (v) the fibre $f(x_1, x_2, x_3)$ is continuous with

$$f_0(x_1, x_2, -c/2) = f_0(x_1, x_2, c/2) \text{ for all } x_1,\ x_2,$$

this will be shown in the structure $f(x_1, x_2, x_3)$ obtained.

2.2. Fibre (freely rotating). Iterative algorithms with oversampling

The phase-retrieval method in Sect. 2.1 is time-consuming. However, it provides the theoretical foundation and serves as a theoretical basis for iterative algorithms with oversampling. From Yuen (2011c), (H), the maximum radius of the fibre $R_{\max}$ is determined. Use an orthogonal unit cell centered at the origin to contain a period of the fibre. Then mathematically add replica of the unit cells successively to give a three-dimensional crystal consisting of parallel freely rotating fibres. The | Fourier coefficients | are given by the magnitude of the Fourier transform of the content of the basic unit cell of the crystal. The in-between | Fourier coefficients | for ($l+\frac{1}{2}$, and/or $m+\frac{1}{2}$, n) are obtained from the experimental diffraction intensity; no interpolation is needed. The in-between | Fourier coefficients | for ($l, m, n+\frac{1}{2}$) are obtained by interpolation in multidimensions. Yuen (2011b), Sect. 2.4 can then be applied.

If we use

1. known value of the maximum radius $R_{\max}$ of the fibre. This is a loose support.
2. the known unit cell constants, the experimentally determined | Fourier coefficients | for (l, m, n), and in-between | Fourier coefficients | for ($l+\frac{1}{2}$,

and/or $m+\frac{1}{2}$, n),

3. random initial phases (0 or π),
4. oversampling ratio $= 2^3 = 8$; use | Fourier coefficients | and in-between | Fourier coefficients | for all (l, m, n),

   and all ($l+\frac{1}{2}$, and/or $m+\frac{1}{2}$, and/or $n+\frac{1}{2}$).

   Some of them are experimental data (see 2.), the rest are obtained by interpolation in multidimensions.
5. constraints (i) to (iv). (v) is also used as a constraint.
6. in the final iterations, the interpolated in-between | Fourier coefficients | are relaxed or adjusted, within their uncertainties, to satisfy the consistent condition (Yuen (2011b), Sect. 2.1); the interpolated in-between | Fourier coefficients | are improved,

the iterative algorithms very probably converge to a non-negative electron density with R -index = 0. The electron density is very probably unique for this set of | Fourier coefficients | and in-between | Fourier coefficients | .

To compromise that proper initial phases are not used, we use known value of the maximum radius $R_{\max}$ of the fibre, and larger oversampling ratio $= 2^3 = 8$; use | Fourier coefficients | and in-between | Fourier coefficients | for all (l, m, n), and all ($l+\frac{1}{2}$, and/or $m+\frac{1}{2}$, and/or $n+\frac{1}{2}$). These will help the iterative algorithms to converge to a unique non-negative electron density with R -index = 0. This convergence can be investigated empirically and theoretically.

As shown by the phase-retrieval method in Sect. 2.1, the interpolated in-between | Fourier coefficients | are, in fact, not needed. After the iterations converge to a non-negative electron density, we stop using all the interpolated in-between | Fourier coefficients | . The iterative algorithms will converge to a **unique** non-negative electron density.

3.1. Fixed fibre (a one-dimensional periodic object). Phase-retrieval method.

This is Yuen (2011a), (F). The fibre is periodic in x_3 with period c. Consider one period of the fibre.

Constraints used:

(i) finite size. $f_0(x_1, x_2, x_3) = 0$ for $x_3 < -c/2$ or $x_3 > c/2$. Value of c is known. Shape and dimensions of the fibre will be determined.

(ii) $f_0(x_1, x_2, x_3)$ is non-negative

(iii) $f_0(x_1, x_2, x_3)$ is centrosymmetric

(iv) $f_0(x_1, x_2, x_3)$ is bounded

Because of the one-dimensional periodic structure, the diffracted intensity is greatly enhanced to form strong diffraction planes which can be measured accurately. Let $\omega_0 = 2\pi / c$. For all l, $l = 0, \pm 1, \pm 2., \ldots$, $|F_0(\omega_1, \omega_2, l\omega_0)|$ are measured experimentally.

$$F_0(\omega_1, \omega_2, l\omega_0)$$

$$= \int\int \left\{ \int_{-c/2}^{c/2} f_0(x_1, x_2, x_3) \exp(-il\omega_0 x_3) dx_3 \right\} \exp[-i(\omega_1 x_1 + \omega_2 x_2)] dx_1 dx_2 \qquad (3)$$

The triple integral is over one period of the fixed fibre. Denote the term in curly brackets by $q(x_1, x_2, l\omega_0)$. From Yuen (2011a), (C) and (F), get $q(x_1, x_2, l\omega_0)$. The unique structure $f(x_1, x_2, x_3)$ is given by (2). The shape and dimensions of one period of the fibre are determined.

If (v) the fibre $f(x_1, x_2, x_3)$ is continuous with

$$f_0(x_1, x_2, -c/2) = f_0(x_1, x_2, c/2) \quad \text{for all } x_1,\ x_2,$$

(vi) the fibre has other symmetry elements,

these will be shown in the structure $f(x_1, x_2, x_3)$ obtained.

3.2. Fixed fibre (a one-dimensional periodic object). Iterative algorithms with

oversampling

The phase-retrieval method in Sect. 3.1 is time-consuming. However, it provides the theoretical foundation and serves as a theoretical basis for iterative algorithms with oversampling. From Yuen (2011d), Sect. 2.2, the maximum lengths of the fibre along all directions normal to the fibre axis are determined. Use an orthogonal unit cell centered at the origin to contain a period of the fibre. Then mathematically add replica of the unit cells successively to give a three-dimensional crystal consisting of parallel fixed fibres.

The | Fourier coefficients | are given by the magnitude of the Fourier transform of the content of the basic unit cell of the crystal. The in-between | Fourier coefficients | for ($l+\frac{1}{2}$, and/or $m+\frac{1}{2}$, n) , are obtained from the experimental diffraction intensity; no interpolation is needed. The in-between | Fourier coefficients | for $(l, m, n+\frac{1}{2})$ are obtained by interpolation in multidimensions. Yuen (2011b), Sect. 2.4 can then be applied.

If we use

1. the known maximum lengths of the fibre along all directions normal to the fibre axis. This is a loose support.
2. the known unit cell constants, the experimentally determined | Fourier coefficients | for (l,m,n), and in-between | Fourier coefficients | for ($l+\frac{1}{2}$, and/or $m+\frac{1}{2}$, n) ,
3. random initial phases (0 or π),
4. oversampling ratio $=2^3=8$; use | Fourier coefficients | and in-between | Fourier coefficients | for all (l,m,n),

and all ($l+\frac{1}{2}$, and/or $m+\frac{1}{2}$, and/or $n+\frac{1}{2}$).

Some of them are experimental data (see 2.), the rest are obtained by interpolation in multidimensions.

5. constraints (i) to (iv) in Sect. 3.1. (v) and (vi) are also used as constraints.
6. in the final iterations, the interpolated in-between | Fourier coefficients | are relaxed or adjusted, within their uncertainties, to satisfy the consistent condition (Yuen (2011b), Sect. 2.1); the interpolated in-between | Fourier coefficients | are improved,

the iterative algorithms very probably converge to a non-negative electron density with R -index = 0. The electron density is very probably unique for this set of | Fourier coefficients | and in-between | Fourier coefficients | .

To compromise that proper initial phases are not used, we use the known maximum lengths of the fibre along all directions normal to the fibre axis, and larger oversampling ratio $= 2^3 = 8$; use | Fourier coefficients | and in-between | Fourier coefficients | for all (l, m, n), and all ($l + \frac{1}{2}$, and/or $m + \frac{1}{2}$, and/or $n + \frac{1}{2}$). These will help the iterative algorithms to converge to a unique non-negative electron density with R -index = 0. This convergence can be investigated empirically and theoretically.

As shown by the phase-retrieval method in Sect. 3.1, the interpolated in-between | Fourier coefficients | are, in fact, not needed. After the iterations converge to a non-negative electron density, we stop using all the interpolated in-between | Fourier coefficients | . The iterative algorithms will converge to a **unique** non-negative electron density.

4. Discussions and conclusion

The phase-retrieval methods in Sect. 2.1 and 3.1 provide the theoretical foundation and give the conditions that iterative algorithms with oversampling will very probably give a unique solution.

Because of the one-dimensional periodic structure, the diffracted intensity is greatly enhanced to form strong diffraction planes. As the oversampling ratio is exactly 2, for a periodic fibre, no interpolation is needed. The intensity in the diffraction planes can be measured experimentally. The electron density obtained is unique and exact. This is an important case of determining unique periodic structures without using interpolation. The *ab initio* electron density is determined by the experimental diffraction intensities alone; no other chemical or physical information (for example, atomicity, chemical formula, bond length, bond angle, etc) is used. It can be regarded as a fundamental property of the fibre. It is just a mathematical transformation of the diffraction intensities to an internal property of the fibre. Theories of molecular structure and properties (for example, quantum theory) should agree with this internal property. It forms a basis for interpretation of experimental data. The electron density obtained shows experimentally whether the fibre has sharp boundary or not.

According to Yuen (2011a), the solution of the phase problem is unique for centrosymmetic, real, bounded and finite-size object; non-negativity is not needed. From the non-negative electron density obtained, we stop using the constraint that the electron density is non-negative, and use the iterations. If we get the same result, this shows that the electron density is indeed non-negative.

For freely rotating fibre, it would be helpful if we can develop or use iterative algorithms with oversampling specially designed for crystals containing a period of the fibre in each unit cell.

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8

Ab initio determination of centrosymmetric one-dimensional electron density profile of a membrane, and two-dimensional projection to the plane of the membrane. Phase-retrieval method and iterative algorithms with oversampling

November 29, 2011

Abstract

This article presents determination of *ab initio* one-dimensional centrosymmetric electron density profile of a membrane by means of phase-retrieval method. The solution is unique and exact. Two-dimensional centrosymmetric electron density of projection of a membrane to the plane of the membrane is also dealt with.

Keywords: Membrane diffraction; diffraction rods; projections; one-dimensional object; phase retrieval; unique solution; exact solution.

1. Introduction

Much work has done in the determination of one-dimensional centrosymmetric electron density profile of membranes. See, for examples, Worthington (1969), Blaurock (1971), Luzzati et al. (1972), McIntosh & Worthington (1974), King (1975), McIntosh et al. (1976), Torbet & Wilkins (1976), Worthington (1981), Worthington (1988), Worcester et al. (1992), Kirchner (1996), Hristova & White (1998), Tristram-Nagle et al. (2002), Zeng & Ungar (2002), Liu & Nagle (2004), Petrache et al. (2004), Kučerka et al. (2005), and Wang et al. (2006).

Reviews are presented in Levine (1972), White (1975), Blaurock (1982), White & Wiener (1996), Nagle & Tristram-Nagle (2000), Tristram-Nagle & Nagle (2004), and Huang (2005).

This article presents determination of *ab initio* one-dimensional centrosymmetric electron density profile of a membrane by means of phase-retrieval method (Sect. 2.1). The solution is unique and exact.

Iterative algorithms with oversampling are presented in Sect. 2.2.

Determination of *ab initio* two-dimensional centrosymmetric electron density of projection of a membrane to the plane of the membrane, by means of phase-retrieval method is presented in Sect. 2.3. The solution is unique for the set of | Fourier coefficients| and in-between | Fourier coefficients| .

Iterative algorithms with oversampling are presented in Sect. 2.4.

2.1. One-dimensional centrosymmetric electron density profile of a membrane. Phase-retrieval method.

Let $f(x_1, x_2, x_3)$ be the electron density distribution of a planar membrane, periodic in x_1 and x_2, with periods a and b. The diffracted wave is given by the Fourier transform of $f(x_1, x_2, x_3)$. Because of the two-dimensional periodic structure, the diffracted intensity is greatly enhanced to form strong diffraction rods which can be measured accurately.

Consider one period of the membrane

$$\iiint f_0(x_1, x_2, x_3)\exp[-i(\omega_1 x_1 + \omega_2 x_2 + \omega_3 x_3)]dx_1 dx_2 dx_3 \qquad (1)$$

The triple integral is over one period of the membrane.

For $\omega_1 = \omega_2 = 0$, (1) becomes

$$\int\left\{\iint f_0(x_1, x_2, x_3)dx_1 dx_2\right\}\exp[-i\omega_3 x_3]dx_3 .$$

Denote the term in curly brackets by $f_n(x_3)$. It is the electron density profile of the membrane, which is the projection of the electron density of one period of the membrane upon an axis normal to the membrane. Assume that it is centrosymmetric. Apply Yuen (2011a), (A) or (B) for the one-dimensional centrosymmetric nonperiodic object with constraints:

(i) size of $f_n(x_3)$ is from –(maximum thickness)/2 to (maximum thickness)/2.

(ii) $f_n(x_3)$ is non-negative

(iii) $f_n(x_3)$ is centrosymmetric,

(iv) $f_n(x_3)$ is bounded

$f_n(x_3)$, including the maximum thickness of the membrane, is determined. The result is unique and exact.

2.2. One-dimensional centrosymmetric electron density profile of a membrane. Iterative algorithms with oversampling

Sect. 2.1 provides the theoretical foundation and serves as a theoretical basis for iterative algorithms with oversampling. Follow Yuen (2011c), (J) to determine the maximum thickness of the membrane. Use a basic unit cell centered at the origin to contain $f_n(x_3)$. Then mathematically add replica of the unit cell successively to give a one-dimensional crystal. Yuen (2011b), Sect. 2.3 or 2.4 can then be applied.

If we use

1. known value of the maximum thickness of the membrane. This is a tight support.
2. the known unit cell constants, the experimentally determined $|$ Fourier coefficients $|$ for $(0,0,n)$, and in-between $|$ Fourier coefficients $|$ for $(0,0,n+\frac{1}{2})$,
3. For an electron density profile of a membrane, reported in the literature, calculate the Fourier coefficients and in-between Fourier coefficients from the electron density in the basic unit cell. Use their phases as initial phases.
   Otherwise, use random initial phases (0 or π),
4. oversampling ratio =2; use $|$ Fourier coefficients $|$ for all $(0,0,n)$, and in-between $|$ Fourier coefficients $|$ for all $(0,0,n+\frac{1}{2})$.
5. constraints (i) to (iv) in Sect. 2.1, plus
   (v) $f_c(x_3)$ is continuous,

the iterative algorithms may or may not converge to a unique non-negative electron density with R-index = 0.

2.3. Projection to the plane of the membrane. Phase-retrieval method.

For $\omega_3 = 0$, (1) becomes

$$\iint \left\{ \int f_0(x_1, x_2, x_3) dx_3 \right\} \exp[-i(\omega_1 x_1 + \omega_2 x_2)] dx_1 dx_2$$

Denote the term in curly brackets by $f_h(x_1, x_2)$. It is the projection of the electron density of one period of the membrane to a plane of the membrane. Assume that it is centrosymmetric. $f_h(x_1, x_2)$ satisfies constraints:

(i) finite size. $f_h(x_1, x_2) = 0$ for $x_1 < -a/2$ or $x_1 > a/2$,
$x_2 < -b/2$ or $x_2 > b/2$,

(ii) $f_h(x_1, x_2)$ is non-negative

(iii) $f_h(x_1, x_2)$ is centrosymmetric

(iv) $f_h(x_1, x_2)$ is bounded

The weak diffraction intensity is collected into strong diffraction rods (Bragg rods) which can be measured accurately. Let $\omega_{10} = 2\pi / a$, $\omega_{20} = 2\pi / b$. For all l and m, the | Fourier coefficients| $|F_0(l\omega_{10}, m\omega_{20}, 0)|$ are measured experimentally. The in-between | Fourier coefficients| $|F_0((l+1/2)\omega_{10}, m\omega_{20}, 0)|$ are obtained by interpolation in multidimensions. Follow Yuen (2011a), (D), $f_h(x_1, x_2)$ is determined. The result is unique for this set of | Fourier coefficients | and in-between | Fourier coefficients| .

2.4. Projection to the plane of the membrane. Iterative algorithms with oversampling

The phase-retrieval method in Sect. 2.4 provides the theoretical foundation and serves as a theoretical basis for iterative algorithms with oversampling. Projection of the membrane to the plane of the membrane is a two-dimensional crystal with $f_h(x_1, x_2)$ a unit cell. Yuen (2011b), Sect. 2.3 or 2.4 can then be applied.

If we use

1. the known unit cell constants with periods a and b, the experimentally determined | Fourier coefficients | for $(l,m,0)$, and in-between | Fourier coefficients | for ($l+\frac{1}{2}$, and/or $m+\frac{1}{2}$, 0) obtained by interpolation in multidimensions,
2. For an electron density of projection of a membrane to the plane of the membrane, reported in the literature, calculate the Fourier coefficients and in-between Fourier coefficients from the electron density in the basic unit cell. Use their phases as initial phases.
   Otherwise, use random initial phases (0 or π),
3. oversampling ratio $=2^2=4$; use | Fourier coefficients | for all $(l,m,0)$, and in-between | Fourier coefficients | for all ($l+\frac{1}{2}$, and/or $m+\frac{1}{2}$, 0).
4. constraints (i) to (iv) in Sect. 2.3, plus
   (v) $f_h(x_1,x_2)$ is continuous,
5. in the final iterations, the interpolated in-between | Fourier coefficients | are relaxed or adjusted, within their uncertainties, to satisfy the consistent condition (Yuen (2011b), Sect. 2.1); the interpolated in-between | Fourier coefficients | are improved,

the iterative algorithms very probably converge to a non-negative electron density with R-index = 0. The electron density is very probably unique for this set of | Fourier coefficients | and in-between | Fourier coefficients | .

3. Discussions and conclusion

By applying phase-retrieval method to the magnitude of the Fourier transform of the one-dimensional centrosymmetric electron density profile of a membrane, an *ab initio* electron density profile of the membrane, including its maximum thickness, is obtained (Sect. 2.1). This maximum thickness should agree with that determined in Yuen (2011c), (J).

Because of the two-dimensional periodic structure, the diffracted intensity is greatly enhanced to form strong diffraction rods which can be measured accurately. No interpolation is needed. The *ab initio* electron density profile obtained is unique and exact. It is determined by the experimental diffraction intensities alone; no other chemical or physical information (for example, atomicity, chemical formula, bond length, bond angle, etc) is used. It can be regarded as a fundamental property of the membrane. It is just a mathematical transformation of the diffraction intensities to an internal property of the membrane. Theories of molecular structure and properties (for example, quantum theory) should agree with this internal property. It forms a basis for interpretation of experimental data.

Iterative algorithms with oversampling are presented in Sect. 2.2. They may or may not converge to a unique non-negative electron density.

For the general case of non-centrosymmetric or complex object, the solution of the one-dimensional phase-retrieval problem may not be unique (Yuen (2011a), Sect. 1, Sect. 2.1.1, (B)). Even though it is unique for a centrosymmetric real object, the iterative algorithms may or may not converge to a unique non-negative electron density (Sect. 2.2). For centrosymmetric real or the general case of non-centrosymmetric or complex object, to help the iterative algorithms to converge to a unique non-negative electron density, very strong container constraint and solvent constraint are used, as in microcavity arrays (Yuen (2011d)).

Determination of *ab initio* two-dimensional centrosymmetric electron density of projection of a membrane to the plane of the membrane, by means of phase-retrieval method is presented in Sect. 2.3. The solution is unique for the set of | Fourier coefficients | and in-between | Fourier coefficients | . Iterative algorithms with oversampling are presented in Sect. 2.4. The iterative algorithms very probably converge to a non-negative electron density with R -index = 0. The electron density is very probably unique.

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9

Ab initio determination of electron densities of centrosymmetric non-negative membrane from diffraction intensities. Iterative algorithms with oversampling

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Abstract

For centrosymmetric non-negative planar periodic membrane, the phase is not lost. It is just hidden in the magnitudes of the diffracted wave. It can be retrieved from the magnitude. Unique result is obtained. The phase-retrieval method provides the theoretical foundation and serves as a theoretical basis for iterative algorithms with oversampling so that the iterative algorithms very probably converge to a unique non-negative electron density with R -index = 0.

Keywords: Membrane diffraction; two-dimensional periodic object; diffraction rods; Bragg rods; phase retrieval; unique solution.

1. Introduction

This is a sequel to (Yuen 2011a, 2011b).

For structures of two-dimensional crystals, see Caspar et al. (1977), Makowski et al. (1977), Zampighi et al. (1984), Dorset et al. (1990), Dorset (1995, 1997), Harper & Gruner (2000), Harper et al. (2000), Wu & Spence (2002). A review is presented in Rappolt (2007).

Spence et al. (2003) uses one-dimensional compact support to investigate the three-dimensional diffractive image for crystalline monolayer. Iterative algorithm with oversampling is employed. Gipson et al. (2011) presents recovery of missing amplitudes and phases in tilt-limited electron crystallography of two-dimensional crystals.

For centrosymmetric non-negative planar periodic membrane, it is found that the phase is not lost. It is just hidden in the magnitudes of the diffracted wave (Yuen (2011a), (G)). It can be retrieved from the magnitude (Sect. 2.1). Unique result is obtained. Hence the electron density can be obtained directly from the diffraction intensities.

The phase-retrieval method in Sect. 2.1 is time-consuming. However, it provides the theoretical foundation and serves as a theoretical basis for iterative algorithms with oversampling so that the iterative algorithms very probably converge to a unique non-negative electron density with R -index = 0 (Sect. 2.2).

2.1. Phase-retrieval method.

This is Yuen (2011a), (G) for planar periodic membrane (a two-dimensional periodic object).

Use constraints:

(i) $f_0(x_1, x_2, x_3)$ has finite size with size = the known unit cell constants, $a, b, c, \pi/2, \pi/2, \gamma$

(ii) $f_0(x_1, x_2, x_3) \geq 0$

(iii) $f_0(x_1, x_2, x_3)$ is centrosymmetric

(iv) $f_0(x_1, x_2, x_3)$ is bounded

Because of the two-dimensional periodic structure, the diffracted intensity is greatly enhanced to form strong diffraction rods (Bragg rods) which can be measured accurately (Mancuso et al. (2009), p. 1). Let $\omega_{10} = 2\pi / a$, $\omega_{20} = 2\pi / b$. For all l and m, $\left| F_0(l\omega_{10}, m\omega_{20}, \omega_3) \right|$ are measured experimentally.

$$F_0(l\omega_{10}, m\omega_{20}, \omega_3)$$

$$= \int \left\{ \int_{-a/2}^{a/2} \int_{-b/2}^{b/2} f_0(x_1, x_2, x_3) \exp\left[-i\left(l\omega_{10}x_1 + m\omega_{20}x_2\right)\right] dx_1 dx_2 \right\} \exp\left(-i\omega_3 x_3\right) dx_3 \tag{1}$$

where the triple integral is over the one period of the membrane. For fixed l' and m', the term in curly brackets, $q(l'\omega_{10}, m'\omega_{20}, x_3)$, is a finite-size one-dimensional centrosymmetric real object of x_3. From Yuen (2011a), (B), get all $q(l\omega_{10}, m\omega_{20}, x_3)$.

$$f(x_1, x_2, x_3) = \frac{1}{ab} \sum_{l=-\infty}^{\infty} \sum_{m=-\infty}^{\infty} q(l\omega_{10}, m\omega_{20}, x_3) \exp\left[i\left(l\omega_{10} x_1 + m\omega_{20} x_2\right)\right]$$

The unique structure $f(x_1, x_2, x_3)$ including shape and dimensions of one period of the membrane are determined.

If (v) $f(x_1, x_2, x_3)$ is continuous with

$f_0(-a/2, x_2, x_3) = f_0(a/2, x_2, x_3)$ for all x_2, x_3

$f_0(x_1, -b/2, x_3) = f_0(x_1, b/2, x_3)$ for all x_1, x_3,

this will be shown in the structure $f(x_1, x_2, x_3)$ obtained.

2.2. Iterative algorithms with oversampling

Use Yuen (2011d), (J) to determine the maximum thickness of the membrane. Use an orthogonal or monoclinic unit cell centered at the origin to contain a period of the membrane. Then mathematically add replica of the unit cell successively to give a three-dimensional crystal consisting of parallel membranes. The | Fourier coefficients | are given by the magnitude of the Fourier transform of the content of the basic unit cell of the crystal. The in-between | Fourier coefficients | for $(l, m, n+\frac{1}{2})$ are obtained from the experimental diffraction intensity; no interpolation is needed. The in-between | Fourier coefficients | for $(l+\frac{1}{2},$ and/or $m+\frac{1}{2},$ $n)$ are obtained by interpolation in multidimensions. Yuen (2011b), Sect. 2.4 can then be applied.

Determine the one-dimensional electron density profile of the membrane by means of phase-retrieval method (Yuen (2011c)). The result is exact and unique. It is a fundamental property of the membrane, and can be used as a constraint for iterative algorithms with oversampling.

If we use

1. known value of the maximum thickness of the membrane. This is a loose support.
2. the known unit cell constants, the experimentally determined | Fourier coefficients | for (l, m, n) and in-between | Fourier coefficients | for $(l, m, n+\frac{1}{2})$,
3. random initial phases (0 or π),
4. oversampling ratio = $2^3 = 8$; use | Fourier coefficients | and in-between | Fourier coefficients | for all (l, m, n),

   and all ($l+\frac{1}{2}$, and/or $m+\frac{1}{2}$, and/or $n+\frac{1}{2}$).

   Some of them are experimental data (see 2.), the rest are obtained by interpolation in multidimensions.
5. constraints (i) to (iv). (v) is also used as a constraint.
6. the one-dimensional electron density profile of the membrane obtained by means of phase- retrieval method is used as a constraint
7. in the final iterations, the interpolated in-between | Fourier coefficients | are relaxed or adjusted, within their uncertainties, to satisfy the consistent condition (Yuen (2011b), Sect. 2.1); the interpolated in-between | Fourier coefficients | are improved.,

the iterative algorithms very probably converge to a non-negative electron density with R -index = 0. The electron density is very probably unique for this set of | Fourier coefficients | and in-between | Fourier coefficients | .

To compromise that proper initial phases are not used, we use known value of the maximum thickness of the membrane, and larger oversampling ratio = $2^3 = 8$; use | Fourier coefficients | and in-between | Fourier coefficients | for all (l, m, n), and all ($l+\frac{1}{2}$, and/or $m+\frac{1}{2}$, and/or $n+\frac{1}{2}$). These will help the iterative algorithms to converge to a unique non-negative electron density with R -index = 0. This convergence can be investigated empirically and theoretically.

As shown by the phase-retrieval method in Sect. 2.1, other than the experimentally determined in-between $|$ Fourier coefficients $|$ for $(l, m, n+\frac{1}{2})$, the interpolated in-between $|$ Fourier coefficients $|$ are, in fact, not needed. After the iterations converge to a unique non-negative electron density, we stop using all the interpolated in-between $|$ Fourier coefficients $|$. The iterative algorithms will converge to a **unique** non-negative electron density.

3. Discussions and conclusion

The phase-retrieval method in Sect. 2.1 provides the theoretical foundation and gives the conditions that iterative algorithms with oversampling will give a unique solution.

Because of the two-dimensional periodic structure, the diffracted intensity is greatly enhanced to form strong diffraction rods. As the oversampling ratio is exactly 2, for a planar periodic membrane, no interpolation is needed. The intensity along the diffraction rods can be measured experimentally. The electron density obtained is unique and exact. This is an important case of determining unique periodic structures without using interpolation. This provides an explanation to the important result of Lyman et al. (2005), p. 4, ‘it is possible to recover a 3D object with just one nonperiodic dimension available for oversampling.’ The *ab initio* electron density is determined by the experimental diffraction intensities alone; no other chemical or physical information (for example, atomicity, chemical formula, bond length, bond angle, etc) is used. It can be regarded as a fundamental property of the membrane. It is just a mathematical transformation of the diffraction intensities to an internal property of the membrane. Theories of molecular structure and properties (for example, quantum theory) should agree with this internal property. It forms a basis for interpretation of experimental data. The electron density obtained shows experimentally whether the membrane has sharp boundary or not.

According to Yuen (2011a), the solution of the phase problem is unique for centrosymmetic, real, bounded and finite-size object; non-negativity is not needed. From the non-negative electron density obtained, we stop using the constraint that the electron density is non-negative, and use the iterations. If we get the same result, this shows that the electron density is indeed non-negative.

It will be interesting if Sect. 2.2 can be applied to the centrosymmetic real anthracene thin film in Wu & Spence (2002). The results obtained can then be compared.

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Non-centrosymmetric non-negative nonperiodic object, crystal, fibre and membrane

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Solutions for the phase problem of n-dimensional non-centrosymmetric non-negative object

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Abstract

With general and reasonable constraints of the object, a solution of phase problem is obtained. The phase-retrieval method applies to non-negative objects, nonperiodic (aperiodic) and periodic, one and multidimensions, small molecule, fibre, membrane and macromolecule. Strong constraint of sharp boundary of an object is used. For periodic object, a unit cell is treated as the object. The known unit cell constants provide a strong constraint. For multidimensional non-negative object, it is very probable that the structure satisfying all the constraints is unique. For periodic object (dimension=1, 2 or 3), the oversampling ratio is exactly 2. The method is very suitable for determination of structures of periodic macromolecules, for example, protein crystals and biological fibres. This article gives the reason that oversampling can provide a solution as shown in many researchers' works. A theoretical basis that iterative algorithms with oversampling ratio=2 may lead to the correct structure is established. Iterative algorithm employing initial phase and gross structure obtained by the phase-retrieval method helps in avoiding stagnation problem with faster convergence to the correct answer. With oversampling ratio= 2, and the constraints presented, the phase-retrieval method, the iterative algorithms or their combinations can be applied for structure determination. Electron density is determined solely from diffraction intensity.

Keywords: Phase retrieval; nonperiodic; aperiodic; periodic; fibre; membrane; macromolecule; sharp boundary; Hilbert transform; discrete Hilbert transform; Shannon's sampling; minimum phase; iterative algorithms; oversampling; imaging; electron density distribution; one-dimensional periodic object; two-dimensional periodic object; diffraction rods; Bragg rods; in-between | Fourier coefficients| ; mid-Bragg intensities; superresolution.

1. Introduction

This is a sequel article of Yuen, 2011a, hereafter referred as I, which presents an analytical and unique solution for the phase problem of centrosymmetric real object. This article deals with non-centrosymmetric, non-negative object. A review of the one-dimensional phase-retrieval problem is presented in Klibanov et al. (1995) (see, e.g., (4.5)). For non-centrosymmetric non-negative object of known size, the structure may not be unique as illustrated in Bruck & Sodin (1979), Huiser & van Toorn (1980) and Lawton (1981). For multidimensional case, Sanz & Huang (1983) show that for irreducible function, solution of the phase problem is unique. Hayes & McClellan (1982) show that the set of reducible polynomials is a set of measure zero. Since it has much higher probability for a function of several complex variables, including polynomial, to be irreducible than reducible, it is 'pathologically' rare to have more than one solution for the phase problem, other than mirror image (Bruck & Sodin, 1979; Hayes, 1982; Barakat & Newsam, 1984; Bates, 1984). Manolitsakis (1982) questions this argument. He suggests that the probability of finding an irreducible function is lower than that of reducible function, because N distinct irreducible functions can be combined to form $2^N - N$ reducible functions. Stefanescu (1985) explains this controversy by showing that the reducible situations are in some sense unstable. Researchers have presented uniqueness conditions (for example, Greenaway, 1977; Crimmins & Fienup, 1983; Fiddy et al., 1983). Scivier et al. (1984) present a phase unwrapping algorithm using the complex zeros of a band-limited function and discuss the ambiguities in two dimensions. Nieto-Vesperinas (1984) investigates inverse scattering in terms of the zeros of entire functions. Nieto-Vesperinas & Dainty (1984),

(1986) study phase retrieval for two-dimensional digital objects by polynomial factorization. Klibanov (2006) presents a uniqueness theorem for recovering of a two-dimensional phase screen function, a complex object, from the magnitude of its Fourier transform. Density modification methods employing experimental constraints and physical constraints are discussed in Podjarny (1987). Fienup (1987), p. 122 demonstrates the importance of the sharpness (or tapering) of the edges of an object (see also Spence et al. (2002), Fienup (2006), He (2006) and Marathe et al. (2010)). For the application of Hilbert transform in phase retrieval problem, see the references in I.

Holmes & Belen'kii (2004) use the Cauchy-Riemann equations for one-dimensional phase retrieval problem. Rusu et al. (2002) derive phase from logarithmic gain derivatives. The phase problem can also be studied by means of zero-crossing (see, for example, Sanz, 1989). Three-beam X-ray diffraction can be used to determine the phase (Tang & Chang, 1990; Weckert & Hümmer, 1997; Chang, 2004; Shen, 2005). Szöke (2001) presents a method of using partially coherent X-ray to solve the crystallographic phase problem. Algebraic method is used to solve the phase problem for crystals with point-like atoms (Cervellino & Ciccariello, 1996, 2005). Direct calculation is used by Hau-Riege et al. (2004) to obtain the electron density represented by Gaussian basis functions from the structure factors. This involves solving a large number of quadratic equations. The iterative process may converge to a local minimum. Isernia et al. (1996) use the square amplitude distribution and a phase retrieval method employing minimization procedure. Crocco et al. (2004) study inverse scattering from phaseless measurements of the total field on a closed curve. Crocco et al. (2007) present a faithful non-linear imaging from amplitudes of incident and total fields. Semichaevsky & Testorf (2004) provide a phase-space interpretation of deterministic phase retrieval. Marengo et al. (2007) present an intensity-only signal-subspace-based imaging. Bates, Dainty, Fiddy, Gureyev, Nieto-Vesperinas, Nikulin, Nugent, Paganin, Wilkins and collaborators have many publications in phase retrieval (see, for example, Gureyev et al., 2004; Nikulin 1998; Nikulin & Steinfeld, 2004; Nikulin et al., 2008; Nugent, 2005; Paganin et al., 2004; Quiney et al., 2006 and their publications in the literature).

Theory of fibre diffraction can be found in Millane (1988) and Millane & Stroud (1995). Namba & Stubbs (1985) solve the phase problem for tobacco mosaic virus. Stroud & Millane (1995), (1996) present theory and simulation of diffraction by disordered polycrystalline fibres. Spence et al. (2003) have presented studies for crystalline monolayer. Liang et al. (2005) have used surface X-ray scattering to study the shape and structure of low-dimensional objects. Shi & Nehorai (2005), (2007) present time-reversal imaging. van der Plas & Millane (2000) present *ab initio* phasing in protein crystallography.

Much work has been done by means of iterative algorithms and oversampling, and has gained much success empirically (for example, Gerchberg & Saxton, 1972; Bates, 1982, 1984; Fienup, 1982; Miao et al., 1998; Miao et al., 1999; Miao et al., 2000; Miao & Sayre, 2000; Elser, 2003a, 2003b; Wu & Spence, 2003; Blankenbecler, 2004; Carrozzini et al., 2004; Oszlányi & Sütö, 2004; Chen et al., 2007; Kordyuk et al., 2007; Song et al., 2007; Elser & Millane, 2008; Baghaei et al., 2009; Shioya et al., 2010). Confined structure is introduced in De Caro et al., 2002. Reviews and tutorial are presented in Sayre (2002), Miao et al. (2004), van der Veen & Pfeiffer (2004), Millane (2006), Marchesini (2007) and Spence (2007). Various oversampling ratios have been used (for example, McBride et al., 2004; Kim et al., 2007; Song et al., 2007). Millane (1996) shows that the oversampling ratio is 4 for three dimensions. Miao et al. (1998) shows that the oversampling ratio >2. In this article, it is shown that in one, two and two dimensions, the oversampling ratio is exactly 2. In spite of these empirical successes, we need to find conditions under which iterative algorithms may lead to the correct answer so that we have confidence that the structure obtained is the correct structure. We deal with this problem in this article for real object, and in a sequel article (Yuen, 2011b) for complex object. Iterative alogorithm with oversampling is a very effective and efficient way for structure determination, both for nonperiodic and periodic objects.

The phase-retrieval problem can be reduced to a mathematical problem: What are the constraints of the object, under which the phase of the Fourier transform of an object can be obtained from the magnitude? Although it has higher probability for a function of several complex variables to be irreducible than reducible, there are still an infinite number of reducible functions. Hence the solution of the phase problem may not be unique. In this article, we use several general and reasonable constraints including the constraint that object has sharp boundary which is a generalization of that initialized by Fienup (1987), p. 122. It is very probable that these constraints will lead to unique solution of the phase problem (see Appendix).

Interactions between molecules depend on the electron densities. Hence it is important to determine the electron density accurately. At present, methods in solving crystal structures employ atomic scattering factors. This place more emphasis on the atomic coordinates. They can be determined accurately, while the electron density is less accurate (Coppens, 1997). By means of the phase-retrieval method presented, electron density is obtained directly from diffraction intensities. The in-between | Fourier coefficients | are very important. They can be obtained by interpolation in multidimensions or experimental measurement.

2.1. deals with one-dimensional non-negative object
2.2.1. General multidimensional non-negative object
2.2.2. Non-negative fibre
2.2.3. Non-negative membrane
2.2.4. Non-negative macromolecule
2.3. Finite $|\boldsymbol{\omega}|$ and combination with iterative algorithm**.** We give the reason that oversampling can provide very probably unique solution. A theoretical basis is established that iterative algorithms with oversampling ratio=2 may lead to the correct structure for nonperiodic or periodic object.

With oversampling ratio = 2, plus the strong constraints, non-negativity and sharp boundary, authors may apply the phase-retrieval method, the iterative algorithms or their combinations to re-determine or refine their structures reported in the literature.

In this article, by uniqueness of the solution, it means unique up to the structure of the object plus the mirror image; the location of the object is not determined. For periodic object, we deal with the content of the basic unit cell as an object, and define $f_0(\mathbf{x}) = f(\mathbf{x})$ for $\mathbf{x}$ ε the basic unit cell, or = 0 otherwise.

2. Method

2.1. One-dimensional non-negative object

The diffraction pattern is given by the Fourier transform of $f(x)$,

$$F(\omega) = \int_{-\infty}^{\infty} f(x)\exp(-i\omega x)dx$$

$$F(\omega) = |F(\omega)|\exp[i\phi(\omega)].$$

Experimentally, we can only measure $|F(\omega)|$; the phase $\phi(\omega), -\infty < \omega < \infty$, is missed. The present article uses constraints of $f(x)$ so that $\phi(\omega)$ can be recovered from $|F(\omega)|, -\infty < \omega < \infty$. The fewer the number of constraints employed, the wider the scope that the method is applicable. The general and reasonable constraints on an object include:

(i) finite size. This constraint is reasonable because either an object has finite size or experimentally we can only deal with finite part of the object. The unit cell of a crystal has finite size.

(ii) $f(x)$ is non-negative, $f(x) \geq 0$

(iii) $f(x)$ is bounded

(iv) $f(x)$ is sectionally continuous

(v) sharp boundary. $f(x) > 0$ at the boundary. In two- and three-dimensional cases, $f(x_1, x_2)$ and $f(x_1, x_2, x_3) > 0$ almost everywhere at the boundary line and boundary surface respectively. The set of boundary points where $f(x_1, x_2)$ and

$f(x_1, x_2, x_3) = 0$ has measure zero. This constraint is reasonable. An object is not related to the background and has sharp boundary; $f(x)$, $f(x_1, x_2)$ and $f(x_1, x_2, x_3)$ are usually > 0 at the boundary. $f(x)$, $f(x_1, x_2)$ and $f(x_1, x_2, x_3)$ in a unit cell of a periodic non-centrosymmetric object is usually, or can be chosen, > 0 at its boundary.

(vi) $f(x)$ is a continuous periodic object with $f_0(0) = f_0(L)$. L is the unit cell constant.

(vii) symmetry elements, if applicable

(viii) $f(x)$ is real

(ix) $f(x) \neq 0$ at the boundary. In two-dimensional case, $f(x_1, x_2) \neq 0$ almost everywhere at the boundary line. The set of boundary points where $f(x_1, x_2) = 0$ has measure zero.

Bilateral Laplace transform of $f(x)$, $F_{II}(s) = \int_{-\infty}^{\infty} f(x) \exp(-sx) dx$

$$(F_{II}(s))^* = F_{II}(s^*) \tag{1}$$

where * denotes complex conjugate. For a non-negative and bounded object $f(x)$ with finite size from $x = 0$ to $x = L$, $F_{II}(s)$ is an entire function of exponential type, order 1 and type L, but not of type less than L (Boas (1954), 6.8.1, Paley-Wiener theorem, Th. 6.8.7). $F(\omega) = F_{II}(i\omega)$ is a square integrable function. $F_{II}(s)$ has an infinite number of zeros (Titchmarsh, 1968, p. 287, Example 23). Let γ_1, γ_2,... be the zeros of $F_{II}(s)$. From Hadamard's factorization theorem,

$$F_{II}(s) = \left[\prod_{n=1}^{\infty} \left(1 - \frac{s}{\gamma_n} \right) \exp(s/\gamma_n) \right] \exp(a + bs) \tag{2}$$

where a and b are real constants.

The zeros occur in conjugate pairs. If we multiply $F_{II}(s)$ in (2) by

$$\frac{\gamma_n\left[\left(-\gamma_n^*\right)-s\right]}{\left(\gamma_n-s\right)\left(-\gamma_n^*\right)}\exp(-s/\gamma_n)\exp\left[s/\left(-\gamma_n^*\right)\right]\cdot$$

$$\frac{\gamma_n^*\left[\left(-\gamma_n\right)-s\right]}{\left(\gamma_n^*-s\right)\left(-\gamma_n\right)}\exp\left(-s/\gamma_n^*\right)\exp\left[s/\left(-\gamma_n\right)\right] \qquad (3)$$

The resulting function has the same magnitude at $i\omega$, but the phase is changed. The function has order = 1. The type is changed. The zero-pair γ_n, γ_n^* is flipped to the right-hand plane.

Let $\dot{\gamma}_m$ denotes a zero to be flipped. If N pairs of zeros are flipped, let

$$\Psi_N(s,\dot{\gamma}_m)=\prod_{m=1}^{N}\left(\frac{\dot{\gamma}_m-s}{\dot{\gamma}_m}\right)\exp\left(s/\dot{\gamma}_m\right)\left(\frac{\dot{\gamma}_m^*-s}{\dot{\gamma}_m^*}\right)\exp\left(s/\dot{\gamma}_m^*\right) \qquad (4)$$

and we obtain $F_{II}(s)\left(\Psi_N(s,\dot{\gamma}_m)\right)^{-1}\Psi_N\left(s,-\dot{\gamma}_m\right)$.

The right-hand side of (2) converges uniformly and absolutely in any bounded closed region of the s-plane (Copson (1962), Sect. 7.5). If N is a finite number, $F_{II}(s)\left(\Psi_N(s,\dot{\gamma}_m)\right)^{-1}\Psi_N\left(s,-\dot{\gamma}_m\right)$ also converges uniformly and absolutely in any bounded closed region of the s-plane. $F_{IIM}(s)\Psi_N(s,\dot{\gamma}_m)^{-1}$, being a partial product of the right-hand side of (2), converges uniformly and absolutely in this region (Forsyth (1965), p. 91). This remains true if $N\rightarrow\infty$. If we substitute $s=-s'$ in $\Psi_N\left(s,-\dot{\gamma}_m\right)$, from (4),

$$\Psi_N\left(s,-\dot{\gamma}_m\right)=\Psi_N(-s',-\dot{\gamma}_m)$$

$$=\prod_{m=1}^{N}\left(\frac{-\dot{\gamma}_m-(-s')}{-\dot{\gamma}_m}\right)\exp\left(s'/\dot{\gamma}_m\right)\left(\frac{-\dot{\gamma}_m^*-(-s')}{-\dot{\gamma}_m^*}\right)\exp\left(s'/\dot{\gamma}_m^*\right)=\Psi_N\left(s',\dot{\gamma}_m\right)$$

$\Psi_N\left(s',\dot{\gamma}_m\right)$, being a partial product of the term in square brackets in the right-hand side of (2), converges uniformly and absolutely in any bounded closed region.

Hence the product $F_{II}(s)\left(\Psi_N(s,\dot{\gamma}_m\right)^{-1}\Psi_N\left(s,-\dot{\gamma}_m\right)$ also converges uniformly and absolutely in any bounded closed region. Therefore, it is an entire function. By the Lindelöf's theorem (Boas (1954), Th. 2.10.1), this entire function is an entire function of exponential type (order=1) (see also Stefanescu (1985), p.2143).

In flipping zero-pairs, the magnitude of $F(\omega)=F_{II}(i\omega)$ is not changed. Hence, after a finite or denumerable number of zero-pairs are flipped, $F_{II}(i\omega)\left(\Psi_N(i\omega,\dot{\gamma}_m)\right)^{-1}\Psi_N\left(i\omega,-\dot{\gamma}_m\right)$ is a square integrable function. It is the two-sided Laplace transform of a finite size and bounded object.

Now, given a finite-size and bounded object $f(x)$, with size from $x=0$ to $x=L$. $f(x)$ is casual. Form its two-sided Laplace transform $F_{II}(s)$. The number of zeros of $F_{II}(s)$ in the right-half plane ($\operatorname{Re} s>0$) may be finite or denumerable. Flip all these zeros to the left-half plane. The resultant function, say $G_{II}(s)$ is the two-sided Laplace transform of a finite-size and bounded object $g(x)$. However, the size of this object may be from $x=c$ to $x=d$ with $c<0$; $g(x)$ is not casual. $G(\omega)=G_{II}(i\omega)$. Form the object $h(x)=g(x-c)$. $h(x)$ is casual. The two-sided Laplace transform of $h(x)$, $H_{II}(s)=G_{II}(s)\exp(-cs)$. The entire function $H_{II}(s)$ is analytic and has no zeros in the right-half plane. The Fourier transform of $h(x)$, $H_M(\omega)=H_{II}(i\omega)$ is a minimum-phase function. M stands for minimum-phase function.

$F_{II}(i\omega)$ is a square integrable function (Nussenzveig, 1972, p. 27, Titchmarsh's theorem). Hence $F(\omega)=F_{II}(i\omega)$ tends to zero as $|\omega|$ approaches ∞. Hence $\ln F(\omega)$ is not square integrable (Burge et al. (1976), p. 195). Titchmarsh's theorem cannot be applied to $\ln F(\omega)$ to obtain the phase of $F(\omega)$ from the magnitude. Burge et al. (1976) and Peřina (1985), Sect. 4.4 have dealt with this problem.

$$|F(\omega)| = |G(\omega)| = |H_M(\omega)|$$

In this article, we will apply Papoulis (1962), Sect. 10-3 to $H_{II}(s)$.

$$H_M(\omega) = |H_M(\omega)| \exp[i\phi_M(\omega)]$$

The phase $\phi_M(\omega)$ is uniquely determined from $\ln|H_M(\omega)| = \ln|F(\omega)|, -\infty < \omega < \infty$,

$$\phi_M(\omega) = -\frac{1}{\pi} P \int_{-\infty}^{\infty} \frac{\ln|F(y)|}{\omega - y} dy \tag{5}$$

This is a Hilbert transform (Papoulis, 1962, (10-67); Hahn, 1996, (7.17)). P stands for Cauchy principal value. Using $\phi_M(\omega)$, we get the minimum-phase function $H_M(\omega)$. By analytic continuation, $H_{II}(s) = H_M(s/i)$ (6)
All the zeros of $H_{II}(s)$ have $\operatorname{Re} s \le 0$. The order of each zero can be determined. For centrosymmetric real object, all the zeros with $\operatorname{Re} s < 0$ have even order (Yuen, 2011a, (B)). If this is not true, the object is not real centrosymmetric. If we flip the appropriate zero-pairs of $H_{II}(s)$ from the left-hand plane to the right-hand plane, we will get the $F_{II}(s)$ and $F(\omega)$ of the object that we are dealing with. The inverse Fourier transform of $F(\omega)$, gives $f(x)$, the desired structure of the object. The number of zero-pairs flipped may be finite or denumerable.

The Fourier transform of a non-negative function is known as a characteristic function. The bilateral Laplace transform of a non-negative function $f(x)$ has no zero on the real axis (Lukacs, 1970, Corollary to Th. 7.1.2). Therefore, if there is any zero on the real axis, $f(x)$ cannot be non-negative.

Remark 1. Let $\dot{\gamma}_m$ denotes a zero to be flipped. If N pairs of zeros are flipped and

N is a finite number,

$$\left(\Psi_N(s,\dot{\gamma}_m)\right)^{-1}\Psi_N\left(s,-\dot{\gamma}_m\right)$$

$$=\left\{\prod_{m=1}^{N}\frac{(-\dot{\gamma}_m{}^* - s)(-\dot{\gamma}_m - s)}{(\dot{\gamma}_m - s)(\dot{\gamma}_m{}^* - s)}\right\}\cdot$$

$$\left\{\prod_{m=1}^{N}\exp(-s/\dot{\gamma}_m)\exp(-s/\dot{\gamma}_m{}^*)\exp\left[s/(-\dot{\gamma}_m{}^*)\right]\exp\left[s/(-\dot{\gamma}_m)\right]\right\} \qquad (7)$$

The first factor of the right-hand side of (7) is known as a Blaschke-factor. If $N \to \infty$, the Blaschke-factor is not absolutely convergent (Titchmarsh (1926), Th. VI, Copson (1962), Sect. 7.1). Hence, if $F_{II}(s)$ has a denumerable number of zero-pairs in the right-hand plane, we cannot just use the Blaschke-factor to flip all these zero-pairs to the left-hand plane. Therefore we cannot use the Hilbert transform to get the phase from the magnitude. In flipping a denumerable number of zeros, the exponential factor in (3) should be included.

Remark 2. When a finite number of zero-pairs are flipped, multiplying $H_{II}(s)$ by the first factor of the right-hand side of (7) does not change the length of the object (Walther, 1963, p. 43, 45). Multiplying by the second factor changes the location of the object, in agreement with change of the type (Boas (1954), Th. 6.8.7).

When a denumerable number of zero-pairs are flipped, in above, we have shown that the product $H_{II}(s)\left(\Psi_N(s,\dot{\gamma}_m)\right)^{-1}\Psi_N\left(s,-\dot{\gamma}_m\right)$ converges uniformly and absolutely in any bounded closed region. Therefore, using the result for finite N, as $N \to \infty$, the length of the object is not changed, while its location changes. In summary, flipping a finite or denumerable number of zeros changes the location of the object, but not its length, as claimed in Walther (1963), p. 45. In particular, the length of the object given by the inverse Fourier transform of the minimum-phase function $H_M(\omega)$ is the length of the non-negative object.

(A) One-dimensional nonperiodic object

Constraints used:

(i) finite size from $x = 0$ to $x = L$. $f(x) = 0$ for x<0 or $x > L$. Value of L will be determined

(ii) $f(x)$ is non-negative, $f(x) \geq 0$

(iii) $f(x)$ is bounded

(iv) $f(x)$ is sectionally continuous

(v) sharp boundary. $f(x)$ >0 at the two boundary points

The above method is used to obtain $H_{II}(s)$. If appropriate pairs of zeros are flipped from the left-hand plane to the right-hand plane, we will obtain the object $f(x)$ with $f(x) \geq 0$, and its mirror image. For each pattern of the zeros, we can obtain a $F(\omega)$, $-\infty < \omega < \infty$. There are studies which decide whether $F(\omega)$ is a characteristic function or not (Papoulis, 1962, p. 225; Stuart & Ord, 1994, p. 133; Lukacs, 1970, Th.4.2.2., 4.2.3. and 4.2.5.). A direct way is to use the negativity index

$$= \left| \int_{-L/2}^{L/2} f_0(x)dx \right| \left[\int_{-L/2}^{L/2} f_0(x)dx \right]^{-1} \geq 0$$

where $f_0(x)$<0 in the first factor, and ≥ 0 in the second factor.

For each pattern of the zeros, we obtain $f(x)$, and calculate the value of the negativity index. The correct structure and mirror image have zero value of the negativity index.

A result of Cartwright is adapted in Nussenzveig (1967), Th. 7: If $f(x)$ is continuous within ($x_0 - a$, $x_0 + a$), and $f(x_0 \pm a) \neq 0$, and if $f'(x)$ is integrable, the zeros of $F_{II}(s)$ are asymptotically given by

$$\gamma_n \approx i\frac{n\pi}{a} - \frac{1}{2a}\ln\left[\frac{f(x_0 - a)}{f(x_0 + a)}\right] + \varepsilon_n$$

where $\varepsilon_n \to 0$ as $n \to \infty$.

In this case, to obtain the object $f(x)$, the "distant" zeros (Nussenzveig (1967), p. 569) can be treated as a group. Hence the number of possible flipping of zero-pairs is reduced from infinity to a finite number. In particular, if $f(x_0 - a) = f(x_0 + a)$, all the distant zeros tend to be on the imaginary axis, and no flipping is needed.

For the general case, $f(x)$ is sectionally continuous. The object has $f(x) > 0$ at its two boundary points. To obtain the object $f(x)$ by means of the inverse transform of $F(\omega)$, integration is used. Intuitively, we may think that the $f(x)$ obtained is continuous at the boundary except for the unique distributions of the zeros for the object and the mirror image which have sharp boundaries. The number of conjugate pairs of zeros is countably infinite. The number of points inside the object is non-denumerable. It is required that $f(x)$ satisfies the five constraints. The solution is probably unique.

If the object is continuous, this will be shown in some of the solutions. If continuity is used as a constraint, it is an additional constraint for uniqueness of the solution.

(B) One-dimensional periodic object (for example, crystal)

Choose the interval $[0, L]$ as the basic unit cell. Constraints used:

(i) $f_0(x)$ has finite size with size = the known unit cell constant, L (tight support, a strong constraint)

(ii) $f_0(x)$ is non-negative

(iii) $f_0(x)$ is bounded

(iv) $f_0(x)$ is sectionally continuous

(v) sharp boundary. $f_0(x) > 0$ at the two boundary points, 0 and L

Let $F_0(\omega)$ be the Fourier transform of $f_0(x)$. Discrete Hilbert transform gives

$$\phi_M(l\omega_{1/2}) = -\frac{2}{\pi}\sum \frac{\log|F_0(m\omega_{1/2})|}{l-m} \tag{8}$$

where $l = 0, \pm1, \pm2, \ldots$, $\omega_{1/2} = 2\pi/(2L) = \pi/L$, and summation over m, $l-m$ odd.

Fourier coefficients of $f(x)$ occur at $m\omega_0$, where $m = 0,\pm 1,\pm 2,...$, $\omega_0 = 2\pi / L$. $\omega_{1/2} = \omega_0 / 2$. We can obtain $|F_0((m+(1/2))\omega_0)|$, $m = 0,\pm 1,\pm 2,...$, by interpolation. The uncertainty introduced by interpolation is similar to experimental uncertainty. As explained in I, the result obtained by interpolation is fairly accurate. With $\phi_M(l\omega_{1/2})$ obtained from (8), we get $H_M(l\omega_{1/2})$. Then, for $-\infty < \omega < \infty$, Shannon's sampling theory gives

$$H_M(\omega) = \sum_{l=-\infty}^{\infty} H_M(l\omega_{1/2}) \frac{\sin(L\omega - l\pi)}{L\omega - l\pi} \tag{9}$$

Use (6). Find all the zeros of $H_{II}(s)$. Following (A), by moving systematically pairs of conjugate zeros so that $f_0(x)$ satisfies the five constraints, we obtain the structure of the object. The known size is a tight support, a strong constraint. If needed, adjust the interpolated $|F_0((m+(1/2))\omega_0)|$, $m = 0,\pm 1,\pm 2,...$. The solution $f_0(x)$ is very probably unique for each set of | Fourier coefficients| and in-between | Fourier coefficients| .

If $f(x)$ is known to be continuous, the interpolated in-between | Fourier coefficients| are adjusted to satisfy the constraint:

(vi) $f_0(x)$ is continuous
$f_0(0) = f_0(L)$

2.2. Multidimensional non-negative object

Assume that $f(x_1, x_2) \neq f_1(x_1) f_2(x_2)$ and
$f(x_1, x_2, x_3) \neq f_1(x_1) f_2(x_2) f_3(x_3)$.

2.2.1. General multidimensional non-negative object

(C) Multidimensional nonperiodic object

Constraints used:

(i) finite size. $f(x_1, x_2) = 0$ for $x_1 < 0$, $x_1 > L$, $x_2 < H(x_1)$ or $x_2 > K(x_1)$. Values of L, $H(x_1)$ and $K(x_1)$ will be determined.

(ii) $f(x_1, x_2)$ is non-negative, $f(x_1, x_2) \geq 0$

(iii) $f(x_1, x_2)$ is bounded

(iv) $f(x_1, x_2)$ is sectionally continuous

(v) sharp boundary. $f(x_1, x_2) > 0$ almost everywhere at the boundary line.

$$F(\omega_1, \omega_2) = \int_0^L \left\{ \int_{H(x_1)}^{K(x_1)} f(x_1, x_2) \exp(-i\omega_2 x_2) dx_2 \right\} \exp(-i\omega_1 x_1) dx_1$$

$$= \int_0^L q(x_1, \omega_2) \exp(-i\omega_1 x_1) dx_1 \qquad (10)$$

For a fixed $\omega_2 = \omega_2'$, this is a one-dimensional problem with $q(x_1, \omega_2')$ regarded as a complex object. Constraints used:

(i) $q(x_1, \omega_2')$ has finite size from $x_1 = 0$ to $x_1 = L$. Value of L will be determined.

(iii) $q(x_1, \omega_2')$ is bounded.

(iv) $q(x_1, \omega_2')$ is sectionally continuous

(1) needs not be satisfied. The zeros need not be symmetrical with respect to the real axis. Use $|F(\omega_1, \omega_2')|$ for all $\omega_1, -\infty < \omega_1 < \infty$. (5) and (8) apply to real or complex $f(x)$ (Papoulis, 1977, p. 251-254). Use treatment similar to Sect. 2.1. Flip the zeros, not zero-pairs. Flipping a finite or denumerable number of zeros does not change the length of the object. In particular, the length of the object given by the inverse Fourier transform of the minimum-phase function is the length of the object. Value of L is found.

Find the structure $q(x_1, \omega_2')$ which is consistent with these three constraints. The number of possible solutions, N is very large. Use the Fourier transform

$F(\omega_1,\omega_2')$ in (10) of each possible structure $q(x_1,\omega_2')$. Let them be ${}^1F(\omega_1,\omega_2')$, ${}^2F(\omega_1,\omega_2')$,..., ${}^NF(\omega_1,\omega_2')$. For all $\omega_2, -\infty<\omega_2<\infty$, $F(\omega_1,\omega_2)$ in (10) is an entire function. ${}^1F(\omega_1,\omega_2)$,..., ${}^NF(\omega_1,\omega_2)$ are obtained from $F(\omega_1,\omega_2)$ by flipping of zeros. Therefore ${}^1F(\omega_1,\omega_2)$, ..., ${}^NF(\omega_1,\omega_2)$, are N continuous functions of ω_1, ω_2. As shown in the Appendix, the solution is very probably unique; only one of the N possible structures $f(x_1,x_2)$ satisfies all the five constraints stated at the beginning of (C). We have to test the possible structures in order to find the structure satisfying the five constraints. Hence the structure $f(x_1,x_2)$ including values of L, $H(x_1)$ and $K(x_1)$ is determined. As the number of possible solutions is very large, much computation is needed.

Similar treatment for the three-dimensional case with constraint (v) sharp boundary, $f(x_1,x_2,x_3)>0$ almost everywhere at the boundary surface. It is very probable that the solution is unique. If the object is continuous or has symmetry elements, these will be shown in the result.

(D) Multidimensional periodic object (for example, crystal)

Choose the cell, $0\le x_1\le a$, $0\le x_2\le b$ as the basic unit cell. Constraints used:

(i) $f_0(x_1,x_2)$ has finite size with size = the known unit cell constants, a,b,α.

(ii) $f_0(x_1,x_2)$ is non-negative, $f_0(x_1,x_2)\ge 0$

(iii) $f_0(x_1,x_2)$ is bounded

(iv) $f_0(x_1,x_2)$ is sectionally continuous

(v) sharp boundary. $f_0(x_1,x_2)>0$ almost everywhere at the boundary parallelogram.

Let $\omega_{10}=2\pi/a$, $\omega_{20}=2\pi/b$. $\left|F_0(l\omega_{10},m\omega_{20})\right|$ are measured experimentally.

$$F_0(l\omega_{10}, m\omega_{20}) = \int_0^a \left\{ \int_0^b f_0(x_1, x_2)\exp(-im\omega_{20}x_2)dx_2 \right\} \exp(-il\omega_{10}x_1)dx_1$$

(11)

Consider a fixed m'. This is a one-dimensional problem with the term in curly brackets $q(x_1, m'\omega_{20})$ regarded as a complex object. Now $f_0(x_1, x_2) \neq 0$ almost everywhere at the boundary. Hence $q(x_1, m'\omega_{20})$ is usually $\neq 0$ at the two boundary points $x_1 = 0$, a.

Constraints used:

(i) $q(x_1, m'\omega_{20})$ has finite size from $x_1 = 0$ to a. Value of a is known (a strong constraint).

(iii) $q(x_1, m'\omega_{20})$ is bounded

(iv) $q(x_1, m'\omega_{20})$ is sectionally continuous

(ix) sharp boundary. $q(x_1, m'\omega_{20}) \neq 0$ at the two boundary points $x_1 = 0$, a.

Obtain $\left|F_0((l+1/2)\omega_{10}, m'\omega_{20})\right|$, $l = 0, \pm 1, \pm 2., \ldots$ by interpolation in multidimensions (Bates, 1984, p. 216; Kincaid & Cheney, 1996, Sect. 6.10; Cheney & Light, 2000; Anita, 2002, Sect. 4.7, 4.8, Hau-Riege et al., 2004) or by experimental measurement (Yuen, 2011d, for a crystal). The oversampling ratio is 2. Similar to (B) with known size a, flip the zeros. Obtain a structure $q(x_1, m'\omega_{20})$. The solution $q(x_1, m'\omega_{20})$ is very probably unique although the non-negativity constraint (ii) is not used. For continuous $f(x_1, x_2)$, the interpolated values of in-between | Fourier coefficients | are adjusted to satisfy the constraint:

(vi) $q(x_1, m'\omega_{20})$ is continuous

$$q(0, m'\omega_{20}) = q(a, m'\omega_{20}) .$$

Hence $F_0(l\omega_{10}, m'\omega_{20})$ is obtained for all l. For all m, $F_0(l\omega_{10}, m\omega_{20})$ is obtained. These give the Fourier coefficients. A structure $f(x_1, x_2)$ is obtained from the Fourier series. Adjust the interpolated in-between | Fourier coefficients | so that $f_0(x_1, x_2)$ satisfies the five constraints stated at the beginning of (D). As shown in the appendix, it is very probable that the structure obtained is unique.

If it happens that N solutions are found, N is small. Use the Fourier transform $F_0(\omega_1, m'\omega_{20})$ in (11) of each possible structure $q(x_1, m'\omega_{20})$. As in (C), for all m, associate the N functions $F_0(\omega_1, m'\omega_{20})$ from individual m to obtain N continuous functions. For each function, with the Fourier coefficients obtained from $F_0(l\omega_{10}, m\omega_{20})$ for all l and m, the Fourier series gives a structure $f(x_1, x_2)$. Hence we get N possible structures. Following the Appendix, it is very probable that the solution is unique; only one of the N structures $f_0(x_1, x_2)$ satisfies all the five constraints.

Similar treatment for the three-dimensional case with constraint (v) sharp boundary, $f_0(x_1, x_2, x_3) > 0$ almost everywhere at the boundary surface. It is very probable that the solution is unique.

In X-ray crystallography, for continuous electron density, the interpolated values are adjusted to satisfy:

(vi) $f_0(x_1, x_2, x_3)$ is continuous with

$$f_0(0, x_2, x_3) = f_0(a, x_2, x_3) \quad \text{for all } x_2,\ x_3$$

$$f_0(x_1, 0, x_3) = f_0(x_1, b, x_3) \quad \text{for all } x_1,\ x_3$$

$$f_0(x_1, x_2, 0) = f_0(x_1, x_2, c) \quad \text{for all } x_1,\ x_2$$

(vii) symmetry elements, if applicable

2.2.2. Non-negative fibre

(E) General non-negative fibre

Fibre has circular symmetry in the x_1x_2 plane with radius $R(x_3)$, and is periodic in x_3 with period c. Choose the cell $(x_1{}^2 + x_2{}^2)^{1/2} \leq R(x_3)$, $0 \leq x_3 \leq c$, as the basic unit cell.

Constraints used:

(i) For each x_3, $f_0(x_1, x_2, x_3)$ has finite radius $R(x_3)$ (value will be determined) in the x_1x_2 plane. $f_0(x_1, x_2, x_3) = 0$ for $x_3 < 0$ or $x_3 > c$. Value of c is known.

(ii) $f_0(x_1, x_2, x_3)$ is non-negative, $f_0(x_1, x_2, x_3) \geq 0$

(iii) $f_0(x_1, x_2, x_3)$ is bounded

(iv) $f_0(x_1, x_2, x_3)$ is sectionally continuous

(v) sharp boundary. $f_0(x_1, x_2, x_3) > 0$ almost everywhere at the two boundary circles at $x_3 = 0$ and c, and at the boundary surface of revolution about the x_3 axis.

The weak diffraction intensity is collected into strong diffraction planes which can be measured accurately. Let $\omega_0 = 2\pi / c$. For all l, $l = 0, \pm 1, \pm 2., \ldots$, $|F_0(\omega_1, \omega_2, l\omega_0)|$ are measured experimentally. It has circular symmetry in the $\omega_1\omega_2$ plane.

$$F_0(\omega_1, \omega_2, l\omega_0)$$

$$= \int_0^c \left\{ \iint f_0(x_1, x_2, x_3) \exp[-i(\omega_1 x_1 + \omega_2 x_2)] dx_1 dx_2 \right\} \exp(-il\omega_0 x_3) dx_3$$

where the double integral is over a circle for a given x_3.

For fixed $\omega_1{}'$ and $\omega_2 = 0$, this is a one-dimensional problem with the term in curly brackets $q(\omega_1',0,x_3)$ regarded as a real object. Constraints used:

(i) $q(\omega_1',0,x_3)$ has finite size from $x_3 = 0$ to c. Value of c is known (a strong constraint).

(iii) $q(\omega_1',0,x_3)$ is bounded

(iv) $q(\omega_1',0,x_3)$ is sectionally continuous

(viii) $q(\omega_1',0,x_3)$ is real

(ix) sharp boundary. $q(\omega_1',0,x_3) \neq 0$ at the two boundary points $x_3 = 0$, c

Obtain $|F_0(\omega_1',0,(l+(1/2))\omega_0)|$, $l = 0,\pm 1,\pm 2.,...$ by interpolation in multidimensions. As in (B) with known size c, find $q(\omega_1',0,x_3)$ and $F_0(\omega_1',0,l\omega_0)$. The solution is very probably unique although the non-negativity constraint (ii) is not used. For continuous $f(x_1,x_2,x_3)$, the interpolated values of in-between | Fourier coefficients | are adjusted to satisfy the constraint:

(vi) $q(\omega_1',0,x_3)$ is continuous

$$q(\omega_1',0,0) = q(\omega_1',0,c)$$

Repeat this for all ω_1.

For a fixed x_3', $f_0(x_1,x_2,x_3')=$ inverse Fourier transform of $q(\omega_1,\omega_2,x_3')$. For all x_3, $f_0(x_1,x_2,x_3)$ is obtained. Alternatively, $f(x_1,x_2,x_3)$ being periodic in x_3 with period c,

$$f_0(x_1,0,x_3) = \frac{1}{c}\sum_{l=-\infty}^{\infty}\left\{\frac{1}{2\pi}\int_0^{\infty} F_0(\omega_1,0,l\omega_0)J_0(x_1\omega_1)\omega_1 d\omega_1\right\}\exp(il\omega_0 x_3)$$

where $J_0(x_1\omega)$ is the 0th order Bessel function of the first kind.

The interpolated values $|F_0(\omega_1,0,(l+(1/2))\omega_0)|$ are further adjusted to satisfy:

(v) $f_0(x_1,x_2,x_3) > 0$ almost everywhere at the boundary surface of revolution about the x_3 axis,

while $q(\omega_1',0,x_3)$ remains satisfying the five constraints and $f_0(x_1,x_2,x_3)$

satisfying the other four constraints. A structure $f(x_1, x_2, x_3)$ is obtained with $R(x_3)$ determined. As shown in the appendix, it is very probable that the structure obtained is unique.

In X-ray crystallography, for continuous electron density, the interpolated values are adjusted to satisfy:

(vi) $f_0(x_1, x_2, x_3)$ is continuous

$$f_0(x_1, x_2, 0) = f_0(x_1, x_2, c) \quad \text{for all } x_1,\ x_2$$

(F) Fixed fibre molecule (a one-dimensional periodic object)

Choose the cell

$$H(x_2, x_3) \le x_1 \le K(x_2, x_3), L(x_3) \le x_2 \le M(x_3), 0 \le x_3 \le c,$$

as the basic unit cell. Constraints used:

(i) $f_0(x_1, x_2, x_3)$ has finite size. Values of $H(x_2, x_3)$, $K(x_2, x_3)$, $L(x_3)$ and $M(x_3)$ will be determined.

$f_0(x_1, x_2, x_3) = 0$ for $x_3 < 0$ or $x_3 > c$. Value of c is known.

(ii) $f_0(x_1, x_2, x_3)$ is non-negative, $f_0(x_1, x_2, x_3) \ge 0$

(iii) $f_0(x_1, x_2, x_3)$ is bounded

(iv) $f_0(x_1, x_2, x_3)$ is sectionally continuous

(v) sharp boundary. $f_0(x_1, x_2, x_3) > 0$ almost everywhere at the two boundary planes at $x_3 = 0$ and c, and at the boundary surface of the fibre.

The weak diffraction intensity is collected into strong diffraction planes which can be measured accurately. Let $\omega_0 = 2\pi / c$. For all l, $l = 0, \pm 1, \pm 2., \ldots$, $|F_0(\omega_1, \omega_2, l\omega_0)|$ are measured experimentally.

$F_0(\omega_1, \omega_2, l\omega_0)$

$$= \int_0^c \left\{ \iint f_0(x_1, x_2, x_3) \exp[-i(\omega_1 x_1 + \omega_2 x_2)] dx_1 dx_2 \right\} \exp(-il\omega_0 x_3) dx_3$$

where the triple integral is over the basic unit cell.

For fixed ω_1' and ω_2', this is a one-dimensional problem with the term in curly brackets $q(\omega_1', \omega_2', x_3)$ regarded as a complex object. Constraints used:

(i) $q(\omega_1', \omega_2', x_3)$ has finite size from $x_3 = 0$ to c. Value of c is known (a strong constraint).

(iii) $q(\omega_1', \omega_2', x_3)$ is bounded

(iv) $q(\omega_1', \omega_2', x_3)$ is sectionally continuous

(ix) sharp boundary. $q(\omega_1', \omega_2', x_3) \neq 0$ at the two boundary points $x_3 = 0$, c.

Obtain $|F_0(\omega_1', \omega_2', (l + (1/2))\omega_0)|$, $l = 0, \pm 1, \pm 2., \ldots$ by interpolation in multidimensions. As in (B) with known size c, find $q(\omega_1', \omega_2', x_3)$ and $F_0(\omega_1', \omega_2', l\omega_0)$. The solution is very probably unique although the non-negativity constraint (ii) is not used. For continuous $f(x_1, x_2, x_3)$, the interpolated values of in-between | Fourier coefficients | are adjusted to satisfy the constraint:

(vi) $q(\omega_1', \omega_2', x_3)$ is continuous

$q(\omega_1', \omega_2', 0) = q(\omega_1', \omega_2', c)$.

Repeat this for all ω_1, ω_2.

For a fixed x_3', $f_0(x_1, x_2, x_3')$ = inverse Fourier transform of $q(\omega_1, \omega_2, x_3')$. For all x_3, $f_0(x_1, x_2, x_3)$ is obtained. Alternatively, $f(x_1, x_2, x_3)$ being periodic in x_3 with period c,

$f(x_1, x_2, x_3) =$

$$\frac{1}{c} \sum_{l=-\infty}^{\infty} \left\{ \frac{1}{(2\pi)^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} F_0(\omega_1, \omega_2, l\omega_0) \exp[i(\omega_1 x_1 + \omega_2 x_2)] d\omega_1 d\omega_2 \right\} \exp(il\omega_0 x_3)$$

The interpolated values $|F_0(\omega_1', \omega_2', (l + (1/2))\omega_0)|$ are further adjusted to satisfy:

(v) $f_0(x_1,x_2,x_3) > 0$ almost everywhere at the boundary surface of the fibre,

while $q(\omega_1', \omega_2', x_3)$ remains satisfying the four constraints and $f_0(x_1,x_2,x_3)$ satisfying the other four constraints. A structure $f(x_1,x_2,x_3)$ is obtained with $H(x_2,x_3)$, $K(x_2,x_3)$, $L(x_3)$ and $M(x_3)$ determined. As shown in the appendix, it is very probable that the structure obtained is unique.

In X-ray crystallography, for continuous electron density, the interpolated values are adjusted to satisfy:

(vi) $f_0(x_1,x_2,x_3)$ is continuous

$$f_0(x_1,x_2,0) = f_0(x_1,x_2,c) \quad \text{for all } x_1,\ x_2$$

(vii) symmetry elements, if applicable

2.2.3. Non-negative membrane

The membrane may be an open surface or a closed surface. The thickness of the membrane may or may not be uniform.

(G) Nonperiodic non-negative membrane

Constraints used:

(i) finite size. The shape, dimensions and thickness of the membrane will be determined.

(ii) $f(x_1,x_2,x_3)$ is non-negative, $f(x_1,x_2,x_3) \geq 0$

(iii) $f(x_1,x_2,x_3)$ is bounded

(iv) $f(x_1,x_2,x_3)$ is sectionally continuous

(v) sharp boundary. $f(x_1,x_2,x_3) > 0$ almost everywhere at the two surfaces of the membrane, and at the edge if the membrane is an open surface.

$$F_0(\omega_1,\omega_2,\omega_3) =$$

$$\int_0^L \left\{ \int_{H(x_1)}^{K(x_1)} \left[\int_{P(x_1,x_2)}^{Q(x_1,x_2)} f(x_1,x_2,x_3)\exp(-i\omega_3 x_3)dx_3 \right] \exp(-i\omega_2 x_2)dx_2 \right\} \exp(-i\omega_1 x_1)dx_1$$

Follow the method in (C). If $f(x_1,x_2,x_3) \neq f_1(x_1,x_2)f_3(x_3)$, it is very probable that the solution is unique. The shape, dimensions and thickness of the membrane are determined. If $f(x_1,x_2,x_3)$ is continuous or has symmetry elements, these will be shown in the result.

Special cases:

(a) Circular membrane, $f(x_1,x_2,x_3) = f_1(x_1,x_2)f_3(x_3)$, where $f_1(x_1,x_2)$ is circularly symmetric.

$$F_0(\omega_1,\omega_2,\omega_3)$$

$$= \iint f_1(x_1,x_2)\exp[-i(\omega_1 x_1 + \omega_2 x_2)]dx_1 dx_2 \int f_3(x_3)\exp(-i\omega_3 x_3)dx_3$$

The solution for $f_1(x_1,x_2)$ is unique (Yuen, 2011a, case (C)). The solution for $f_3(x_3)$ is very probably unique. (see (A)).

(b) Thin membrane, the thickness of the membrane approaches 0. $f(x_1,x_2,x_3)$ becomes a function of two variables on the surface. It is very probable that the solution is unique (see (C)).

(H) Planar periodic non-negative membrane (a two-dimensional periodic object)

The object is periodic in x_1 and x_2 with periods a and b respectively. Choose the cell $0 \le x_1 \le a$, $0 \le x_2 \le b$, $H(x_1,x_2) \le x_3 \le K(x_1,x_2)$ as the basic unit cell. Constraints used:

(i) $f_0(x_1,x_2,x_3)$ has finite size with size = the known unit cell constants, a,b,α . $f_0(x_1,x_2,x_3) = 0$ for $x_3 < H(x_1,x_2)$ or $x_3 > K(x_1,x_2)$.

Values of $H(x_1,x_2)$ and $K(x_1,x_2)$ will be determined.

(ii) $f_0(x_1,x_2,x_3)$ is non-negative, $f_0(x_1,x_2,x_3) \ge 0$

(iii) $f_0(x_1, x_2, x_3)$ is bounded

(iv) $f_0(x_1, x_2, x_3)$ is sectionally continuous

(v) sharp boundary. $f_0(x_1, x_2, x_3) > 0$ almost everywhere at the six boundary surfaces of the basic unit cell

The weak diffraction intensity is collected into strong diffraction rods (Bragg rods) which can be measured accurately. Let $\omega_{10} = 2\pi / a$, $\omega_{20} = 2\pi / b$. For all l and m, $|F_0(l\omega_{10}, m\omega_{20}, \omega_3)|$ are measured experimentally.

$$F_0(l\omega_{10}, m\omega_{20}, \omega_3)$$

$$= \int_0^a \int_0^b \left\{ \int_{H(x_1,x_2)}^{K(x_1,x_2)} f_0(x_1, x_2, x_3) \exp(-i\omega_3 x_3) dx_3 \right\} \exp[-i(l\omega_{10}x_1 + m\omega_{20}x_2)] dx_1 dx_2$$

For fixed $\omega_3 = \omega_3'$, this is a two-dimensional case with the term in curly brackets $q(x_1, x_2, \omega_3')$ regarded as a complex object. Constraints used:

(i) $q(x_1, x_2, \omega_3')$ has finite size $0 \le x_1 \le a$, $0 \le x_2 \le b$. Known a, b, α (a strong constraint).

(iii) $q(x_1, x_2, \omega_3')$ is bounded

(iv) $q(x_1, x_2, \omega_3')$ is sectionally continuous

(ix) sharp boundary. $q(x_1, x_2, \omega_3') \ne 0$ almost everywhere at the four boundary lines $x_1 = 0$, a, $x_2 = 0$, b.

For example, $a > b$. Obtain $|F_0((l + 1/2)\omega_{10}, m\omega_{20}, \omega_3')|$, $l = 0, \pm 1, \pm 2., \ldots$, $m = 0, \pm 1, \pm 2., \ldots$ by interpolation in multidimensions. Follow (D), find $q(x_1, x_2, \omega_3')$ and $F_0(l\omega_{10}, m\omega_{20}, \omega_3')$. The solution is very probably unique although the non-negativity constraint (ii) is not used. For continuous $f(x_1, x_2, x_3)$, the interpolated values of in-between | Fourier coefficients | are adjusted to satisfy the constraints:

(vi) $q(x_1, x_2, \omega_3')$ is continuous

$$q(0, x_2, \omega_3') = q(a, x_2, \omega_3') \text{ for all } x_2$$

$$q(x_1, 0, \omega_3') = q(x_1, b, \omega_3') \text{ for all } x_1$$

(vii) symmetry elements, if applicable

Repeat this for all ω_3.

For fixed x_1', x_2', $f_0(x_1', x_2', x_3)$ = inverse Fourier transform of $q(x_1', x_2', \omega_3)$. For all x_1, x_2, $f_0(x_1, x_2, x_3)$ is obtained. Alternatively, $f(x_1, x_2, x_3)$ being periodic in x_1 and x_2 with periods a and b respectively,

$f(x_1, x_2, x_3) =$

$$\frac{1}{2\pi ab}\sum_{l=-\infty}^{\infty}\sum_{m=-\infty}^{\infty}\left[\int_{-\infty}^{\infty} F_0(l\omega_{10}, m\omega_{20}, \omega_3)\exp(i\omega_3 x_3)d\omega_3\right]\exp\left[i(l\omega_{10}x_1 + m\omega_{20}x_2)\right]$$

The interpolated values of $\left|F_0((l+1/2)\omega_{10}, m\omega_{20}, \omega_3')\right|$ are further adjusted to satisfy:

(v) $f_0(x_1, x_2, x_3) > 0$ almost everywhere at the other two boundary surfaces of the basic unit cell,

while $q(x_1, x_2, \omega_3')$ remains satisfying the four constraints and $f_0(x_1, x_2, x_3)$ satisfying the other four constraints. A structure $f(x_1, x_2, x_3)$ is obtained with $H(x_1, x_2)$ and $K(x_1, x_2)$ determined. As shown in the appendix, it is very probable that the structure obtained is unique.

In X-ray crystallography, for continuous electron density, the interpolated values are adjusted to satisfy:

(vi) $f_0(x_1, x_2, x_3)$ is continuous

$f_0(0, x_2, x_3) = f_0(a, x_2, x_3)$ for all x_2, x_3

$f_0(x_1, 0, x_3) = f_0(x_1, b, x_3)$ for all x_1, x_3

(vii) symmetry elements, if applicable.

2.2.4. Non-negative macromolecule

For nonperiodic and periodic non-negative macromolecule ((C) to (H)), it is very probable that the structure obtained is unique.

For periodic object (D), the in-between | Fourier coefficients| for one Fourier index l are obtained by interpolation in multidimensions. For crystal of a macromolecule, the unit cell is large. Hence the reciprocal lattice points are closely spaced. The interpolated value can be more accurate (Article I). This helps the determination of the structure. Hence this method is very suitable for the determination of structures of periodic macromolecules, for example, protein crystals and biological fibres with long period.

2.3. Finite $|\boldsymbol{\omega}|$ and combination with iterative algorithm

We cannot have both $f(\mathbf{x}) = 0$ for $|\mathbf{x}| > L$ and $F(\boldsymbol{\omega}) = 0$ for $|\boldsymbol{\omega}| > \omega_{\max}$ (Papoulis, 1962, p. 219, Prob. 39(b)). To calculate a finite-size object $f(\mathbf{x})$ from $F(\boldsymbol{\omega})$, $F(\boldsymbol{\omega})$ for all $\boldsymbol{\omega}$ has to be used. Suppose $f(\mathbf{x})$ is smooth. In practice, we use $|F(\boldsymbol{\omega})|$ for $|\boldsymbol{\omega}| \le \omega_{\max}$, where $\omega_{\max}$ is sufficiently large. The effect of the tail, $|F(\boldsymbol{\omega})|$ for $|\boldsymbol{\omega}| > \omega_{\max}$ although nonzero, should be negligible. Accurate structure $f(\mathbf{x})$ can be obtained. The smoother $f(\mathbf{x})$, the more rapidly $F(\boldsymbol{\omega})$ approaches zero with increasing $|\boldsymbol{\omega}|$ (Gaskill, 1978, pp. 191-192). If $F(\boldsymbol{\omega})$ for all $\boldsymbol{\omega}$, $0 < |\boldsymbol{\omega}| < \infty$, are used, this gives ultimate resolution. For example, even the electron density in the atoms can be determined. But this is not needed in our daily life. **Every $\omega_{\max}$ gives a structure including shape and dimensions with a definite resolution.** If experimentally we deal with $N/2$ pairs of conjugate zeros of $F_{IIM}(s)$, there are $2^{N/2}$ combinations. N depends on the resolution needed. If N is large, much computation will be involved. We may start with a small $\omega_{\max}$. Set $|F(\boldsymbol{\omega})| = 0$ for $\omega_{\max} < |\boldsymbol{\omega}| < \infty$. Apply the phase-retrieval method and neglect the contribution of the tail in (5). Corresponding to the small $\omega_{\max}$ is a small value of $|\mathbf{s}|_{\max}$ and a gross structure with low resolution. The number of zeros of $F_{IIM}(s)$ will be small. For centrosymmetric object, the zeros are symmetric with respect to the origin. Objects in nature have tendency to be centrosymmetric (Brames, 1986, p. 63). If we are dealing with molecules, because atoms have roughly spherical shape and the bond lengths and bond angles have certain values, we expect that the zeros for a non-centrosymmetric object have roughly same distributions in the left-hand plane and right-hand plane. Only in rare and extreme cases there are very few or no zero in one half plane. Therefore we arrange the conjugate pairs of zeros in ascending order according to their distances from the origin. This may be used as the initial structure for iterative algorithms. Then we move alternately pairs of zeros to the right-hand plane to satisfy the constraints and obtain a gross structure (Nussenzveig (1967), p. 570). In determination of molecular structure, the atoms appear very blurry with the heavier atoms being predominant. This may be useful in structure determination. If we need only a gross structure including shape and dimensions, we can stop here (for example, detection of an obstructing object in the course of an aeroplane). Otherwise we can extend the value of $|\mathbf{s}|_{\max}$, deal with more zeros, and obtain more refined structures.

From gross structure to more refined structure is a logical sequence in structure determination (Millane, 1988, Sect. 5.2). The refinement stops at the resolution needed. Alternatively, we may use a narrow strip centered at the imaginary axis with length given by the experimentally attained $\omega_{\max}$, so that it contains all the zeros of the experimentally observed $|F(\boldsymbol{\omega})|$ (Wackerman & Yagle (1994)). The width of the strip can then be increased. In all cases, $|\boldsymbol{\omega}|$ $\leq$ the experimentally attained $\omega_{\max}$.

An alternate method for phase retrieval is iterative algorithm (Gerchberg & Saxton, 1972, Fienup, 1982, other references in the Introduction). Use $|\boldsymbol{\omega}|$ $\leq$ a finite $\omega_{\max}$ means that the structure of the object obtained is of infinite extent. This violates the constraint of finite-size object. Hence we use a finite $\omega_{\max}$ corresponding to the resolution needed and use iterative algorithm. This fits in the tail, $F(\boldsymbol{\omega})$, $|\boldsymbol{\omega}| > \omega_{\max}$ by extrapolation (Goodman, 2005, Sect. 6.6, superresolution or bandwidth extrapolation; Gerchberg, 1974; Papoulis, 1977, p. 244). This also applies to centrosymmetric real object (Yuen, 2011a), and complex object with non-negative real and imaginary parts (Yuen, 2011b). Two criteria are used: (a) the observed $|F(\boldsymbol{\omega})|$, $|\boldsymbol{\omega}| \leq \omega_{\max}$, (b) the above constraints on the object. As a combination of the phase-retrieval method and the iterative algorithm, initially, use a small $\omega_{\max}$. The phase-retrieval method gives a gross structure of the object. Iterative algorithm is then applied. The phase of the Fourier transform of the gross structure can be taken as the initial phase. It may be better than the commonly employed random initial phase (Gerchberg & Saxton, 1972, p. 239; Sayre et al., 1998, p. 238; Miao et al., 2001; Wu & Spence, 2005, p. 195; Chapman et al., 2006a, p. 843). The gross structure, including shape and dimensions, of the object is used as initial input to the iterative process. This supplies more information than the known support employed in Fienup et al. (1982), p. 610. These initial inputs may help in avoiding the stagnation problem with faster convergence to the global minimum, the correct answer (Fienup & Wackerman, 1986). Use the initial phase and the gross structure avoids stagnation due to the possibility of the iterative process to proceed towards the object or its mirror image. Use the observed $\left|F(\boldsymbol{\omega})\right|$, $\left|\boldsymbol{\omega}\right| \leq$ the experimentally attained $\omega_{\max}$. Apply the iterative algorithm. The final result obtained must be consistent with the two criteria.

In one, two and three dimensions ((B), (D), (E), (F) and (H)), the oversampling ratio is exactly 2. A unit cell of a multidimensional periodic object is usually, or can be chosen, to have sharp boundary. Hence using an oversampling ratio= 2 is sufficient to give a solution of the phase problem. This is the reason that oversampling can provide a solution for periodic objects. For a multidimensional nonperiodic object with sharp boundary, replica can be used to give a periodic object. If we can choose a unit cell such that its boundary is sharp, oversampling can provide a solution for this nonperiodic object, which is very probably unique. (However, without *a priori* knowledge of the object, this condition will not be satisfied). Much work has done by many researchers with much empirical success. As it is very probable that the solution is unique for an object satisfying the constraints in this article, this provides a theoretical basis that iterative algorithms with oversampling ratio=2 may lead to the correct structure. When an iterative algorithm is applied to a multidimensional periodic object (D), the known unit cells constants are used as a tight support. The convergence will be faster and probably converges to the correct structure.

In the calculation of the inverse Fourier transform, occurrence of overshooting or undershooting (Gibbs' phoenomenon) demonstrates that sharp boundary of the object is encountered (see Appendix). For numerical treatment of discontinuity, see Sakurai (2004).

3. Discussion

In I, for centrosymmetric real object, the constraint, finite size, is employed. The object can taper to zero. In this article for non-centrosymmetric non-negative object, we use an additional constraint, the object has sharp boundary. This may be due to experimental limitation, or the object does have sharp boundary. If atoms and molecules do have sharp boundaries, this is very interesting and useful. It is different from quantum mechanical results. Sharp boundary may also exhibit in centrosymmetric real object (see I). For nonperiodic object, only finite size is used as a constraint for uniqueness. Value of the size (shape and dimensions) need not be known; this value is not used as a constraint. The size (shape and dimensions) is determined. In many cases, this is the result needed. The remote-sensing instrument in an aeroplane needs to detect the gross structure, including shape and dimensions, of an obstructing object in a short time. This is provided by employing a small $\omega_{\max}$ (Sect. 2.3).

For periodic object, a unit cell is treated as the object (Millane, 1996, p. 730). The size of this object is a unit cell with known unit cell constants. The difference between this and a nonperiodic object is that the periodicity of the structure greatly enhances the diffracted intensity which can be measured accurately. The known size is a very strong constraint. It also simplifies the reconstruction of the object by the iterative method (Fienup et al., 1982, p. 610 for tight support). An object may contain internal cavities where $f(\mathbf{x}) = 0$. Hence the size of an object contains its support where $f(\mathbf{x}) \neq 0$, and the internal cavities.

For periodic object (D), the strong constraint of known size can be applied. Because of the periodicity, sharp boundary is maintained for $q(x_1, m'\omega_{20})$ so that it is very probable the $q(x_1, m'\omega_{20})$ obtained is unique. This greatly simplifies the calculations. This also applies to (E), (F) and (H). For multidimensional nonperiodic object (C), a very large number of possible structures are obtained. We have to test the structures and find the structure which satisfies all the constraints.

In one, two and two dimensions ((B), (D), (E), (F) and (H)), the oversampling ratio is exactly 2.

The in-between | Fourier coefficients| (or mid-Bragg intensities) are very important quantities. May be in the future, articles in X-ray crystallography could include these values. Besides obtaining these values by interpolation in multidimensions, Yuen (2011d) presents an attempt to measure them experimentally.

As mentioned in Sect. 2.2.4, the method used in this article is very suitable for structure determination of protein crystals and biological fibres. For these macromolecules, structure determination had been fairly complicate or difficult.

The phase-retrieval method employed in this article uses flipping of zeros. It will be very useful if we can find an expression for the solution so that flipping of zeros is not needed. Mathematical proof that the constraints used in the Appendix do lead to unique solution, is needed. Method of projection may be considered (Youla, 1987; Levi & Stark, 1987; Elser, 2003a; Elser et al., 2007).

In flipping a denumerable number of zeros, the exponential factor in (3) is included (Remark 1). In the literature, articles involving flipping of zeros may include this exponential factor in their derivations, not just the Blaschke-factor.

In Remark 2, we have shown that the length of the object given by the inverse Fourier transform of the minimum-phase function $H_M(\omega)$ is the length of the non-negative object. This important result for a real or complex object will be used in Yuen (2011c).

Iterative algorithm employing initial phase and gross structure obtained by the phase-retrieval method helps in avoiding stagnation problem with faster convergence to the correct answer. Use oversampling ratio=2 applied to the longest unit cell edge, and the constraints in this article, including the strong constraints, non-negativity and sharp boundary. Algorithms applicable to real data may be developed. The authors may apply the phase-retrieval method presented in above, the iterative algorithms (X-ray diffraction microscopy) or their combinations to re-determine their structures in the literature. The results can be compared. Because of noise in the experimental data, more refined sampling with higher sampling ratio will be more effective for the iterative process to converge to the global minimum (Miao et al., 1998, p. 1667). The structure determined will be electron density from diffraction intensity only. Concept of atom and other knowledge of physics and chemistry are not used. The chemical formula of the molecule, bond lengths and bond angles need not be known. In fact, this can be used as a tool for identifying the atoms and establishing molecular formula. For freely rotating molecule and fibre, the electron density obtained may be used as fundamental data for model fitting. Determination of the structures of macromolecules, for example, proteins, nuclei acids, protein-nuclei acid complexes, viruses, is very important, particularly in medicine and biology. Electron density determines the active site of the molecule. These help the understanding of biochemical processes.

The phase-retrieval method or iterative algorithms may be used in the imaging processes involving various kinds of waves, electromagnetic wave, acoustic wave, etc. (Bates et al., 1991, Table 6.2). In fact, many disciplines involve wave or particle scattered from an object, these methods will be applicable. Finding the solutions of the phase problem or inverse scattering problem is just like getting the key to the treasury of Knowledge. We can enter into the room and have many new and exciting discoveries.

4. Conclusions

As a conclusion, in scattering and diffraction for multidimensional non-negative object, the phase may not be lost; it is just hidden in the magnitude. Using five general and reasonable constraints of finite-size object, non-negative, bounded, sectionally continuous and sharp boundary, it is very probable that a unique solution of the phase problem may be obtained. A phase-retrieval method is presented to recover the phase from the magnitude. The method is very suitable for determination of structures of periodic macromolecules, for example, protein crystals and biological fibres. It is important to determine whether atoms and molecules do have sharp boundary. For a nonperiodic or periodic object satisfying these five constraints, a theoretical basis has been established that iterative algorithms with oversampling ratio=2 may lead to the correct structure. The in-between | Fourier coefficients| are very important quantities. In the future, they may be included in articles in X-ray crystallography. It is worthwhile to spend effort to develop the technology of measuring the in-between | Fourier coefficients | . The phase-retrieval method, the iterative algorithms or their combinations determine electron density from diffraction intensity only. These methods also serve as a tool for identifying atoms and establishing molecular formula. They are useful in many fields including structure determination, imaging, biology and medicine. Researchers may develop algorithms applicable to real data or find an expression for the solution. The authors may apply the phase-retrieval method, the iterative algorithms or their combinations to re-determine or refine their nonperiodic and periodic structures in the literature. It remains to find an expression for the solution of the phase problem so that flipping of zeros is not needed, and to establish mathematical proof that the constraints used indeed lead to a unique solution.

Appendix

The zeros of a function of several complex variables are never isolated. Let $\mathbf{s} = (s_1,...,s_n)$. By Osgood's theorem (Manolitsakis, 1982, (3.3), Appendix A), an entire function $F_{II}(\mathbf{s})$ with domain C^n has unique factorization into a finite or infinite number of irreducible factors.

$$F_{II}(\mathbf{s}) = \prod_{j=1}^{N} \left\{P_j(\mathbf{s}) \exp\left[Q_j(\mathbf{s})\right]\right\}^{l_j} \qquad (N \le \infty) \tag{12}$$

where $P_j(\mathbf{s})$ are irreducible entire functions but not necessarily of exponential type

(Scivier & Fiddy, 1985, p. 694). $Q_j(\mathbf{s})$ are polynomials in $\mathbf{s}$. $\exp\lfloor Q_j(\mathbf{s})\rfloor$ are convergence factors and l_j are integers. We call the zeros of an irreducible factor, a zero-cluster. The number of zero-clusters may be finite or infinite.

If $F_{II}(\mathbf{s})$ is reducible, all solutions of the phase problem are obtained by flipping the zero-clusters (Manolitsakis, 1982, p. 2296, Stefanescu, 1985, Prop. 2). Manolitsakis, 1982, p. 2296 and Stefanescu, 1985, p.2142 show that a whole zero-cluster should be flipped and not part of it. For real object, a pair of conjugate zero-clusters should be flipped. A zero-cluster can be conjugate to itself. If $F_{II}(\mathbf{s})$ has $N/2$ pairs of zero-clusters, the number of possible structures is at most $2^{N/2}$ (Stefanescu, 1985, p. 2144). Hence if $F_{II}(\mathbf{s})$ has one zero-cluster, or two conjugate zero-clusters, the solution of the phase problem is unique, plus the mirror image. $F_{II}(\mathbf{s})$ is most likely irreducible (Bruck & Sodin, 1979; Hayes, 1982; Barakat & Newsam, 1984; Bates, 1984). In this case, there is only one zero-cluster. It is 'pathologically' rare for $F_{II}(\mathbf{s})$ to be reducible into two factors with a pair of conjugate zero-clusters. Even if this occurs, the solution of the phase problem is still unique. It is even rarer that $F_{II}(\mathbf{s})$ is reducible into two conjugate factors each of them is reducible so that there are more than one pair of zero-clusters, and the solution is not unique.

Let $F(\boldsymbol{\omega})$ be the Fourier transform of a finite-size multidimensional object. If $F(\boldsymbol{\omega})$ is reducible into two factors, $F(\boldsymbol{\omega}) = F_1(\boldsymbol{\omega})F_2(\boldsymbol{\omega})$
The object $f(\mathbf{x}) = f_1(\mathbf{x}) \otimes f_2(\mathbf{x})$
where $f_1(\mathbf{x})$ and $f_2(\mathbf{x})$ are the inverse Fourier transforms of $F_1(\boldsymbol{\omega})$ and $F_2(\boldsymbol{\omega})$ respectively. $\otimes$ denotes convolution.
If $f_1(\mathbf{x})$ and $f_2(\mathbf{x})$ are objects of finite and non-zero size, $f_1(\mathbf{x}) \otimes f_2(\mathbf{x}) = 0$ at its boundary (Papoulis, 1968, p. 32).

Consider a nonperiodic non-negative object. Constraints used:

(i) finite size. $f(x_1, x_2) = 0$ for $x_1 < 0$, $x_1 > L$, $x_2 < H(x_1)$ or $x_2 > K(x_1)$. Values of L, $H(x_1)$ and $K(x_1)$ will be determined.

(ii) $f(x_1, x_2)$ is non-negative, $f(x_1, x_2) \geq 0$

(iii) $f(x_1, x_2)$ is bounded

(iv) $f(x_1, x_2)$ is sectionally continuous

(v) sharp boundary. $f(x_1, x_2) > 0$ almost everywhere at the boundary line. The set of boundary points where $f(x_1, x_2) = 0$ has measure zero. The object has sharp boundary. For three-dimensional object, boundary surface is used.

Lawton (1981), p. 1519 and Fienup (1983), p. 100 have constructed objects by means of convolution of components. The Fourier transforms of these objects have the same magnitudes. These objects = 0 at the boundary and are excluded.

To obtain the object $f(\mathbf{x})$ by means of the inverse transform of $F(\boldsymbol{\omega})$, integration is used. Intuitively, we may think that the $f(\mathbf{x})$ obtained is continuous at the boundary except for the unique distributions of the zero-clusters for the object and the mirror image which have sharp boundaries. The number of points inside the object is non-denumerable. In two-dimensional case, the boundary is a closed curve. In three-dimensional case, it is a closed surface. The number of boundary points is also non-denumerable. The number of pairs of conjugate zero-clusters is one, finite or countably infinite. For $N/2$ pairs of zero-clusters, the number of possible structures is at most $2^{N/2}$. Constraints (ii) and (v) are very strong (Fienup, 1987). Bruck & Sodin (1979) have shown that the structure may not be unique for non-negative discrete object which consists of finite number of points. This result cannot be generalized to continuous object consisting of non-denumerable number of points. This generalization is questioned by Manolitsakis (1982), p. 2291.

If the two-sided Laplace transform $F_{II}(\mathbf{s})$ of a real object has more than one pair of zero-clusters, each pair can be flipped. We have to check whether the new object obtained satisfy the five constraints, in particular, the strong constraints of finite size, non-negativity and sharp boundary.

Because of the discontinuity at the boundary of the object, in the calculation of the inverse Fourier transform, overshooting or undershooting (Gibbs' phoenomenon) will occur (Papoulis, 1962, p. 30). In one dimension, the overshoot or undershoot is approximately 9% of the discontinuity. In two-dimension, an example of overshooting is ringing in optical imaging (Goodman, 2005, p. 160).

Huiser & van Toorn (1980) have constructed objects by means of flipping of zero clusters. These objects satisfy constraints (i), and probably (iii) and (iv). The objects (Fig. 2(a)-3(a)) are claimed to be non-negative. Because numerical method is used, there may be negative regions of small magnitude which are neglected. Moreover, Fig. 1(b)-3(a) do not show whether constraint (v) is satisfied. If these objects do have sharp boundary, there should be negative region due to undershooting. This is not mentioned in their article.

Combine with the finding that a function of several complex variables is pathologically rare to be reducible, and more unlikely to have more than one pair of zero-clusters. By flipping the pairs of zero-clusters, there is very small probability of finding more than one structures which satisfy all the five constraints. It is very probable that in multidimensions, the solution of the phase problem is unique, plus the mirror image. It may even be possible that constraints (i)-(v) plus the finding that a whole zero-cluster has to be flipped are sufficient for the solution to be unique.

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11

Solutions for the phase problem of n-dimensional complex object with non-negative real and imaginary parts

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Abstract

With general and reasonable constraints of complex object with non-negative real and imaginary parts, a solution of phase problem is obtained. The phase-retrieval method applies to centrosymmetric and non-centrosymmetric objects, nonperiodic and periodic, one and multidimensions, small molecule, fibre, membrane and macromolecule. For multidimensional object, it is very probable that the structure satisfying the ten constraints is unique. For periodic object (dimension=1, 2 or 3), the oversampling ratio is exactly 2. The method is very suitable for determination of structures of periodic macromolecules, for example, protein crystals and biological fibres. This article gives the reason that oversampling can provide a solution as shown in many researchers' works. For nonperiodic or periodic complex object, a theoretical basis is established that iterative algorithms with oversampling ratio=2 may lead to the correct structure. With oversampling ratio=2, plus strong constraints, non-negativity and sharp boundary of the real and imaginary parts, the phase-retrieval method, the iterative algorithms or their combinations can be applied for structure determination.

Keywords: Phase retrieval; nonperiodic; aperiodic; periodic; fibre; membrane; macromolecule; sharp boundary; Hilbert transform; discrete Hilbert transform; Shannon's sampling; minimum phase; iterative algorithms;

oversampling; electron density distribution; one-dimensional periodic object; two-dimensional periodic object; diffraction rods; Bragg rods; in-between｜Fourier coefficients｜; mid-Bragg intensities.

1. Introduction

This is a sequel article of Yuen (2011a) and Yuen (2011c), hereafter referred as I and II respectively. This article deals with centrosymmetric and non-centrosymmetric complex object with non-negative real and imaginary parts.

Studies of anomalous scattering in fibers are presented in Nave (1987). Wang et al. (2006) present phase determination for membrane diffraction of a centrosymmetric system by anomalous dispersion and Patterson function. Anomalous scattering in macromolecular structure analysis is discussed in Hendrickson (1987), Karle (1989), Matthews (2001) and Smith et al. (2001). Single-wavelength anomalous dispersion method is discussed in Rice et al. (2000) and Dauter et al. (2002). Multiwavelength anomalous dispersion method is presented in Philips et al. (1977), Philips & Dodgson (1980), Hendrickson & Teeter (1981) and Hendrickson (1998). Anomalous dispersion of macromolecules in solution is presented in Stuhrmann & Notbohm (1981). Parametric X-radiation is presented in Feranchuk & Ulyanenkov (2005). In the optical model of nuclear structure, complex model is used (Feshbach et al., 1954).

Lawton (1981) shows that for radially symmetric complex function, there are an infinite number of solutions [see also Kiedron (1981) and Ehm et al. (2004)]. Klibanov (2006) presents a uniqueness theorem for recovering of a two-dimensional phase screen function, a complex object, from the magnitude of its Fourier transform. If an object consists of sufficiently separated parts, the solution is almost always unique (Greenaway, 1977; Crimmins & Fienup, 1983). Fiddy et al. (1983) enforce irreducibility of a polynomial and find that the correct solution can be obtained by Fienup algorithm. Fienup (1983a) and (1983b) show that the shape of an object is important in determining whether a solution is unique, and provide experimental evidence for the uniqueness. Dainty & Fiddy (1984) and Brames (1986) have investigated the problem of uniqueness in terms of the support of two-dimensional object. Fienup (1987) shows that it is possible to reconstruct a complex-valued object for separated or special shapes of supports. Isernia et al. (1999) present a unified discussion of support information and zero locations in phase retrieval by a quadratic approach. Yagle & Bell, 1999 have developed algorithm for solving the phase problem by solving linear systems of equations. A review of diffraction with a coherent X-ray beam is presented in Livet (2007).

The relative complex dielectric constant,

$$\varepsilon_r(\mathbf{x}) = \varepsilon(\mathbf{x})/\varepsilon_0 - j\sigma(\mathbf{x})/(\omega\varepsilon_0) \tag{1}$$

where $\varepsilon(\mathbf{x})$ is the permittivity, $\sigma(\mathbf{x})$ the conductivity , ω the angular frequency (Ishimaru, 1991, (2-37)). The magnitude of the Fourier transform of $\varepsilon_r(\mathbf{x})$ is obtained in optical scattering. Both the terms $\varepsilon(\mathbf{x})/\varepsilon_0$ and $\sigma(\mathbf{x})/(\omega\varepsilon_0)$ are non-negative, hence it is meaningful to deal with the phase problem of n-dimensional complex object with non-negative real and imaginary parts. In this article, we show that with general and reasonable constraints applied to the object, the solution is very probably unique.

This article does not apply to a conductor at frequencies with small depth of penetration.

Use of convex constraints in phase retrieval is presented in Youla, 1987 and Weierstall et al., 2002 (Spence (2011)). Iterative algorithms and oversampling for complex object have been used by many researchers with much empirical success (for example, Gerchberg & Saxton, 1972; Bates, 1982, 1984; Fienup & Kowalczyk, 1990; Marchesini et al., 2003; Wu & Spence, 2005; Chapman et al., 2006a, 2006b; Fienup, 2006; He, 2006; Williams et al., 2007; Montiel et al., 2008; Takahashi, et al., 2008; Volkov et al., 2008; Kim et al., 2010; Wu et al., 2010). Non-negative constraints on the real or imaginary part of complex object have been used by Miao et al. (1998), Sayre et al. (1998) and Miao & Sayre (2000). For complex object, importance of the sharpness (or tapering) of the edges of an object is investigated in Fienup (1987). This is discussed in the review by Spence et al. (2002). McBride et al. (2004) have presented retrieval of periodic complex-valued object. Marchesini (2007) presents phase retrieval and saddle-point optimization. Guizar-Sicairos & Fienup (2008) presents phase retrieval with Fourier-weighted projections. Review of the X-ray diffraction microscopy is presented in Miao et al. (2004). In spite of these empirical successes, we need to find conditions under which iterative algorithms may lead to the correct answer so that we have confidence that the structure obtained is the correct structure. In the present article, we deal with this problem for complex object.

In this article,
2.1. deals with one-dimensional complex object
2.2.1. General multidimensional complex object
2.2.2. Complex fibre
2.2.3. Complex membrane
2.2.4. Complex macromolecule
2.3. Finite $|\boldsymbol{\omega}|$ and combination with iterative algorithm**.** A theoretical basis is established that iterative algorithms with oversampling ratio=2 may lead to the correct structure for nonperiodic or periodic complex object.

In this article, by uniqueness of the solution, it means unique up to the absolute configuration of the object; the location of the object is not determined. The size of the object is the union of sizes of the real part and the imaginary part. For periodic object, we deal with the content of the basic unit cell as an object, and define $f_0(\mathbf{x}) = f(\mathbf{x})$ for $\mathbf{x}$ εthe basic unit cell, or = 0 otherwise. $f_0(\mathbf{x}) = f_{r0}(\mathbf{x}) + jf_{i0}(\mathbf{x})$ where $f_{r0}(\mathbf{x})$ and $f_{i0}(\mathbf{x})$ are the real and imaginary parts respectively. The real part and imaginary part have the same basic unit cell.

2. Method

Let $\mathbf{x},\boldsymbol{\omega}\varepsilon R^n$, $\mathbf{s}\varepsilon C^n$. Let $f(\mathbf{x}) = f_r(\mathbf{x}) + jf_i(\mathbf{x})$ be a complex object, where $f_r(\mathbf{x})$ and $f_i(\mathbf{x})$ are real. Let $F(\boldsymbol{\omega})$, $F_r(\boldsymbol{\omega})$ and $F_i(\boldsymbol{\omega})$ be the Fourier transforms of $f(\mathbf{x})$, $f_r(\mathbf{x})$ and $f_i(\mathbf{x})$ respectively, and $F_{II}(\mathbf{s})$, $F_{rII}(\mathbf{s})$, $F_{iII}(\mathbf{s})$ their bilateral Laplace transforms. $F_{rII}(\mathbf{s})$ and $F_{iII}(\mathbf{s})$ are real analytic functions. $F(\boldsymbol{\omega}) = F_r(\boldsymbol{\omega}) + jF_i(\boldsymbol{\omega})$.

$F(-\boldsymbol{\omega}) = (F_r(\boldsymbol{\omega}) - jF_i(\boldsymbol{\omega}))^*$.

$F(-\boldsymbol{\omega})$ is, in general, not equal to $(F(\boldsymbol{\omega}))^*$ for non-centrosymmetric object . Therefore Friedal's law is not satisfied.

If $g(\mathbf{x})$ is the mirror image of the object, $g(\mathbf{x}) = f(-\mathbf{x})$. $G(\boldsymbol{\omega}) = F(-\boldsymbol{\omega})$. Therefore from the experimental $|F(\boldsymbol{\omega})|$, the absolute configuration of the object can be determined.

If the object is centrosymmetric, $f(\mathbf{x}) = f(-\mathbf{x})$, then $F(\boldsymbol{\omega}) = F(-\boldsymbol{\omega})$, although it is usually complex. (See anomalous scattering in X-ray crystallography).

Let $h(\mathbf{x}) = (f(-\mathbf{x}))^*$ be the twin of $f(\mathbf{x})$ (Fienup & Wackerman, 1986, p. 1897), and $H(\boldsymbol{\omega})$ its Fourier transform.

$H(\boldsymbol{\omega}) = (F(\boldsymbol{\omega}))^*$ with $|H(\boldsymbol{\omega})| = |F(\boldsymbol{\omega})|$

Hence, given $|F(\boldsymbol{\omega})|$, we cannot recover a complex object uniquely .

In the special case of real $f(\mathbf{x})$, the twin $(f(-\mathbf{x}))^*$ reduces to the mirror image, $f(-\mathbf{x})$.

To resolve the conjugate ambiguity and obtain the unique solution, the non-negativity or non-positivity constraint on either the real part or imaginary part must be employed (Miao et al., 2000, p. 598; Spence et al., 2002, p. 880). In X-ray diffraction, the imaginary part of $f(\mathbf{x})$ is always non-negative (Miao et al., 1998, p. 1664). Hence unique solution of the phase problem can be obtained.

Consider a nonperiodic complex object $f(x_1, x_2) = f_r(x_1, x_2) + jf_i(x_1, x_2)$. For the real part $f_r(x_1, x_2)$, constraints used:

(i) finite size. $f_r(x_1, x_2) = 0$ for $x_1 < 0$, $x_1 > L_r$, $x_2 < H_r(x_1)$ or $x_2 > K_r(x_1)$. Values of L_r, $H_r(x_1)$ and $K_r(x_1)$ will be determined.

(ii) $f_r(x_1, x_2)$ is non-negative, $f_r(x_1, x_2) \geq 0$

(iii) $f_r(x_1, x_2)$ is bounded

(iv) $f_r(x_1, x_2)$ is sectionally continuous

(v) sharp boundary. $f_r(x_1, x_2) > 0$ almost everywhere at the boundary lines. The set of boundary points where $f_r(x_1, x_2) = 0$ has measure zero. The object has sharp boundary. For three-dimensional object, boundary surfaces are used,

with similar constraints for the imaginary part $f_i(x_1, x_2)$.

The two-sided Laplace transform $F_{II}(s_1, s_2)$ of $f(x_1, x_2)$ has unique factorization of a finite or countably infinite number of irreducible factors ((14) of article II). If $F_{II}(s_1, s_2)$ has more than one factor, the zero-clusters can be flipped. We have to check whether the new object obtained satisfy the five constraints for $f_r(x_1, x_2)$ and the five constraints for $f_i(x_1, x_2)$, in particular, the six strong constraints of finite size, non-negativity and sharp boundary. The number of zero-clusters is one, finite or countably infinite. The number of points inside the object and at the boundary line or surface is non-denumerable. Combine with the finding that a function of several complex variables is pathologically rare to be reducible. By flipping the zero-clusters, there is very small probability of finding more than one structures which satisfy all the ten constraints. It is very probable that in multidimensions, the solution of the phase problem is unique.

2.1. One-dimensional complex object

(A) One-dimensional nonperiodic complex object

For the real part $f_r(x)$, constraints used:

(i) finite size from $x = 0$ to $x = L_r$. $f_r(x) = 0$ for $x < 0$ or $x > L_r$. Value of L_r will be determined

(ii) $f_r(x)$ is non-negative, $f_r(x) \geq 0$

(iii) $f_r(x)$ is bounded

(iv) $f_r(x)$ is sectionally continuous

(v) sharp boundary. $f_r(x) > 0$ at the two boundary points,

with similar constraints for the imaginary part $f_i(x)$.

The Fourier transform of $f(x)$,

$$F(\omega) = \int_0^L f(x)\exp(-j\omega x)dx$$

where L is the larger of L_r, L_i.

Follow (A) of article II. Use Hilbert transform to obtain the phase of the minimum-phase function $H_M(\omega)$ from the magnitude $|H_M(\omega)| = |F(\omega)|$. Flip the zeros of $H_{II}(s)$. Note that the zeros need not be symmetrical with respect to the real axis. Hence zeros, not zero-pairs are flipped. Find the structure $f(x)$ with its real and imaginary parts each satisfying the five constraints. As in (A) of article II, the solution is probably unique. Thus the structure $f(x)$ including values of L_r and L_i for the real and imaginary parts is determined. If the object is continuous, this will be shown in the result.

(B) One-dimensional periodic complex object (for example, crystal)

Choose the interval $[0, L]$ as the basic unit cell for both the real and imaginary parts.

For the real part $f_{r0}(x)$, constraints used:

(i) $f_{r0}(x)$ has finite size with size = the known unit cell constant, L (tight support, a strong constraint)

(ii) $f_{r0}(x)$ is non-negative

(iii) $f_{r0}(x)$ is bounded

(iv) $f_{r0}(x)$ is sectionally continuous

(v) sharp boundary. $f_{r0}(x) > 0$ at the two boundary points, 0 and L,

with similar constraints for the imaginary part $f_{i0}(x)$.

Use interpolated in-between | Fourier coefficients | (or mid-Bragg intensities). Let $F_0(\omega)$ be the Fourier transform of $f_0(x)$. Follow (B) of article II. Discrete Hilbert transform and Shannon's sampling are used to obtain the phase of the minimum-phase function $H_M(\omega)$ from the magnitude $\left|H_M(\omega)\right| = \left|F_0(\omega)\right|$. Flip the zeros of $H_{II}(s)$. Find the structure $f_0(x)$ with its real and imaginary parts each satisfying the five constraints. If needed, adjust the interpolated in-between | Fourier coefficients | . The solution $f_0(x)$ is very probably unique for each set of | Fourier coefficients | and in-between | Fourier coefficients | . Thus the structure $f_0(x)$ is determined.

If $f(x)$ is known to be continuous, the interpolated values of in-between | Fourier coefficients | are adjusted to satisfy the constraint:

(vi) $f_0(x)$ is continuous

$f_0(0) = f_0(L)$

2.2. Multidimensional complex object

Assume that $f(x_1, x_2) \neq f_1(x_1) f_2(x_2)$ and
$f(x_1, x_2, x_3) \neq f_1(x_1) f_2(x_2) f_3(x_3)$.

2.2.1. General multidimensional complex object

(C) Multidimensional nonperiodic complex object.

For the real part $f_r(x_1, x_2)$, constraints used:

(i) finite size. $f_r(x_1, x_2) = 0$ for $x_1 < 0$, $x_1 > L_r$, $x_2 < H_r(x_1)$ or $x_2 > K_r(x_1)$. Values of L_r, $H_r(x_1)$ and $K_r(x_1)$ will be determined.

(ii) $f_r(x_1, x_2)$ is non-negative, $f_r(x_1, x_2) \geq 0$

(iii) $f_r(x_1, x_2)$ is bounded

(iv) $f_r(x_1, x_2)$ is sectionally continuous

(v) sharp boundary. $f_r(x_1, x_2) > 0$ almost everywhere at the boundary line,

with similar constraints for the imaginary part $f_i(x_1, x_2)$.

The Fourier transform of $f(x_1, x_2)$,

$$F(\omega_1, \omega_2) = \int_0^L \left\{ \int_{H(x_1)}^{K(x_1)} f(x_1, x_2) \exp(-j\omega_2 x_2) dx_2 \right\} \exp(-j\omega_1 x_1) dx_1$$

where L is the larger of L_r, L_i, $H(x_1)$ the smaller of $H_r(x_1)$, $H_i(x_1)$,

and $K(x_1)$ the larger of $K_r(x_1)$, $K_i(x_1)$.

For a fixed $\omega_2 = \omega_2'$, this is a one-dimensional problem with the term in curly brackets $q(x_1, \omega_2')$ regarded as a complex object. As in (C) of article II, flipping a finite or denumerable number of zeros does not change the length of the object. In particular, the length of the object given by the inverse Fourier transform of the minimum-phase function is the length of the object. Value of L is found.

Find the structure $f(x_1, x_2)$ with its real and imaginary parts each satisfying the five constraints. The solution is very probably unique. We have to test the possible structures in order to find the structure satisfying the five constraints. Hence the structure $f(x_1, x_2)$ including values of L_r, $H_r(x_1)$, $K_r(x_1)$, L_i, $H_i(x_1)$ and $K_i(x_1)$ for the real and imaginary parts is determined. As the number of possible solutions is very large, much computation is needed.

Similar treatment for the three-dimensional case with constraints (v) sharp boundary, $f_r(x_1, x_2, x_3) > 0$ and $f_i(x_1, x_2, x_3) > 0$ almost everywhere at the boundary surfaces. It is very probable that the solution is unique. If the object is continuous or has symmetry elements, these will be shown in the result.

(D) Multidimensional periodic complex object (for example, crystal)

Choose the cell, $0 \le x_1 \le a$, $0 \le x_2 \le b$ as the basic unit cell for both the real and imaginary parts. For the real part $f_{r0}(x_1, x_2)$, constraints used:

(i) $f_{r0}(x_1, x_2)$ has finite size with size = the known unit cell constants, a, b, α (tight support, a strong constraint)

(ii) $f_{r0}(x_1, x_2)$ is non-negative, $f_{r0}(x_1, x_2) \ge 0$

(iii) $f_{r0}(x_1, x_2)$ is bounded

(iv) $f_{r0}(x_1, x_2)$ is sectionally continuous

(v) sharp boundary. $f_{r0}(x_1, x_2) > 0$ almost everywhere at the boundary parallelogram,

with similar constraints for the imaginary part $f_{i0}(x_1, x_2)$.

Let $\omega_{10} = 2\pi / a$, $\omega_{20} = 2\pi / b$. $|F_0(l\omega_{10}, m\omega_{20})|$ are measured experimentally.

$$F_0(l\omega_{10}, m\omega_{20}) = \int_0^a \left\{ \int_0^b f_0(x_1, x_2) \exp(-jm\omega_{20} x_2) dx_2 \right\} \exp(-jl\omega_{10} x_1) dx_1$$

Consider a fixed m'. This is a one-dimensional problem with the term in curly brackets $q(x_1, m'\omega_{20})$ regarded as a complex object. Obtain $|F_0((l+1/2)\omega_{10}, m'\omega_{20})|$, $l = 0, \pm 1, \pm 2., ...$ by interpolation in multidimensions or by experimental measurement (Yuen (2011d) for a crystal). The oversampling ratio is 2. As in (D) of article II, find the structure $f_0(x_1, x_2)$ with its real and imaginary parts each satisfying the five constraints. The solution is very probably unique.

Similar treatment for the three-dimensional case with constraints (v) sharp boundary,

$f_{r0}(x_1, x_2, x_3) > 0$ and $f_{i0}(x_1, x_2, x_3) > 0$ almost everywhere at the boundary surfaces. It is very probable that the solution is unique.

In X-ray crystallography, for continuous electron density, the interpolated | Fourier coefficients | are adjusted to satisfy:

(vi) $f_0(x_1, x_2, x_3)$ is continuous with

$$f_0(0, x_2, x_3) = f_0(a, x_2, x_3) \quad \text{for all } x_2,\ x_3$$

$$f_0(x_1, 0, x_3) = f_0(x_1, b, x_3) \quad \text{for all } x_1,\ x_3$$

$$f_0(x_1, x_2, 0) = f_0(x_1, x_2, c) \quad \text{for all } x_1,\ x_2$$

(vii) symmetry elements, if applicable

2.2.2. Complex fibre

(E) General complex fibre

Fibre has circular symmetry in the x_1x_2 plane with radius $R_r(x_3)$ for the real part and $R_i(x_3)$ for the imaginary part. It is periodic in x_3 with period c for both the real and imaginary parts. Choose the cell $(x_1{}^2 + x_2{}^2)^{1/2} \le R_r(x_3)$, $0 \le x_3 \le c$, as the basic unit cell for the real part, and the cell $(x_1{}^2 + x_2{}^2)^{1/2} \le R_i(x_3)$, $0 \le x_3 \le c$, as the basic unit cell for the imaginary part.

For the real part, constraints used:

(i) For each x_3, $f_{r0}(x_1, x_2, x_3)$ has finite radius $R_r(x_3)$ (value to be determined) in the x_1x_2 plane. $f_{r0}(x_1, x_2, x_3) = 0$ for $x_3 < 0$ or $x_3 >$ c. Value of c is known.

(ii) $f_{r0}(x_1, x_2, x_3)$ is non-negative, $f_{r0}(x_1, x_2, x_3) \geq 0$

(iii) $f_{r0}(x_1, x_2, x_3)$ is bounded

(iv) $f_{r0}(x_1, x_2, x_3)$ is sectionally continuous

(v) sharp boundary. $f_{r0}(x_1, x_2, x_3) > 0$ almost everywhere at the two boundary circles at $x_3 = 0$ and c, and at the boundary surface of revolution of the real part about the x_3 axis,

with similar constraints for the imaginary part $f_{i0}(x_1, x_2, x_3)$.

The weak diffraction intensity is collected into strong diffraction planes which can be measured accurately. Let $\omega_0 = 2\pi / c$. For all l, $l = 0, \pm 1, \pm 2., \ldots$, $|F_0(\omega_1, \omega_2, l\omega_0)|$ are measured experimentally. It has circular symmetry in the $\omega_1 \omega_2$ plane.

$$F_0(\omega_1, \omega_2, l\omega_0)$$

$$= \int_0^c \left\{ \iint f_0(x_1, x_2, x_3) \exp[-j(\omega_1 x_1 + \omega_2 x_2)] dx_1 dx_2 \right\} \exp(-jl\omega_0 x_3) dx_3$$

where the double integral is over a circle for a given x_3 with radius the larger of $R_r(x_3)$, $R_i(x_3)$.

For fixed ω_1' and $\omega_2 = 0$, this is a one-dimensional problem with the term in curly brackets $q(\omega_1', 0, x_3)$ regarded as a complex object. Obtain $|F_0(\omega_1', 0, (l + (1/2))\omega_0)|$, $l = 0, \pm 1, \pm 2., \ldots$ by interpolation in multidimensions.

Follow (E) of II. Find the structure $f_0(x_1, x_2, x_3)$ with its real and imaginary parts each satisfying the five constraints. The solution is very probably unique. Thus the structure $f(x_1, x_2, x_3)$ including $R_r(x_3)$ and $R_i(x_3)$ is determined. For continuous $f(x_1, x_2, x_3)$, the interpolated values are adjusted to satisfy: $f_0(x_1, x_2, x_3)$ is continuous and $f_0(x_1, x_2, 0) = f_0(x_1, x_2, c)$ for all x_1, x_2, and symmetry elements.

(F) Fixed complex fibre molecule (a one-dimensional periodic object).

Choose the cell $H_r(x_2,x_3) \le x_1 \le K_r(x_2,x_3)$, $L_r(x_3) \le x_2 \le M_r(x_3)$, $0 \le x_3 \le c$, as the basic unit cell for the real part, and the cell $H_i(x_2,x_3) \le x_1 \le K_i(x_2,x_3)$, $L_i(x_3) \le x_2 \le M_i(x_3)$, $0 \le x_3 \le c$, as the basic unit cell for the imaginary part .

For the real part, constraints used:

(i) $f_{r0}(x_1,x_2,x_3)$ has finite size. Values of $H_r(x_2,x_3)$, $K_r(x_2,x_3)$, $L_r(x_3)$ and $M_r(x_3)$ will be determined.

$f_{r0}(x_1,x_2,x_3) = 0$ for $x_3 < 0$ or $x_3 > c$. Value of c is known.

(ii) $f_{r0}(x_1,x_2,x_3)$ is non-negative, $f_{r0}(x_1,x_2,x_3) \ge 0$

(iii) $f_{r0}(x_1,x_2,x_3)$ is bounded

(iv) $f_{r0}(x_1,x_2,x_3)$ is sectionally continuous

(v) sharp boundary. $f_{r0}(x_1,x_2,x_3) > 0$ almost everywhere at the two boundary planes at $x_3 = 0$ and c, and at the boundary surface of the real part of the fibre,

with similar constraints for the imaginary part $f_{i0}(x_1,x_2,x_3)$.

The weak diffraction intensity is collected into strong diffraction planes which can be measured accurately. Let $\omega_0 = 2\pi / c$. For all l, $l = 0,\pm1,\pm2.,\ldots$, $|F_0(\omega_1,\omega_2,l\omega_0)|$ are measured experimentally.

$F_0(\omega_1,\omega_2,l\omega_0)$

$$= \int_0^c \left\{ \iint f_0(x_1, x_2, x_3) \exp[-j(\omega_1 x_1 + \omega_2 x_2)] dx_1 dx_2 \right\} \exp(-jl\omega_0 x_3) dx_3$$

where the triple integral is over the union of the basic unit cells for the real part and the imaginary part.

For fixed ω_1' and ω_2', this is a one-dimensional problem with the term in curly brackets $q(\omega_1', \omega_2', x_3)$ regarded as a complex object. Obtain $|F_0(\omega_1', \omega_2', (l + (1/2))\omega_0)|$, $l = 0, \pm 1, \pm 2., ...$ by interpolation in multidimensions.. Follow (F) of II. Find the structure $f_0(x_1, x_2, x_3)$ with its real and imaginary parts each satisfying the five constraints. The solution is very probably unique. Thus the structure $f(x_1, x_2, x_3)$ including $H_r(x_2, x_3)$, $K_r(x_2, x_3)$, $L_r(x_3)$, $M_r(x_3)$, $H_i(x_2, x_3)$, $K_i(x_2, x_3)$, $L_i(x_3)$ and $M_i(x_3)$ is determined. If applicable, the interpolated values are adjusted to satisfy: $f_0(x_1, x_2, x_3)$ is continuous and $f_0(x_1, x_2, 0) = f_0(x_1, x_2, c)$ for all x_1, x_2, and symmetry elements.

2.2.3. Complex membrane

The membrane may be an open surface or a closed surface. The thickness of the membrane may or may not be uniform.

(G) Nonperiodic complex membrane

For the real part $f_r(x_1, x_2, x_3)$, constraints used:

(i) finite size. The shape, dimensions and thickness of the real part of the membrane will be determined.

(ii) $f_r(x_1, x_2, x_3)$ is non-negative, $f_r(x_1, x_2, x_3) \geq 0$

(iii) $f_r(x_1, x_2, x_3)$ is bounded

(iv) $f_r(x_1, x_2, x_3)$ is sectionally continuous

(v) sharp boundary. $f_r(x_1, x_2, x_3) > 0$ almost everywhere at the two surfaces of the real part of the membrane, and at the edge if the membrane is an open surface, with similar constraints for the imaginary part $f_i(x_1, x_2, x_3)$.

$$F_0(\omega_1, \omega_2, \omega_3) =$$

$$\int_0^L \left\{ \int_{H(x_1)}^{K(x_1)} \left[\int_{P(x_1,x_2)}^{Q(x_1,x_2)} f(x_1, x_2, x_3) \exp(-j\omega_3 x_3) dx_3 \right] \exp(-j\omega_2 x_2) dx_2 \right\} \exp(-j\omega_1 x_1) dx_1$$

Follow the method in (C). If $f(x_1, x_2, x_3) \neq f_1(x_1, x_2) f_3(x_3)$, it is very probable that the solution is unique. The shape, dimensions and thickness of the real and imaginary parts of the membrane are determined. If $f(x_1, x_2, x_3)$ is continuous or has symmetry elements, these will be shown in the result.

Special cases:

(a) Circular complex membrane, $f(x_1, x_2, x_3) = f_1(x_1, x_2) f_3(x_3)$, where $f_1(x_1, x_2)$ is circularly symmetric.

$$F_0(\omega_1, \omega_2, \omega_3)$$

$$= \iint f_1(x_1, x_2) \exp[-j(\omega_1 x_1 + \omega_2 x_2)] dx_1 dx_2 \int f_3(x_3) \exp(-j\omega_3 x_3) dx_3$$

It is very probable that the solution for $f_1(x_1, x_2)$ is unique (see (C)). The solution for $f_3(x_3)$ is very probably unique (see (A)).

(b) Thin complex membrane, the thickness of the membrane approaches 0. $f(x_1, x_2, x_3)$ becomes a function of two variables on the surface. It is very probable that the solution is unique (see (C)).

(H) Planar periodic complex membrane (a two-dimensional periodic object)

The object is periodic in x_1 and x_2 with periods a and b respectively for both the real and imaginary parts. Choose the cell $0 \le x_1 \le a$, $0 \le x_2 \le b$, $H_r(x_1, x_2) \le x_3 \le K_r(x_1, x_2)$ as the basic unit cell for the real part, and the cell

$0 \le x_1 \le a$, $0 \le x_2 \le b$, $H_i(x_1,x_2) \le x_3 \le K_i(x_1,x_2)$ as the basic unit cell for the imaginary part.

For the real part, constraints used:

(i) $f_{r0}(x_1,x_2,x_3)$ has finite size with known unit cell constants, a,b,α. $f_{r0}(x_1,x_2,x_3)=0$ for $x_3 < H_r(x_1,x_2)$ or $x_3 > K_r(x_1,x_2)$. Values of $H_r(x_1,x_2)$ and $K_r(x_1,x_2)$ will be determined.

(ii) $f_{r0}(x_1,x_2,x_3)$ is non-negative, $f_{r0}(x_1,x_2,x_3) \ge 0$

(iii) $f_{r0}(x_1,x_2,x_3)$ is bounded

(iv) $f_{r0}(x_1,x_2,x_3)$ is sectionally continuous

(v) sharp boundary. $f_{r0}(x_1,x_2,x_3) > 0$ almost everywhere at the six boundary surfaces of the basic unit cell of the real part,

with similar constraints for the imaginary part $f_{i0}(x_1,x_2,x_3)$.

The weak diffraction intensity is collected into strong diffraction rods (Bragg rods) which can be measured accurately. Let $\omega_{10} = 2\pi/a$, $\omega_{20} = 2\pi/b$. For all l and m, $\left|F_0(l\omega_{10}, m\omega_{20}, \omega_3)\right|$ are measured experimentally.

$$F_0(l\omega_{10}, m\omega_{20}, \omega_3)$$

$$= \int_0^a \int_0^b \left\{ \int_{H(x_1,x_2)}^{K(x_1,x_2)} f_0(x_1,x_2,x_3)\exp(-j\omega_3 x_3)dx_3 \right\} \exp\left[-j(l\omega_{10}x_1 + m\omega_{20}x_2)\right]dx_1 dx_2$$

where $H(x_1,x_2)$ is the smaller of $H_r(x_1,x_2)$, $H_i(x_1,x_2)$, and $K(x_1,x_2)$ the larger of $K_r(x_1,x_2)$, $K_i(x_1,x_2)$.

For fixed $\omega_3 = \omega_3'$, this is a two-dimensional case with the term in curly brackets $q(x_1,x_2,\omega_3')$ regarded as a complex object. For example, $a > b$. Obtain $\left|F_0((l+1/2)\omega_{10}, m\omega_{20}, \omega_3')\right|$, $l = 0,\pm1,\pm2.,...$, $m = 0,\pm1,\pm2.,...$ by interpolation in multidimensions. Follow (H) of II. Find the structure $f_0(x_1,x_2,x_3)$

with its real and imaginary parts each satisfying the five constraints. The solution is very probably unique. Thus the structure $f(x_1, x_2, x_3)$ including $H_r(x_1, x_2)$, $K_r(x_1, x_2)$, $H_i(x_1, x_2)$ and $K_i(x_1, x_2)$ is determined. If applicable, the interpolated values are adjusted to satisfy:

(vi) $f_0(x_1, x_2, x_3)$ is continuous

$$f_0(0, x_2, x_3) = f_0(a, x_2, x_3) \quad \text{for all } x_2,\ x_3$$

$$f_0(x_1, 0, x_3) = f_0(x_1, b, x_3) \quad \text{for all } x_1,\ x_3$$

(vii) symmetry elements

2.2.4. Complex macromolecule

For nonperiodic and periodic complex macromolecule ((C) to (H)), it is very probable that the structure obtained is unique. As in I and II, the method in above is very suitable for determination of structures of periodic macromolecules, for example, protein crystals and biological fibres.

2.3. Finite $|\omega|$ and combination with iterative algorithm

As mentioned in Yuen (2011c), Sect. 2.3, in practice, we use a finite $\omega_{\max}$ corresponding to the resolution needed. The phase-retrieval method is applied with smaller $\omega_{\max}$ to determine a gross structure of the object. The zeros are arranged in ascending order according to their distances from the origin. Then we move alternately the zeros to the right-hand plane to satisfy the constraints and obtain a gross structure. We then extend the value of $\omega_{\max}$, deal with more zeros, and obtain more refined structures. Alternatively, we may use a narrow strip centered at the imaginary axis with length given by the experimentally attained $\omega_{\max}$, so that it contains all the zeros of the experimentally observed $|F(\boldsymbol{\omega})|$ (Wackerman & Yagle (1994)). The width of the strip can then be increased. In all cases, $|\boldsymbol{\omega}| \leq$ the experimentally attained $\omega_{\max}$.

Much empirical success has been attained by means of iterative algorithms and oversampling by many researchers. Various values of oversampling ratio are employed. For periodic object, a unit cell is usually, or can be chosen, to have sharp boundary. Hence if we use oversampling ratio=2, and the constraints in this article, including the strong constraints of finite size, non-negativity and sharp boundary for the real and imaginary parts, it is very probable that the solution of the phase problem is unique. This provides a theoretical basis that iterative algorithms with oversampling ratio=2 may lead to the correct structure for nonperiodic or periodic complex object. If iterative algorithm is used, it fits in the tail, $F(\boldsymbol{\omega})$, $\omega_{\max} < |\boldsymbol{\omega}|$ by extrapolation.

As a combination of the phase-retrieval method and iterative algorithm, the phase obtained for the gross structure can be used as the initial phase of the iterative algorithm. This is better than the random initial phase commonly employed. The gross structure is also used as initial input to the iterative process. This contains more information than just the known support of the object. These initial inputs may help in avoiding stagnation with faster convergence to the global minimum, the correct solution. Stagnation may be due to the possibility of the iterative process to proceed towards the object or its twin (Fienup & Wackerman, 1986, p. 1897), the insensitivity of the Fourier modulus versus a translational operation on an object, the support not tight enough or the boundary not sharp enough (He, 2006, p. 554).

3. Discussion

For the phase-retrieval method or combined with iterative algorithm (X-ray diffraction microscopy), algorithms applicable to real data need to be developed. Constraints used in this article will be applied. For periodic object, the oversampling ratio is 2, applied to the longest unit cell edge. Because of noise in the experimental data, more refined sampling with higher sampling ratio will be more effective for the iterative process to converge to the global minimum. The authors may apply the phase retrieval method, the iterative algorithms or their combinations to re-determine or refine their structures in the literature. It is very probable that the structure obtained is unique. Electron density is determined solely from diffraction intensity. Absolute configuration is determined. Electron density determines the active site of the molecule. Absolute configuration of a biological macromolecule is vital to its biological activity. It is important to determine the structure of a macromolecule in solution because its configuration may be different from that in crystal.

For multidimensional nonperiodic object (C), support of the complex object need not be known, as long as it is finite. There is no restriction on the shape of the support. It can be very smooth and highly symmetrical. For example, the object can be spherically symmetric. This is contrary to the belief that special types of support are needed for reconstruction of complex objects while simple symmetric supports do not work well. The support will be determined. As a very large number of possible structures are obtained. We have to test the structures and find the structure which satisfies all the five constraints.

In one, two and three dimensions ((B), (D), (E), (F) and (H)), the oversampling ratio is exactly 2.

The method is very suitable for determination of structures of periodic macromolecules, for example, protein crystals and biological fibres.

The phase-retrieval method presented uses flipping of zeros. It will be very useful if an expression without using zero-flipping can be obtained. An additional constraint may be included: The size (shape and dimensions), and the regions of zeros (not isolated points) of the real part and the imaginary part are the same. This is a strong constraint. For a crystal (D), same basic unit cell is used for both the real part and the imaginary part. A mathematical proof that the ten or eleven constraints employed indeed lead to unique solution is needed. If such proof can be established, the constraints of sharp boundary, non-negativity, same size and same regions of zeros of the real and imaginary parts can be disregarded successively. Their individual effect to the uniqueness of the solution can be examined. Projection method may be employed (Youla, 1987; Levi & Stark, 1987; Stark & Yang (1998); Weierstall et al., 2002; Elser, 2003a, 2003b; Elser et al., 2007). Ten or eleven constraints are used for complex object while five constraints are used for real object (article II). Intuitively we may think that it is more probable for uniqueness of solution for complex object. In fact, the system may be over-determined. The computer modeling of Miao & Sayre (2000), p. 598 ‘seems to show that enforcing the positivity constraints on both parts is not as effective as enforcing them on either the real or the imaginary part’; non-negativity constraint is applied on either the real part or the imaginary part. In constraint (iv), the real or imaginary parts of the object may be continuous. It will be very useful to determine the different sets of minimum number of constraints which will lead to unique solution. It is mentioned at the beginning of Sect. 2 that the non-negativity or non-positivity constraint on either the real part or imaginary part must be employed. The above argument suggests that it is more probable to obtain a unique solution for the phase problem of complex object with non-negative real and imaginary parts, than for real non-negative object. This is contrary to the belief that phase retrieval is more difficult for complex object than for real object. If the constraint of sharp boundary can be disregarded, replicas of a nonperiodic object can be added and a three-dimensional object is obtained. Yuen (2011b) for iterative algorithms with oversampling can then be applied. If the object is centrosymmetric, there is one more constraint. The system is more over-determined. For a three-dimensional continuous periodic object satisfying all these constraints plus, if applicable, other symmetry elements, it remains to investigate the restriction of these constraints upon the interpolated values (Palmer, 1994). Method of projection may be applied.

Use the non-positivity constraint on the imaginary part of the object (Sect. 2). The phase-retrieval method, the iterative algorithm or their combinations can be applied in optical scattering to determine the relative complex dielectric constant (see (1)), which is very probably unique. The constraints in Yuen (2011a, 2011c) and this article may also be employed for objects for which weak scattering (the first Born approximation) cannot be applied (Crocco et al., 2004).

4. Conclusions

As a conclusion, the phase of the wave diffracted by a multidimensional complex object with non-negative real and imaginary parts may not be lost; it is just hidden in the magnitude. With general and reasonable constraints on the object, it is very probable that the solution of the phase problem is unique. A phase-retrieval method is presented to recover the phase from the magnitude. On this basis, for nonperiodic or periodic complex object, iterative algorithms with oversampling ratio =2 may lead to the correct structure. The phase-retrieval method is very suitable for determination of structures of periodic macromolecules, for example, protein crystals and biological fibres. For the phase-retrieval method or combined with iterative algorithm, researchers may develop algorithms applicable to real data. They may also find an expression for the solution. The authors may apply the phase retrieval method, the iterative algorithms or their combinations to re-determine or refine their nonperiodic and periodic structures in the literature.

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12

Ab initio determination of electron densities of non-centrosymmetric non-negative or complex crystals from diffraction intensities in X-ray crystallography. Iterative algorithms with oversampling

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Abstract

For non-centrosymmetric non-negative or complex crystals, the phase of the diffracted wave can be retrieved from the magnitudes. The result is very probably unique. Hence the electron density can be obtained directly from the diffraction intensities in X-ray crystallography. The oversampling ratio is exactly 2. Values of in-between | Fourier coefficients | for one index l, m or n is sufficient for determination of the electron density. Values of the in-between | Fourier coefficients | for the three indices l, m and n are not independent. They must be consistent so that the electron densities obtained for each index are identical. This consistent condition must be satisfied. The process of phase retrieval is time-consuming. However, it provides the theoretical foundation and gives the conditions that iterative algorithms with oversampling will give a probably unique solution. The in-between Fourier coefficients calculated from crystal structure reported in the literature are reasonably good. For crystal structure reported in the literature (R-index $\neq 0$), iterative algorithms with oversampling can be used to refine the electron density. The iterative algorithms will converge to a unique *ab initio* non-negative or complex electron density with R-index = 0. This is an improvement of the electron density reported in the literature. For crystal structure which has not been solved, its electron density with R-index = 0 can also be determined by means of iterative algorithms with oversampling. The *ab initio* electron density may be unique. It is determined by the experimental lattice constants, diffraction intensities and initial in-between | Fourier coefficients | (or mid-Bragg intensities); no other chemical or physical information (for example, atomicity, chemical formula, bond length, bond angle, etc) is used. From the refined electron density with R-index = 0, atomic coordinates are obtained which is an improvement of the atomic coordinates obtained by conventional methods in X-ray crystallography.

If some of the light atoms do not appear, or some bond lengths and bond angles are different from those obtained by other means, we can use chemical or physical informations, including the atomic coordinates and electron density determined in conventional X-ray crystallography to improve the in-between｜Fourier coefficients｜ and/or the｜Fourier coefficients｜ within their uncertainties to obtain all the atoms and reasonable bond lengths and bond angles. Iterative algorithms with oversampling will be a very useful and important tool in crystal structure analysis.

Keywords: Phase retrieval; consistent condition; iterative algorithms; oversampling; in-between｜Fourier coefficients｜; mid-Bragg intensities; refinement of electron density; improvement of｜Fourier coefficients｜; macromolecule; protein; polypeptide; DNA.

1. Introduction

This is a sequel of Yuen (2011a, 2011d. 2011e).

For crystal structure determination, direct methods is used in Dorset (2000, 2001) and reviewed in Usón & Sheldrick (1999). Direct methods and applications to electron crystallography are reviewed in Giacovazzo et al. (2002). Use of anomalous X-ray diffraction in protein crystallography is reviewed in Kahn et al. (2000). Maximum-likelihood refinement is used in solving a protein structure in Tame (2000). Experimental phasing is presented in Read (2003). Phase determination by entropy maximization and likelihood ranking is reviewed in Bricogne (1993). Bayesian statistical viewpoint on structure determination is reviewed in Bricogne (1997a, 1997b, 1997c). Group-subgroup relation is used in Di Contanzo et al. (2003). Webster & Hilgenfeld (2001) presents an evolutionary computational approach to the phase problem in macromolecular X-ray crystallography. Kolinski & Skolnick (1997) discusses determinants of secondary structure of polypeptide chains. Ortiz et al. (1998) presents studies of fold assembly of small proteins. Protein structure alignment is presented in Blankenbecler et al. (2003). Thumiger & Zanotti (2009) presents a review of iterative methods. Specific radiation damage is used in Ravelli et al. (2003). *Ab initio* phasing starting from low resolution is presented in Lunin et al. (2007). Ockwell et al. (2000) and Hao (2001) present phasing from an envelope. Hong & Hao (2009) determinates molecular envelope and heavy-atom sites by combining solution wide-angle X-ray scattering and crystallography. Cooper et al. (2010) uses a multiplayer online game to predict protein structures. Serial nanocrystallography method is presented in Hunter et al. (2011) and Kirian et al. (2011). Phasing methods for protein crystallography are reviewed in Hauptman (1997) and Taylor (2003).

Real space refinement for proteins is presented in Diamond (1971, 1985). Refinement of protein structure is presented in Qian et al. (2007). Role of stereochemical restraints is presented in Jaskolski et al. (2007). Quantum refinement is reviewed in Ryde & Nilsson (2003). Scheres & Gros (2001) presents conditional optimization in protein structure refinement.

Keel et al. (2007) presents a general strategy to solve the phase problem in RNA crystallography. A general method for phasing complex RNA crystal structures is presented in Robertson & Scott (2008).

A real-space approach to the phase problem is presented in Su (2008). Low-density elimination method is used in Matsugaki & Shiono (2001). Macromolecular crystallography is discussed in the review of Jaskolski (2010).

Structural proteomics are reviewed in Liu & Hsu (2005).

Diffraction properties of periodic lattices under free electron laser radiation is presented in Rajkovic et al. (2010).

Binary superposed phase retrieval problem is presented in Alpers et al. (2010).

For non-centrosymmetric and non-negative, and complex crystals, by imposing some general and reasonable constraints on the objects, the phase of the diffracted wave can be retrieved from the magnitude (Sect. 2.1). The solution is very probably unique.

The in-between Fourier coefficients calculated from crystal structure reported in the literature is more reliable than individual value of the electron density. They are reasonably good. Magnitudes of these in-between Fourier coefficients can be used for phase retrieval.

Much work has been done by many researchers with iterative algorithms and oversampling, and has gained much empirical success. The process of phase retrieval is time-consuming. However, it provides the theoretical foundation and gives the conditions that iterative algorithms with oversampling will give a solution which is probably unique. For iterative algorithms and oversampling, see Pitney et al. (1999), Robinson et al. (2001), Miao et al. (2002), Weierstall et al. (2002), Miao et al. (2003), Nishino et al. (2003), Williams et al. (2003, 2004), Pfeifer et al. (2006), Williams et al. (2006), Minkevich et al. (2007), Stadler et al. (2007), Boutet & Robinson (2008), Jiang et al. (2008), Minkevich et al. (2008), Huang et al. (2009), Fleischer et al. (2010), Gulden et al. (2010), Harder et al. (2010), Newton et al. (2010), Takahashi et al. (2010), Vaxelaire et al. (2010), Chapman et al. (2011) and references in Yuen (2011a, 2011c, 2011d. 2011e, 2011f). Coherent X-ray diffraction imaging of strain is reviewed in Robinson & Harder (2009).

For a crystal structure reported in the literature, the electron density can be refined by iterative algorithms with oversampling (Sect. 2.2). The iterative algorithms will converge to a unique *ab initio* non-negative or complex electron density with R -index = 0. This is an improvement of the electron density reported in the literature. The in-between | Fourier coefficients | are also improved.

For crystal structure which has not been solved, its electron density with R -index = 0 can also be determined by means of iterative algorithms with oversampling (Sect 2.3). The *ab initio* electron density may be unique.

The electron density obtained has R -index = 0. This means that the uncertainty of the experimentally measured | Fourier coefficients | is not taken into account. If some of the light atoms do not appear, or some bond lengths and bond angles are different from those obtained by other means, we can use chemical or physical informations, including the atomic coordinates and electron density determined in conventional X-ray crystallography to improve the in-between | Fourier coefficients | and/or the | Fourier coefficients | within their uncertainties to obtain all the atoms and reasonable bond lengths and bond angles (Sect. 2.4).

This article can be applied for determination or refinement of electron density of proteins, polypeptides and DNA.

2.1 Phase-retrieval method

Similar to Yuen (2011b), Sect. 2.1.

For a crystal with electron density $f(x_1, x_2, x_3)$, we deal with the content of the basic unit cell of the crystal as an object, and define $f_0(x_1, x_2, x_3) = f(x_1, x_2, x_3)$

for (x_1, x_2, x_3) ε the basic unit cell, or = 0 otherwise. Choose the cell, $0 \le x_1 \le a$, $0 \le x_2 \le b, 0 \le x_2 \le c$ as the basic unit cell. The phase of the diffraction points will be retrieved from the magnitude by imposing some general and reasonable constraints upon the object.

For the real part $f_{r0}(x_1, x_2, x_3)$ of a complex crystal, use the constraints (Yuen (2011e), (D)):

(i) $f_{r0}(x_1, x_2, x_3)$ has finite size with size = the known unit cell constants, $a, b, c, \alpha, \beta, \gamma$

(ii) $f_{r0}(x_1, x_2, x_3) \ge 0$

(iii) $f_{r0}(x_1, x_2, x_3)$ is bounded

(iv) sharp boundary. The component function $f_{r0}(x_1, x_2, x_3) \ne 0$ almost everywhere at the six boundary surfaces of the three-dimensional unit cell (or four boundary lines of the two-dimensional unit cell for planar crystal)

(v) $f_{r0}(x_1, x_2, x_3)$ is continuous with

$$f_{r0}(0, x_2, x_3) = f_{r0}(a, x_2, x_3) \quad \text{for all } x_2,\ x_3$$

$$f_{r0}(x_1, 0, x_3) = f_{r0}(x_1, b, x_3) \quad \text{for all } x_1,\ x_3$$

$$f_{r0}(x_1, x_2, 0) = f_{r0}(x_1, x_2, c) \quad \text{for all } x_1,\ x_2$$

(vi) the crystal may have other symmetry elements,

with similar constraints for the imaginary part $f_{i0}(x_1, x_2, x_3)$.

(vii) the size (shape and dimensions), and the regions of zeros (not isolated points) of the real part and the imaginary part are the same. This is a strong constraint.

For a non-centrosymmetric non-negative crystal, use constraints (i) – (vi) (Yuen (2011d), (D)).

According to Yuen (2011d, 2011e), (D), the solution of the phase-retrieval method is very probably unique. For a three-dimensional crystal, because of the constraint of sharp boundary and the regular shape of a unit cell with eight sharp corners and twelve sharp edges, it is very probably that the solution is unique. Millane (1996), p.726 states that one might suspect that in going from two dimensions to three, the phase problem my go from being unique to being “more than unique”.

The | Fourier coefficients | are obtained from the experimentally measured intensity. As the | Fourier coefficients | are used in obtaining the electron density, the R -index = 0.

Values of in-between | Fourier coefficients | for one index l, m or n (oversampling ratio = 2) is sufficient for determination of the unique electron density. Values of the in-between | Fourier coefficients | for the three indices l, m and n are not independent. They must be consistent so that the electron densities obtained for each index are identical (Yuen (2011b), Sect. 2.1). This consistent condition must be satisfied.

2.2. Iterative algorithms with oversampling. Refinement of electron density of crystal structure reported in the literature (R -index $\neq 0$)

Similar to Yuen (2011b), Sect. 2.2, 2.3.

The phase-retrieval method in Sect. 2.1 provides the theoretical foundation and gives the conditions that iterative algorithms with oversampling ratio =2 will lead to the electron density which is very probably unique. As the process in Sect. 2.1 is time-consuming, iterative algorithms with oversampling may be employed.

As shown in Yuen (2011b), Sect. 2.2, the in-between Fourier coefficients calculated from the electron density reported in the literature are reasonably good. For a crystal structure reported in the literature, calculate the Fourier coefficients and in-between Fourier coefficients from the electron density in the basic unit cell. Use their phases as initial phases.

Hence we have an important conclusion of this article:

Use

1. the experimentally determined unit cell constants and | Fourier coefficients | ,
2. initial in-between | Fourier coefficients | obtained from the electron density reported in the literature with R -index $\neq 0$,

3. the initial phases,
4. | Fourier coefficients | and initial in-between | Fourier coefficients | for all (l,m,n), and all ($l+\frac{1}{2}$, and/or $m+\frac{1}{2}$, and/or $n+\frac{1}{2}$). The oversampling ratio is $2^3 = 8$.
5. the six constraints for non-centrosymmetric non-negative crystal or thirteen constraints for complex crystal,
6. in the final iterations, the in-between | Fourier coefficients | are relaxed or adjusted, within their uncertainties, to satisfy the consistent condition; the in-between | Fourier coefficients | are improved.

The iterative algorithms will converge to a non-negative or complex electron density with R -index = 0. The *ab initio* electron density is unique for this set of | Fourier coefficients | and initial in-between | Fourier coefficients | . This is an improvement of the electron density reported in the literature.

Crystal structures determined by conventional methods in X-ray crystallography provide accurate atomic ordinates. The electron density is less accurate. Since the structure has been refined by conventional methods in X-ray crystallography, the electron density should not be very different from the true electron density. Using the electron density reported in the literature as the initial electron density, it is reasonable to expect that the iterations will converge to a unique and improved electron density.

As explained in Yuen (2011b), Sect. 2.2, the initial in-between | Fourier coefficients | obtained from the reported electron density is more reliable than individual value of the electron density. Using these initial in-between | Fourier coefficients | , the electron density obtained by iterative algorithms with oversampling may show some light atoms which are missed in the electron density reported in the literature. From the refined electron density with R -index = 0, atomic coordinates are obtained which is an improvement of the atomic coordinates reported.

2.3. Iterative algorithms with oversampling. Determination of electron density of crystal structure which has not been solved

Similar to Yuen (2011b), Sect. 2.4.

For a crystal structure which has not been solved, obtain the initial in-between $|$ Fourier coefficients $|$ by interpolation in multidimensions or by experimental measurement (Yuen (2011g)). Follow Sect 2.2. We use random initial phases which are commonly employed. The phase-retrieval method in Sect. 2.1 shows that oversampling ratio =2 leads to a very probably unique *ab initio* electron density. To compromise that proper initial phases are not used, more oversampling can be employed. We use $|$ Fourier coefficients $|$ and initial in-between $|$ Fourier coefficients $|$ for all (l,m,n), and all ($l+\frac{1}{2}$, and/or $m+\frac{1}{2}$, and/or $n+\frac{1}{2}$). The oversampling ratio $=2^3=8$. Iteration algorithms in Sect. 2.2 will then be applied. The iterations will converge to an electron density with R-index = 0. The electron density may be unique. This is because of the known unit cell constants which is a tight support, and a very strong and powerful constraint, in particular for three-dimensional crystal. This convergence can be investigated empirically and theoretically.

As experience accumulates, we will learn how much oversampling is needed for the fastest and most efficient convergence of the iterations.

From the refined electron density with R-index = 0, atomic coordinates are obtained which is an improvement of the atomic coordinates obtained by conventional methods in X-ray crystallography. The in-between $|$ Fourier coefficients $|$ are also improved because of the consistent condition.

2.4. Recovery of missing atoms and improvement of bond lengths and bond angles

Use Yuen (2011b), Sect. 2.5.

The electron density obtained in Sect. 2.2 or 2.3 has R-index = 0. This means that the uncertainty of the experimentally measured intensity (or $|$ Fourier coefficients $|$) is not taken into account. The experimentally measured $|$ Fourier coefficients $|$ do have uncertainties. The uncertainties of the in-between $|$ Fourier coefficients $|$ may be larger than those of the $|$ Fourier coefficients $|$. If some of the light atoms (for example, hydrogen atom) do not appear, or some bond lengths and bond angles are different from those obtained by other means (for example, neutron diffraction, spectroscopy), and if we cannot find any good reason, such discrepancies may be due to uncertainties of the in-between | Fourier coefficients | and/or the | Fourier coefficients | . Hence we can use chemical or physical informations, including the atomic coordinates and electron density determined in conventional X-ray crystallography to improve first the in-between | Fourier coefficients | , then the | Fourier coefficients | . They are adjusted within their uncertainties to obtain all the atoms and reasonable bond lengths and bond angles.

3. Discussions and conclusion

The phase-retrieval method in Sect. 2.1 provides the theoretical foundation and gives the conditions that iterative algorithms with oversampling will give a unique solution.

Hence, an important conclusion is obtained:

For refinement of electron density of crystal structure reported in the literature R-index $\neq 0$), if we follow the process in Sect. 2.2, **the iterative algorithms will converge to a non-negative or complex electron density with R-index = 0. The *ab initio* electron density is unique for this set of | Fourier coefficients | and initial in-between | Fourier coefficients | . This is an improvement of the electron density reported in the literature.** The electron density obtained by iterative algorithms with oversampling may show some light atoms which are missed in the electron density reported in the literature.

The in-between Fourier coefficients calculated from crystal structure reported in the literature are reasonably good. We urge that crystallographers will apply iterative algorithms with oversampling (Sect. 2.2) to refine the electron density of structures of the many non-centrosymmetric non-negative or complex crystals reported in the literature, in particular, those crystal structures solved by themselves. Improved in-between | Fourier coefficients | and improved electron density with R-index = 0 can be obtained. The *ab initio* electron density is determined by the experimental lattice constants and diffraction intensities alone; no other chemical or physical information (for example, atomicity, chemical formula, bond length, bond angle, etc) is used. This is different from conventional X-ray crystallography, where atomicity, atomic scattering factor and chemical formula are used in structure determination. From the electron density with R-index = 0, the atoms, their species and coordinates can be determined. The atomic coordinates obtained is an improvement of the atomic coordinates obtained by conventional methods in X-ray crystallography. The chemical formula can then be obtained. This serves as an experimental method of obtaining the atomic species and the chemical formula, and an experimental verification that atomicity holds in crystals.

For forthcoming articles in X-ray crystallography, after the electron densities have been refined by conventional methods, iterative algorithms with oversampling (Yuen (2011b), Sect. 2.3, and Sect. 2.2 of this article) may be applied to further refine, and to obtain the *ab initio* electron densities with R-index = 0. The *ab initio* electron densities will be very useful in studies of distribution of electrons in crystals, and chemical reactions, in particular, biochemical reactions. May be this can be adopted as a standard procedure.

As we have obtained an electron density with R-index = 0, we have made full use of the｜Fourier coefficients｜. Hence it is meaningful to have Sect. 2. 4: Recovery of missing atoms and improvement of bond lengths and bond angles. If any light atom is missed, or some bond lengths and bond angles are different from those obtained by other means (for example, neutron diffraction, spectroscopy), such discrepancies may be due to uncertainties of the in-between｜Fourier coefficients｜and/or the｜Fourier coefficients｜. These discrepancies are corrected by adjusting first the in-between ｜Fourier coefficients｜, then the｜Fourier coefficients｜, within their uncertainities.

For determining the electron density of crystals, the structures of which have not been solved by conventional methods in X-ray crystallography, random initial phases may be adopted. To compromise that proper initial phases are not used, more oversampling can be employed by using all the initial in-between｜Fourier coefficients｜. The oversampling ratio is $2^3 = 8$. No chemical information is used. The iterations may converge to a unique *ab initio* electron density because of the known unit cell constants, a very strong and powerful constraint. This convergence can be investigated empirically and theoretically. Yuen (2011a), Appendix shows that for crystals of large unit cells, the in-between | Fourier coefficients | obtained by interpolation is more accurate. In contrary to conventional methods in X-ray crystallography, the phase-retrieval method and the iterative algorithms with oversampling is very suitable for crystals of macromolecules, for example, proteins.

Oversampling ratio = 2 is sufficient for determination of the unique electron density. Values of the in-between | Fourier coefficients | are not independent. They must be consistent so that the electron densities obtained for each index are identical. In the final iterations, the in-between | Fourier coefficients | are relaxed or adjusted, within their uncertainties, to satisfy this consistent condition. The in-between | Fourier coefficients | are improved.

Crystallographers who have reported preliminary X-ray diffraction studies of non-centrosymmetric non-negative crystals or complex crystals in the literature may apply Sect. 2.3 to determine the electron density of their crystals.

For a complex object, as mentioned in Yuen (2011e), Sect. 3, the system may be over-determined. From the converged complex electron density, we drop the non-negativity constraint (ii) for the real or imaginary part or both. Iterative algorithms can then be applied. If we get the same result, this shows that this part of the electron density is indeed non-negative.

As a conclusion, it is reasonable to believe that iterative algorithms with oversampling will be a very useful and important tool in the determination of crystal electron density (see also Yuen (2011h)).

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13

Direct determination of length of an one-dimensional object, or diameter of an object with shape of a circle, a cylinder, a sphere or an axisymmetric object, and the thickness of a membrane by means of the minimum-phase function

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This article presents a method for direct determination of the length of an one-dimensional object, or diameter of an object with shape of a circle, a cylinder, a sphere or an axisymmetric object, and the maximum thickness of a layer by means of the minimum-phase function. It applies to a real or complex, centrosymmetric or non-centrosymmetric object.

Keywords: Inverse problem; scattering; diffraction; light scattering; entire function of exponential type; Hilbert transform; discrete Hilbert transform; flipping of zeros; flipping of zero-pairs; diffraction rods; Bragg rods.

1. Introduction

This is a sequel article of Yuen (2011c, 2011d). It applies to a real or complex, centrosymmetric or non-centrosymmetric object.

This article presents a method for direct determination of the length of an one-dimensional object, or diameter of an object with shape of a circle, a cylinder, a sphere or an axisymmetric object, and the maximum thickness of a membrane by means of the minimum-phase function. The diameter of an object with shape of an axisymmetric object refers to the maximum diameter. The object may or may not be homogeneous, or continuous. For objects with the shape of a circle, cylinder or axisymmetric, it may or may not be circularly or cylindrically symmetric. For object with the shape of a sphere, it may or may not be spherically symmetric. The result is independent of the material of the object. Only the external dimensions are obtained. These are independent of the internal structure of the object. This article does not apply to a conductor at frequencies with small depth of penetration.

In Yuen (2011a, 2011c, 2011d), the structure of the object is determined. This involves analytic continuation, locating and flipping of zeros. Much computation is needed. However, if only the length of an object, diameter of an object with shape of a circle, a cylinder, a sphere or an axisymmetric object, and the maximum thickness of a membrane is needed, the computation can be much reduced.

The inverse problem of determining the length of an object, the diameter of a circle, a sphere or a fibre, and the thickness of a membrane is very important in many disciplines: meteorology, ice-thickness, geology, membranes and films in industry, atoms and molecules in chemistry and biology, radii of nuclei and elementary particles, etc. This is a fundamental problem in, for example, light scattering. Much research has been done. For examples:

For scattering from spheres, see Kerker et al. (1978), Roumeliotis et al. (1995), Yi & Stafsudd (1998), Siu & Cheng (2002) and Li et al. (2005). Inverse scattering is presented in Brovko et al. (2008) and Tsitas & Athanasiadis (2008). Luo et al. (2007) uses Fourier transform of light scattering spectrum for size determination. Gerchberg-Saxton method is employed in Dilanian & Nikulin (2005). Kozak (2007) presents structural investigation of biological systems such as radius of gyration and maximum size by means of small-angle X-ray scattering. X-ray diffraction is employed in Kurlov & Gusev (2007). Evaporative light-scattering detection method is presented in Emmenegger et al. (2007). Haiss et al. (2007) presents size and concentration determination from UV-vis spectra. Terahertz spectroscopy is presented in Bandyopadhyay et al. (2007). Extinction spectra are employed in Khlebtsov (2008). Hollow spheres are investigated in Huang et al. (2009).

For scattering from cylinders, see (Wait (1955), Roumeliotis & Kakogiannos (1994), Stiles & Sarabandi (1996), Ye (1997), Brancaccio et al. (1998), Stanton et al. (1998), Bernabeu et al. (1999), Pierri & Leone (1999), Polat (1999), Wear & Harris (2008)). Phase/Doppler light-scattering is employed in Schaub et al. (1998) and occlusion of a laser beam in Butler & Forbes (1998). For inverse scattering, see Budko & van den Berg (1998) and Agarwal & Chen (2008)). For axisymmetric body, see Reeder & Stanton (2004). For application of diffraction-shadow technique to sphere and cylinder, see Rinkevichyus et al. (2006).

Books and reviews in scattering are provided by Kerker (1969), van de Hulst (1981), Jones (1999), Nieto-Vesperinas (2006), and Kokhanovsky (2006, 2007, 2008).

For electron diffraction of molecules, see references in Yuen (2011b).

For optical diffraction pattern analysis of filament, see Belczyk et al. (2006). For scattering curve and radius of gyration of a straight chain of identical spheres, see Kawaguchi (2001). For diffraction from a layer of spheres see Mills et al. (2008). Retinal vessel diameter is measured in Joos et al (1997). Size and embedding depth of blood vessels are determined in Bykov et al. (2007).

For determination of layer and film thickness, see Jablonski & Zemek (2009) and You et al. (2009)). For Langmuir-Blodgett film thickness, see Ifuku at al. (2005). For lipid bilayer thickness, see Kučerka et al. (2004) and Chen et al. (2009). For determination of lubricant layer thickness, see Hoshino & Kimachi (1996) For foam thickness, see Raišutis et al. (2008). For thermal boundary layer thickness, see Ashjaee & Mahmoudi (2007). For insulation thickness, see Cheng (1994). For asphalt pavement layer thickness, see Choubane et al. (2003).

Cloud thickness is determined in Cahalan et al. (2005) and Lamquin et al. (2008). For cloud drop size distribution, see Freud et al. (2008), and raindrop size distribution, see Smith et al. (2009). Radius of aerosol particle is determined in Stothers (2001). Aerosol size distribution is measured in Bäumer et al. (2008). For hailstone size distribution, see Sánchez et al. (2009). Venkataraman et al. (2007) measures snow grain size. Snow and sea ice thickness is determined in Galley et al. (2009).

For estimation of crustal thickness, see Maden et al. (2009). For mantle transition zone thickness, see Lawrence & Shearer (2008). For core radius, see Hales & Roberts

(1970). For 3D depth imaging, see Nestvold et al. (1996). For petroleum depth, see Ejedawe & Okoh (1981). Dippenaar et al. (2005) presents a review of the determination of light non-aqueous phase liquid plume thickness. For coal-seam thickness, see Gang & Goulty (1997). For ore thickness, see Murphy et al. (2005).

For nuclear radius, see Angeli (2004) and Mueller et al. (2007). For proton and neutron radii, see Sick (2005).

2. Method

(A) One-dimensional nonperiodic object $f(x)$.

Unknown real or complex $f(x)$.

Constraints used:

(i) finite size from $x = 0$ to $x = L$. $f(x) = 0$ for $x < 0$ or $x > L$. The purpose is to determine the value of L.

(ii) $f(x)$ is bounded

The diffracted wave is given by the Fourier transform of $f(x)$,

$$F(\omega) = \int_0^L f(x)\exp(-i\omega x)dx \tag{1}$$

$$F(\omega) = |F(\omega)|\exp[i\phi(\omega)].$$

Bilateral Laplace transform of $f(x)$,

$$F_{II}(s) = \int_0^L f(x)\exp(-sx)dx \tag{2}$$

$F_{II}(s)$ is an entire function of exponential type, order 1 and type L, but not of type less than L. It has an infinite number of zeros. Let γ_1, γ_2,… be the zeros of $F_{II}(s)$ not at the origin. From Hadamard's factorization theorem,

$$F_{II}(s) = s^{\alpha}\left[\prod_{n=1}^{\infty}\left(1 - \frac{s}{\gamma_n}\right)\exp(s/\gamma_n)\right]\exp(a + bs) \tag{3}$$

where α is an integer ≥ 0 for the multiplicity of the origin as a zero of $F_{II}(s)$,

a and b are real or complex constants

Use treatment similar to that in Yuen(2011c), Sect. 2.1. Zeros for complex object, or zero-pairs for real object are flipped. The minimum-phase function

$$H_M(\omega) = |H_M(\omega)| \exp[i\phi_M(\omega)].$$

$|H_M(\omega)| = |F(\omega)|$ which is obtained from the experimentally measured intensity.

The phase $\phi_M(\omega)$ is uniquely determined from $\ln|H_M(\omega)| = \ln|F(\omega)|, -\infty < \omega < \infty$,

$$\phi_M(\omega) = -\frac{1}{\pi} P \int_{-\infty}^{\infty} \frac{\ln|F(y)|}{\omega - y} dy \tag{4}$$

This is a Hilbert transform. P stands for Cauchy principal value. Using $\phi_M(\omega)$, we get the minimum-phase function $H_M(\omega)$.

The length of the object is not changed in flipping of zeros. Let $h(x)$ be the inverse Fourier transform of $H_M(\omega)$. The length of the object $h(x)$ is equal to the length L of the object $f(x)$.

Hence the length L of object $f(x)$ is obtained from the minimum-phase function. Note that locations of the zeros of $F_{II}(s)$ and flipping of zeros or zero-pairs are not performed explicitly.

Remark 1. Although the length of the object is not changed in flipping of zeros, in one distribution of the zeros, the object obtained may consist of two disconnected intervals, while in another distribution of the zeros, the null interval of the object may change in length, or disappear so that the object obtained is connected. Hence the treatment presented can only determine the total length of the object, not the internal null interval. $f(x)$ is also not determined.

(B) Treating an one-dimensional nonperiodic object as a crystal

Instead of (A), to facilitate the computations and reduce the number of measurements, use a sampling method. Size of the object is unknown. Use a sufficiently large L to contain the object. Then mathematically add its replica successively to give a periodic object. Instead of (4), discrete Hilbert transform gives

$$\phi_M(l\omega_{1/2}) = -\frac{2}{\pi}\sum \frac{\log|F(m\omega_{1/2})|}{l-m} \tag{5}$$

where $l = 0,\pm1,\pm2,\ldots$, $\omega_{1/2} = 2\pi/(2L) = \pi/L$, and summation over m , $l-m$ odd.

With $\phi_M(l\omega_{1/2})$ obtained from (5), we get $H_M(l\omega_{1/2})$. Then, for $-\infty < \omega < \infty$, Shannon's sampling theory gives the minimum-phase function

$$H_M(\omega) = \sum_{l=-\infty}^{\infty} H_M(l\omega_{1/2}) \frac{\sin(L\omega - l\pi)}{L\omega - l\pi} \tag{6}$$

Let $h(x)$ be the inverse Fourier transform of $H_M(\omega)$. As in (A), the length of the object $h(x)$ is the length of the object.

(C) A two-dimensional object with shape of a circle

Unknown $f(x_1, x_2)$.

Constraints used:

(i) finite size with radius R. The purpose is to determine the value of R.

(ii) $f(x_1, x_2)$ is bounded

$f(x_1, x_2)$ may or may not be circularly symmetric.

$$F(\omega_1, \omega_2) = \iint f(x_1, x_2)\exp[-i(\omega_1 x_1 + \omega_2 x_2)]dx_1 dx_2$$

where the double integral is over the area of the object.

$$F(\omega_1, \omega_2) = \int_{-R}^{R} \left\{\int f(x_1, x_2)\exp(-i\omega_2 x_2)dx_2\right\}\exp(-i\omega_1 x_1)dx_1$$

Choose $\omega_2 = 0$.

$$F(\omega_1,0)=\int_{-R}^{R}\left\{\int f(x_1,x_2)dx_2\right\}\exp(-i\omega_1 x_1)dx_1 \tag{7}$$

The term in the curly brackets is a function of x_1 representing an one-dimensional object with size from $x_1=-R$ to $x_1=R$. Follow (A) or (B), the value of R is determined.

(D) A two-dimensional object with shape of a rectangle

Unknown $f(x_1,x_2)$.

Constraints Used:

(i) finite size with edge lengths a and b. The purpose is to determine the values of a and b.

(ii) $f(x_1,x_2)$ is bounded

$$F(\omega_1,\omega_2)=\int_0^a\int_0^b f(x_1,x_2)\exp[-i(\omega_1 x_1+\omega_2 x_2)]dx_1 dx_2$$

$$=\int_0^a\left\{\int_0^b f(x_1,x_2)\exp(-i\omega_2 x_2)dx_2\right\}\exp(-i\omega_1 x_1)dx_1$$

Choose $\omega_2=0$.

$$F(\omega_1,0)=\int_0^a\left\{\int_0^b f(x_1,x_2)dx_2\right\}\exp(-i\omega_1 x_1)dx_1 \tag{8}$$

The term in the curly brackets is a function of x_1 representing an one-dimensional object with size from $x_1=0$ to $x_1=a$. Follow (A) or (B), the value of a is determined. Similarly, the value of b is determined.

(E) A two-dimensional object with shape of a cylinder

Unknown $f(x_1,x_2,x_3)$.

Constraints used:

(i) finite size with radius R and length L. The purpose is to determine the values of R and L. The axis of the cylinder is along the x_3-axis.

For each x_3, $f(x_1, x_2, x_3)$ has the shape of a circle with radius R. $f(x_1, x_2, x_3)$ may or may not be circularly symmetric.

(ii) $f(x_1, x_2, x_3)$ is bounded

$$F(\omega_1, \omega_2, \omega_3) = \iiint f(x_1, x_2, x_3) \exp[-i(\omega_1 x_1 + \omega_2 x_2 + \omega_3 x_3)] dx_1 dx_2 dx_3$$

where the triple integral is over the volume of the object.

$$F(\omega_1, \omega_2, \omega_3)$$

$$= \int_{-R}^{-R} \left\{ \int \left[\int_0^L f(x_1, x_2, x_3) \exp(-i\omega_3 x_3) dx_3 \right] \exp(-i\omega_2 x_2) dx_2 \right\} \exp(-i\omega_1 x_1) dx_1$$

Choose $\omega_2 = \omega_3 = 0$.

$$F(\omega_1, 0, 0) = \int_{-R}^{-R} \left\{ \int \left[\int_0^L f(x_1, x_2, x_3) dx_3 \right] dx_2 \right\} \exp(-i\omega_1 x_1) dx_1 \tag{9}$$

The term in the curly brackets is a function of x_1 representing an one-dimensional object with size from $x_1 = -R$ to $x_1 = R$. Follow (A) or (B), the value of R is determined.

To determine the value of L, let

$$F(\omega_1, \omega_2, \omega_3)$$

$$= \int_0^L \left\{ \int \left[\int_{-R}^{R} f(x_1, x_2, x_3) \exp(-i\omega_1 x_1) dx_1 \right] \exp(-i\omega_2 x_2) dx_2 \right\} \exp(-i\omega_3 x_3) dx_3$$

Choose $\omega_1 = \omega_2 = 0$.

$$F(0, 0, \omega_3) = \int_0^L \left\{ \int \left[\int_{-R}^{R} f(x_1, x_2, x_3) dx_1 \right] dx_2 \right\} \exp(-i\omega_3 x_3) dx_3 \tag{10}$$

The term in the curly brackets is a function of x_3 representing an one-dimensional object with size from $x_3 = 0$ to $x_3 = L$. Follow (A) or (B), the value of L is determined.

Remark 2. $\boldsymbol{\omega}$ = wavevector of the scattered wave – wavevector of the incident wave. Hence, for object with shape of a cylinder, as long as $\boldsymbol{\omega}$ is in a direction normal to the cylinder, it is immaterial whether the incident wave is normal or oblique to the cylinder.

(F) An object with shape of a sphere

Unknown $f(x_1, x_2, x_3)$.

Constraints used:

(i) finite size with radius R. The purpose is to determine the value of R.
$f(x_1, x_2, x_3)$ may or may not be spherically symmetric.

(ii) $f(x_1, x_2, x_3)$ is bounded

$$F(\omega_1, \omega_2, \omega_3) = \iiint f(x_1, x_2, x_3) \exp[-i(\omega_1 x_1 + \omega_2 x_2 + \omega_3 x_3)] dx_1 dx_2 dx_3$$

where the triple integral is over the volume of the object.

$$F(\omega_1, \omega_2, \omega_3)$$

$$= \int_{-R}^{R} \left\{ \iint f(x_1, x_2, x_3) \exp[-i(\omega_2 x_2 + \omega_3 x_3)] dx_2 dx_3 \right\} \exp(-i\omega_1 x_1) dx_1$$

Choose $\omega_2 = \omega_3 = 0$.

$$F(\omega_1, 0, 0) = \int_{-R}^{R} \left\{ \iint f(x_1, x_2, x_3) dx_2 dx_3 \right\} \exp(-i\omega_1 x_1) dx_1 \qquad (11)$$

The term in the curly brackets is a function of x_1 representing an one-dimensional object with size from $x_1 = -R$ to $x_1 = R$. Follow (A) or (B), the value of R is determined.

(G) Maximum diameter and length of a non-periodic object with axisymmetric

shape

Unknown $f(x_1, x_2, x_3)$.

Constraints used:

(i) For each x_3, the object $f(x_1, x_2, x_3)$ has finite radius $R(x_3)$ in the x_1x_2 plane. Length of the object is finite. The purpose is to determine the maximum diameter and the length L. The axis of the object is along the x_3-axis.

For each x_3, $f(x_1, x_2, x_3)$ may or may not has circular symmetry in the x_1x_2 plane.

(ii) $f(x_1, x_2, x_3)$ is bounded

$$F(\omega_1, \omega_2, \omega_3)$$

$$= \int\int\left\{\int_0^L f(x_1, x_2, x_3)\exp(-i\omega_3 x_3)dx_3\right\}\exp[-i(\omega_1 x_1 + \omega_2 x_2)]dx_1 dx_2$$

where the triple integral is over the volume of the object. Choose $\omega_3 = 0$.

$$F(\omega_1, \omega_2, 0) = \int\int\left\{\int_0^L f(x_1, x_2, x_3)dx_3\right\}\exp[-i(\omega_1 x_1 + \omega_2 x_2)]dx_1 dx_2 \tag{12}$$

The term in curly brackets is a two-dimensional object with the shape of a circle in the x_1x_2 plane. Its diameter is the maximum diameter of the object. Follow (C), the value of this maximum diameter is determined.

To determine the length L, let

$$F(\omega_1, \omega_2, \omega_3)$$

$$= \int_0^L\left\{\int\int f(x_1, x_2, x_3)\exp[-i(\omega_1 x_1 + \omega_2 x_2)]dx_1 dx_2\right\}\exp(-i\omega_3 x_3)dx_3$$

Choose $\omega_1 = \omega_2 = 0$.

$$F(0,0,\omega_3) = \int_0^L \left\{ \iint f(x_1, x_2, x_3) dx_1 dx_2 \right\} \exp(-i\omega_3 x_3) dx_3 \tag{13}$$

The term in the curly brackets is a function of x_3 representing an one-dimensional object with size from $x_3 = 0$ to $x_3 = L$. Follow (A) or (B), the value of L is determined.

Similar to (E), as long as $\boldsymbol{\omega}$ is in a direction normal to the axis of the object with axisymmetric shape, it is immaterial whether the incident wave is normal or oblique to the axis.

(H) Maximum diameter of an one-dimensional periodic object with axisymmetric shape

The unknown object is periodic in x_3 with period c. Value of the period c is known; it can be determined as in X-ray crystallography. For each x_3, the object may or may not has circular symmetry in the $x_1 x_2$ plane. The purpose is to determine the maximum diameter. $f_0(x_1, x_2, x_3)$ is one period of the object.

Constraints used:

(i) For each x_3, $f_0(x_1, x_2, x_3)$ has finite radius $R(x_3)$ in the $x_1 x_2$ plane.

(ii) $f_0(x_1, x_2, x_3)$ is bounded

Because of the one-dimensional periodic structure, the diffracted intensity is greatly enhanced to form strong diffraction planes. Let $\omega_0 = 2\pi / c$.

$$F_0(\omega_1, \omega_2, n\omega_0)$$

$$= \iint \left\{ \int_0^c f_0(x_1, x_2, x_3) \exp(-in\omega_0 x_3) dx_3 \right\} \exp[-i(\omega_1 x_1 + \omega_2 x_2)] dx_1 dx_2$$

where the triple integral is over the volume of one period of the object.

Choose $n = 0$.

$$F_0(\omega_1,\omega_2,0)=\int\int\left\{\int_0^c f_0(x_1,x_2,x_3)dx_3\right\}\exp\left[-i\left(\omega_1x_1+\omega_2x_2\right)\right]dx_1dx_2 \tag{14}$$

The term in curly brackets is a two-dimensional object with the shape of a circle in the x_1x_2 plane. Its diameter is the maximum diameter of the object. Follow (C), the value of this maximum diameter is determined.

Similar to (E), as long as $\boldsymbol{\omega}$ is in a direction normal to the axis of the object with axisymmetric shape, it is immaterial whether the incident wave is normal or oblique to the axis.

(I) Maximum thickness of a planar non-periodic membrane, film or layer

Consider a membrane in the x_1,x_2 plane. Constraints used:

(i) the membrane $f(x_1,x_2,x_3)$ has finite size. The purpose is to determine the maximum thickness of the membrane.

(ii) $f(x_1,x_2,x_3)$ is bounded

$$F(\omega_1,\omega_2,\omega_3)$$

$$=\int\left\{\int\int f(x_1,x_2,x_3)\exp[-i(\omega_1x_1+\omega_2x_2)]dx_1dx_2\right\}\exp(-i\omega_3x_3)dx_3$$

where the triple integral is over the volume of the membrane.

Choose $\omega_1=\omega_2=0$.

$$F(0,0,\omega_3)=\int\left\{\int\int f(x_1,x_2,x_3)dx_1dx_2\right\}\exp(-i\omega_3x_3)dx_3 \tag{15}$$

The term in the curly brackets is an one-dimensional object in x_3. Its length is the maximum thickness of the membrane. Follow (A), the value of this maximum thickness is determined.

Similar to (E), as long as $\boldsymbol{\omega}$ is in a direction normal to the membrane, it is immaterial whether the incident wave is normal or oblique to the membrane.

(J) Maximum thickness of a planar two-dimensional periodic membrane, film or

layer

The membrane is periodic in x_1 and x_2 with periods a and b respectively. $f_0(x_1, x_2, x_3)$ is one period of the membrane.

Constraints used:

(i) $f_0(x_1, x_2, x_3)$ has finite size with known unit cell constants, a, b, α, which can be determined as in X-ray crystallography. The purpose is to determine the maximum thickness of the membrane.

(ii) $f_0(x_1, x_2, x_3)$ is bounded.

Because of the two-dimensional periodic structure, the diffracted intensity is greatly enhanced to form strong diffraction rods (Bragg rods). Let $\omega_{10} = 2\pi / a$, $\omega_{20} = 2\pi / b$.

$$F_0(l\omega_{10}, m\omega_{20}, \omega_3)$$

$$= \int \left\{ \iint f_0(x_1, x_2, x_3) \exp[-i(l\omega_{10} x_1 + m\omega_{20} x_2)] dx_1 dx_2 \right\} \exp(-i\omega_3 x_3) dx_3$$

where the triple integral is over the volume of one period of the membrane.

Choose $l = m = 0$.

$$F_0(0,0,\omega_3) = \int \left\{ \iint f_0(x_1, x_2, x_3) dx_1 dx_2 \right\} \exp(-i\omega_3 x_3) dx_3 \tag{16}$$

The term in the curly brackets is an one-dimensional object in x_3. Its length is the maximum thickness of the membrane. Follow (A), the value of this maximum thickness is determined.

Similar to (E), as long as $\boldsymbol{\omega}$ is in a direction normal to the membrane, it is immaterial whether the incident wave is normal or oblique to the membrane.

Remark 3. In (1), (7) - (16), if we use $|F(\boldsymbol{\omega})|$ or $|F_0(\boldsymbol{\omega})|$ for all $|\boldsymbol{\omega}| < \infty$, the inverse Fourier transform gives a structure which is exact, corresponding to ultimate accuracy. This is not needed in practice. In length measurement in daily life, we do not need accuracy at, say, angstroms. Depending on the accuracy needed, we use $|F(\boldsymbol{\omega})|$

or $|F_0(\boldsymbol{\omega})|$ for $|\boldsymbol{\omega}| \le \omega_{\max}$, where $\omega_{\max}$ is sufficiently large (Yuen (2011c), Sect. 2.3).

Remark 4. As the functions $f(x)$, $f(x_1, x_2)$, $f(x_1, x_2, x_3)$ or $f_0(x_1, x_2, x_3)$ are not obtained, the internal structure of the object, including cavities, are not obtained. Only the external dimensions, diameter, length and thickness, are obtained. This also means that the dimensions obtained are independent of the material of the object.

3. Discussions and conclusion

A method is presented for direct determination of the external dimensions, diameter, length and thickness of an one-dimensional object, or objects with shapes of a circle, cylinder, sphere, axisymmetric object and membrane. The object may or may not be homogeneous, or continuous. For objects with the shape of a circle, cylinder or axisymmetric object, it may or may not be circularly or cylindrically symmetric. For object with the shape of a sphere, it may or may not be spherically symmetric. The result is independent of the material of the object. Only the external dimensions are obtained. These are independent of the internal structure of the object. The incident wave may be normal or oblique to a cylinder or a membrane. In (H) and (J) for one-dimensional and two-dimensional periodic structures, the diffracted intensity is greatly enhanced into strong diffraction planes or diffraction rods. These facilitate the measurement of intensities.

Unlike Yuen (2011a, 2011c, 2011d), analytical continuation of $H_M(\omega)$, location of zeros and flipping of zeros for complex object, or zero-pairs for real object, are not used. The amount of computation is much reduced. This is because, in the treatment presented, only the length and the radii are obtained. The functions $f(x)$, $f(x_1, x_2)$ and $f(x_1, x_2, x_3)$ are not obtained. Hence the radius of gyration is not obtained. If the objects have null interval or internal empty cavities, they are not obtained. To obtain these informations, the treatment in Yuen (2011a, 2011c, 2011d) can be used.

Constraint (i) of finite size is used. As mentioned in Yuen (2011b), it is very interesting to find out whether finite radii of gas atom or rotating molecule is obtained. This is different from quantum chemistry which states that the electron densities of atoms and molecules extend to infinity. It is more interesting if sharp boundary of atom or molecule is found.

It is interesting if the treatment presented can be extended to obtaining the size distribution or a mixture of gases.

This article only presents the basic principle of measuring the maximum thickness of a membrane. For the many applications in meteorology and geology, much research is needed.

In this book, we try to extend the phase-retrieval method in daily scale and atomic scale to nuclear scale and elementary-particle scale. Much research is needed. The results can be compared with results from other theoretical or experimental methods. If there is any discrepancy, we can seek for modifications and improvements. These provide more understanding of nuclear physics and elementary-particle physics. Since nuclear physics and elementary-particle physics are very important and complicate, every approach is invaluable and may be attempted. The phase-retrieval approach is direct, efficient and quantitative.

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14

Direct determination of the maximum length of a real or complex object along any direction, and the reconstruction of the shape (and dimensions) of the convex hull (envelope) of the object

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This article presents a method for direct determination of the maximum length of a real or complex object along any direction, and for reconstruction of the shape of the convex hull (envelope) of the object from the set of maximum lengths.

Keywords: Inverse problem; inverse scattering; convex object; concave object; multi-object; convex hull; minimum-phase function; central slice theorem; Fourier projection theorem; projection-slice theorem; Radon transform; tomography.

1. Introduction

This is a sequel of Yuen (2011). The term 'shape' used in this article includes dimensions. If an object is represented by $f(x_1, x_2, x_3)$, the shape of the object refers to the boundary of the union of the support of $f(x_1, x_2, x_3)$ and, if any, internal cavities.

For a three-dimensional object, and any direction, use a straight line along this direction. Project the three-dimensional object on this line. We are interested in the length of this projection, and call it the maximum length of the object along this direction. Similar definition is used for a two-dimensional object.

Following Yuen (2011) with the use of the minimum-phase function, the maximum length of an object along any direction is determined. Using the central slice theorem, the shape of the convex hull (envelope) of the object (may be a multi-object) is reconstructed. The shape of the convex hull of a two-dimensional or three-dimensional centrosymmetric object can be reconstructed graphically.

Reconstruction of the shape of the convex hull of an object is dealt with, not internal cavity. The boundary may have sharp corners.

This article does not apply to a conductor at frequencies with small depth of penetration.

Determination of the shape of an object is very important in many disciplines. Much research has been done in the determination of the shape of an object, for examples:

For inferring phase information from modulus information in two-dimensional aperture synthesis, Napier & Bates (1974) have used the one-dimensional projections of a two-dimensional object upon lines of arbitrarily small angular increments. Fiddy (1987), p. 503 envisages that the asymptotic behavior of the zeros from the one-dimensional Fourier transforms may reflect the support shape. Fienup et al. (1982) reconstructs the support of an object from the support of its autocorrelation.

Hoenders (1978) discusses the uniqueness of inverse problems. Inverse problems in imaging are presented in the book, Bertero & Boccacci (2002).

For inverse scattering, see Colton & Kirsch (1996), Ikehata (1998), Potthast (2000), Belkebir & Tijhuis (2001), Luke & Potthast (2003), Cakoni & Colton (2005), the review van den Berg (2000), and the book Cakoni & Colton (2006).

For shape determination, see Colton (1988), Xu (1996), Belkebir et al. (1997), Rozier et al. (1997), Kirsch (1998), Abubakar et al. (2000), Buchanan et al. (2000), Brancaccio et al. (2002), Liseno & Pierri (2003), Liu et al. (2003), Kusyi & Voitovich (2005), Liang et al. (2005) and Yaman & Şimşek (2009).

Convex scattering support is presented in Kusiak & Sylvester (2003) and Potthast et al. (2003). Campos-García & Díaz-Uribe (2005) presents error analysis in evaluation of the shape of a surface.

Radon transform and tomography are presented it the books, Markoe (2006) and Deans (2007).

For nuclear deformation, see Nazarewicz & Ragnarsson (1996). An oblate ellipsoid is presented in Erlykin & Wolfendale (2004)). Nuclear shape is presented in the book, Nilsson & Ragnarsson (1995).

Chacón et al. (1998) presents low-resolution structures of proteins in solution retrieved from X-ray scattering. Koch et al. (2003) presents studies of structures of macromolecules in solution by means of small-angle scattering.

Medical imaging is presented in the book, Epstein (2008).

2. Method

2.1. A two-dimensional object $f(x_1, x_2)$

Locate the origin of the x_1, x_2 coordinate system at the boundary of the object.

Constraints used:

(i) finite size

(ii) $f(x_1, x_2)$ is bounded

The diffracted wave is given by the Fourier transform of $f(x_1, x_2)$,

$$F(\omega_1, \omega_2) = \iint f(x_1, x_2) \exp[-i(\omega_1 x_1 + \omega_2 x_2)] dx_1 dx_2$$

where the double integral is over the area of the object.

$$F(\omega_1, \omega_2) = \int_0^L \left\{ \int f(x_1, x_2) \exp(-i\omega_2 x_2) dx_2 \right\} \exp(-i\omega_1 x_1) dx_1$$

where L is the maximum length of the object along the x_1-direction.

Choose $\omega_2 = 0$.

$$F(\omega_1, 0) = \int_0^L \left\{ \int f(x_1, x_2) dx_2 \right\} \exp(-i\omega_1 x_1) dx_1$$

The term in the curly brackets is a function of x_1 representing an one-dimensional object with size from $x_1 = 0$ to $x_1 = L$. It is the projection of the object on the x_1-axis. Following Yuen (2011), (A) or (B), with the use of the minimum-phase function, the value of L is determined. For all orientations of the x_1, x_2 coordinate system, the maximum lengths of the object along all directions are obtained.

2.2. A three-dimensional object $f(x_1, x_2, x_3)$

Locate the origin of the x_1, x_2, x_3 coordinate system at the boundary of the object.

Constraints used:

(i) finite size

(ii) $f(x_1, x_2, x_3)$ is bounded

The diffracted wave is given by the Fourier transform of $f(x_1, x_2, x_3)$,

$$F(\omega_1, \omega_2, \omega_3)$$

$$= \int_0^L \left\{ \iint f(x_1, x_2, x_3) \exp[-i(\omega_1 x_1 + \omega_2 x_2)] dx_1 dx_2 \right\} \exp(-i\omega_1 x_1) dx_1$$

where L is the maximum length of the object along the x_1-direction.

Choose $\omega_2 = \omega_3 = 0$.

$$F(\omega_1, 0, 0) = \int_0^L \left\{ \iint f(x_1, x_2, x_3) dx_1 dx_2 \right\} \exp(-i\omega_1 x_1) dx_1$$

The term in the curly brackets is a function of x_1 representing an one-dimensional object with size from $x_1 = 0$ to $x_1 = L$. It is the projection of the object on the x_1-axis. Following Yuen (2011), (A) or (B), with the use of the minimum-phase function, the value of L is determined. For all orientations of the x_1, x_2, x_3 coordinate system, the maximum lengths of the object along all directions are obtained.

2.3. Concave object and multi-object:

The maximum length in any direction of a concave object is the same as the maximum length of its convex hull (envelope) in the same direction. Hence for a concave object or multi-object, the results obtained in this article are for its convex hull.

2.4. Reconstruction of the shape of the convex hull (envelope) of an object

The central slice theorem or Fourier projection theorem or projection-slice theorem (Fiddy (1987), p. 502, Lim (1990), Sect. 1.4.3, Nieto-Vesperinas (2006), Sect. 10.8.1) will be used.

Two-dimensional object:

The maximum lengths of the object $f(x_1, x_2)$ along all directions are given. Let $g(x_1, x_2) \equiv 1$ be an object with the same shape as the convex hull of the object $f(x_1, x_2)$. $g(x_1, x_2)$ is a characteristic function with support the convex hull of the object $f(x_1, x_2)$. Hence the maximum lengths of the object $g(x_1, x_2)$ along all directions are known. Using the central slice theorem, the object $g(x_1, x_2)$ is obtained. Hence the shape of the convex hull of the object $f(x_1, x_2)$ is determined.

The shape of the convex hull of a two-dimensional centrosymmetric object can be reconstructed graphically by using a parallel strip with width equal to the maximum length along the direction normal to the strip, and centered at the origin. The intersection of all strips for all maximum lengths has the shape of the object.

Three-dimensional object:

The maximum lengths of the object $f(x_1, x_2, x_3)$ along all directions are given. Let $g(x_1, x_2, x_3) \equiv 1$ be an object with the same shape as the convex hull of the object $f(x_1, x_2, x_3)$. $g(x_1, x_2, x_3)$ is a characteristic function with support the convex hull of the object $f(x_1, x_2, x_3)$. Hence the maximum lengths of the object $g(x_1, x_2, x_3)$ along all directions are known. Using the central slice theorem, the object $g(x_1, x_2, x_3)$ is obtained. Hence the shape of the convex hull of the object $f(x_1, x_2, x_3)$ is determined.

Alternatively,

$$F(\omega_1, \omega_2, \omega_3)$$

$$= \iint \left\{ \int f(x_1, x_2, x_3) \exp(-i\omega_3 x_3) dx_3 \right\} \exp[-i(\omega_1 x_1 + \omega_2 x_2)] dx_1 dx_2$$

Choose $\omega_3 = 0$

$$F(\omega_1, \omega_2, 0) = \iint \left\{ \int f(x_1, x_2, x_3) dx_3 \right\} \exp[-i(\omega_1 x_1 + \omega_2 x_2)] dx_1 dx_2$$

The term in curly brackets is a planar object. Use the reconstruction for two-dimensional object in above and find an object $g_p(x_1, x_2) \equiv 1$ with the shape of the convex hull of the planar object $\left\{ \int f(x_1, x_{,2}\, x_3) dx_3 \right\}$. For all orientations of the x_1, x_2, x_3 coordinate system, find the set of all objects $g_p(x_1, x_2) \equiv 1$. Use reconstruction in three dimensions by linear inversion (Allison (2006), p. 242), the object $g(x_1, x_2, x_3)$ is obtained.

The shape of the convex hull of a three-dimensional centrosymmetric object can be reconstructed graphically by using a parallel slab with thickness equal to the maximum length along the direction normal to the slab surfaces, and centered at the origin. The slab surfaces are the two-dimensional planes. The intersection of all slabs for all maximum lengths has the shape of the object.

An example of a three-dimensional object is a membrane.

3. Discussions and conclusion

In Hoenders (1978), p.58, it is mentioned that the solution of inverse problem of determining an unknown function $f(x_1, x_2, x_3)$ uses three independent parameters, while the boundary can be described by two independent parameters. It seems plausible that less data will suffice to determine the boundary. In this article, we find that the set of maximum length of a real or complex object in all directions is sufficient to determine the shape of the object. The boundary may have sharp corners. The internal content of the object, $f(x_1, x_2)$ or $f(x_1, x_2, x_3)$, is not determined.

A method is presented for direct determination of the maximum length of an object along any direction, and for reconstruction of the shape (and dimensions) of the convex hull (envelope) of an object from the set of maximum lengths.

This article can be applied to find the shape of a nucleus. A nucleus may be complex according to the optical model (Feshbach et al. (1954)).

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15

Ab initio determination of electron densities of non-centrosymmetric non-negative or complex nonperiodic objects from diffraction intensities. Iterative algorithms with oversampling

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Abstract

The phase of the diffracted waves of non-centrosymmetric non-negative or complex nonperiodic object can be retrieved from the magnitudes. The oversampling ratio is exactly 2. The solution is very probably unique. Phase retrieval method provides the theoretical foundation and gives the conditions that iterative algorithms with oversampling will give a probably unique solution. The basic idea is to use a basic unit cell which has six sharp boundary surfaces to contain the nonperiodic object. Then mathematically add replica of the unit cell successively to give a crystal. Electron densities of the crystals can then be determined from diffraction intensities by means of iterative algorithms with oversampling. In order to have a basic unit cell which has six sharp boundary surfaces, we can use a rectangular container containing the object. The shape and dimensions of the container walls form a very strong container constraint. The region of constant electron density of the solvent provides a solvent constraint. Alternatively we can embed the object in a liquid which is then solidified. For a macromolecule, protection against radiation damage is provided by the frozen solution in the container, or the solidified liquid. If a reference object is used, because of the very strong constraint of reproducing the structure of the reference object, it is most probable that the iterations will converge to the unique electron density. For objects such as astronomical objects, we have to use just the objects. Electron density of object reported in the literature can be refined by means of iterative algorithms with oversampling. The iterative algorithms probably converge to a unique, *ab initio* and improved electron density with R-index = 0. For electron density of object which has not been solved, its electron density with R-index = 0 can also be determined by means of iterative algorithms with oversampling. The *ab initio* electron density obtained may not be unique. A container, or solidified liquid or just the nonperiodic

object can be used. In all cases, the experimental ｜Fourier coefficients｜ and in-between ｜Fourier coefficients｜ are improved by means of the consistent condition. For complex object, the system may be over-determined. If the constraint of sharp boundary can be disregarded, procedures similar to determination of electron densities of centrosymmetric real nonperiodic objects by means of iterative algorithms with oversampling can be applied.

Keywords: Phase retrieval; iterative algorithms; oversampling; container constraint; solvent constraint; macromolecules; solidified liquid; radiation damage; protection; refinement of electron density.

1. Introduction

This is a sequel of Yuen (2011b, 2011c. 2011d).

Much work has been done by many researchers in the determination of the electron density of a nonperiodic object.

Dielectric properties and molecular structure are presented in Valiyev et al. (2005). Chiroptical methods are used in Julínek et al. (2009). Infrared spectra and microwave spectra are employed in Borba et al. (2009) and Okabayashi et al. (2009). Wide field single molecule microscopy is employed in Mackowiak et al. (2009). Molecular dynamics is employed in De Simone et al. (2008). Ab initio Hartree-Fock and density functional theory investigations on molecular structure are presented in Sıdır et al. (2010). Density functional theory for molecular structure calculations is reviewed in Chen & Zhou (2008). Optimization techniques is presented in Sahinidis (2009). Single-molecule imaging is presented in Schwartz et al. (2010). Reviews of single-molecule imaging are presented in Engel & Müller (2000) and Kellermayer (2005). Solution structures of proteins are presented in Goroncy et al. (2010) and Lee et al. (2010). A review of macromolecules in solution is presented in Svergun & Koch (2003). For reviews about molecular structures, see Böhm (1996), Lal & Lin (2001), Prassl & Laggner (2009), Rudd et al. (2009), Konermann et al. (2010) and Perry et al. (2010). X-Ray microdiffraction imaging of bone and tissue-engineered bone is reviewed in Guagliardi et al. (2009).

Spence et al. (2004) presents coherence and sampling requirements for diffractive imaging. Data preparation and evaluation techniques for x-ray diffraction microscopy are presented in Steinbrener et al. (2010). Requirement of empty space surrounding an object in coherent X-ray diffraction is discussed in Hajdu & Maia (2008). A reconstruction algorithm is presented in Loh & Elser (2009). A new statistically regularized amplitude constraint is presented in Dilanian et al. (2010). Kawahara et al. (2010) presents beam-divergence deconvolution for diffractive imaging. Chu et al. (2011) presents in-plane rotation classification for coherent X-ray imaging of single biomolecules. Dynamic sample imaging is presented in Clark et al. (2011). Huang et al. (2011a) presents propagation uniqueness in three-dimensional coherent diffractive imaging. Single molecule diffraction is presented in Spence & Doak (2004). Coherent x-ray diffraction imaging of silicon oxide growth is presented in Robinson et al. (1999). Chen et al. (2011) presents three-dimensional image reconstruction of radiation-sensitive samples with x-ray diffraction microscopy. Roy et al. (2011) presents lensless X-ray imaging in reflection geometry. Coherent diffractive imaging with X-rays and electrons is reviewed in Spence & Howells (2004). Falcone et al. discusses diffraction microscopy.

Femtosecond diffractive imaging is presented in Chapman et al. (2006, 2009a, 2009b), Barty et al. (2008) and Barty (2010). Song et al. (2008a) presents resonant X-ray diffraction microscopy. Song et al. (2008b) and Seibert et al. (2011) present imaging of viruses. Seibert et al. (2010) presents femtosecond diffractive imaging of biological cells. Huang et al. (2009a) presents signal-to-noise and radiation exposure considerations. Anti-contamination device for cryogenic soft X-ray diffraction microscopy is presented in Huang et al. (2011b). Dose requirements for resolving a given feature in an object by coherent X-ray diffraction imaging is presented in Schropp & Schroer (2010). Howell et al. (2009) presents an assessment of the resolution limitation due to radiation-damage in X-ray diffraction microscopy. Ponomarenko et al. (2011) investigates radiation-induced melting. Schroer et al. (2008) and Takahashi et al. (2009b) present diffraction imaging with focused illumination. Peterson et al. (2008) and Ho et al. (2009) present X-ray probe and molecular structure determination of laser-aligned molecules. Filsinger et al. (2011) presents aligned and oriented molecules for ultrafast diffraction studies. Incoherent addition of scattered intensities in single-particle diffraction is discussed in Maia et al. (2009). Pabst et al. (2010) discusses x-ray scattering from three-dimensionally- aligned asymmetric-top molecules. Wang et al. (2011) studies impact of a water layer on orientational information. Classification and averaging of random orientation single macromolecular diffraction patterns at atomic resolution is presented in Bortel et al. (2009). Shneerson et al. (2008) presents the common-line method for assembling a three-dimensional diffraction volume from single-particle scattering. Fung et al. (2009) demonstrates the recovery of the structure of a weakly scattering macromolecule. Schwander et al. (2010) deal with mapping the conformations of biological assemblies. Bogan et al. (2010) simulates coherent X-ray diffractive imaging of combustion particle (soot) morphology. Kirian et al. (2011) discusses signal, noise, and resolution in correlated fluctuations. Saldin et al. presents reconstruction of an icosahedral virus. Larabell & Nugent (2010) reviews imaging cellular architecture with X-rays. Feasibility of imaging living cells at subnanometer resolutions by ultrafast X-ray diffraction is discussed in the review, Bergh et al. (2008). Coherent diffractive imaging of biological samples is presented in Mancuso et al. (2010a) and Mancuso et al. (2010b). Ade & Stoll (2009) reviews near-edge X-ray absorption fine-structure microscopy.

For non-centrosymmetric and non-negative, and complex object, by imposing some general and reasonable constraints on the object, the phase of the diffracted wave can be retrieved from the magnitude (Sect. 2.1). The solution is very probably unique.

Much work has been done by many researchers with iterative algorithms and oversampling, and has gained much empirical success. The process of phase retrieval is time-consuming. However, it provides the theoretical foundation and gives the conditions that iterative algorithms with oversampling will give a solution which is probably unique. For iterative algorithms and oversampling, see Miao et al. (2003), Shapiro et al. (2005), Miao et al. (2006), Thibault et al. (2006), Huang et al. (2007), Kohli (2007), Sandberg et al. (2007), Thibault & Rankenburg (2007), Boutet et al. (2008), Morishita et al. (2008), Sandberg et al. (2008), Takahashi et al. (2008), Bostedt et al. (2009), Huang et al. (2009b), Takahashi et al. (2009a), Chapman & Nugent (2010), Doh et al. (2010), Nelson et al. (2010), Takahashi et al. (2010), Pelliccia et al.

(2011), Turner et al. (2011), Zhou et al. (2011) and references in Yuen (2011a, 2011b, 2011c, 2011d).

A review of X-ray phase imaging is presented in Peele & Nugent (2008) .

To apply iterative algorithms with oversampling, the basic idea is to use a unit cell to contain the nonperiodic object. Then mathematically add replica of the unit cell successively to give a crystal. In this crystal, we choose a basic unit cell which has six sharp boundary surfaces. For general shape of the object, for example, a sphere with a concentric cavity, we may not be able to choose a basic unit cell which has six sharp boundary surfaces (Sect. 2.2).

In order to have a basic unit cell which has six sharp boundary surfaces, we can use an orthogonal container to contain the nonperiodic object in solution (Sect. 2.3). The container plus the solvent and the nonperiodic object is considered as the basic unit cell and a new object, the structure of which will be determined. The container provides six sharp boundary surfaces. The shape and dimensions of the container walls form a container constraint, a very strong constraint. The orientations of the solvent molecules vary randomly with time. It produces a region of constant electron density (see solvent flattening in Wang (1985), Woolfson & Fan, 1995, Sect. 5.2, Bunk et al. (2007), p. 311). This provides a solvent constraint. Alternatively, we can embed the object in a liquid in a rectangular container, which is then solidified (Sect. 2.4). The rectangular block containing the nonperiodic object forms the basic unit cell, of which the structure will be determined. If a reference object is used, because of the very strong constraint of reproducing the structure of the reference object, it is most probable that the iterations will converge to the unique electron density.

For some objects, for example an astronomical object, a container or solidified liquid cannot be employed. We just use the nonperiodic object (Sect. 2.5).

For electron density of object reported in the literature, iterative algorithms with oversampling can be used to refine the electron density (Sect. 2.5.1). The iterative algorithms probably converge to a unique *ab initio* electron density with R -index = 0. This is an improvement of the electron density reported in the literature.

For electron density of object which has not been solved, its electron density with

R -index = 0 can also be determined by means of iterative algorithms with oversampling (Sect 2.5.2). The *ab initio* electron density obtained may not be unique.

In all cases, the experimental | Fourier coefficients | and in-between | Fourier coefficients | are improved by means of the consistent condition.

For complex object, the system may be over-determined. If the constraint of sharp boundary can be disregarded, procedures similar to Yuen (2011a) can be applied (Sect. 2.6).

This article does not apply to a conductor at frequencies with small depth of penetration.

2.1. Phase-retrieval method.

The phase of the diffracted wave will be retrieved from the magnitude by imposing some general and reasonable constraints upon the object. For non-centrosymmetric non-negative or complex nonperiodic objects, the constraints used are:

For the real part $f_r(x_1, x_2, x_3)$ of a complex nonperiodic object $f(x_1, x_2, x_3)$, use the constraints (Yuen (2011c), (C)):

(i) $f_r(x_1, x_2, x_3)$ has finite size. The size (shape and dimensions) will be determined.

(ii) $f_r(x_1, x_2, x_3) \geq 0$

(iii) $f_r(x_1, x_2, x_3)$ is bounded

(iv) $f_r(x_1, x_2, x_3)$ is continuous

(v) sharp boundary. The component function $f_r(x_1, x_2, x_3) \neq 0$ almost everywhere at the boundary surface (or boundary line for planar object)

with similar constraints for the imaginary part $f_i(x_1, x_2, x_3)$.

For the real part and the imaginary part together, we have a constraint:

(vi) the size and the regions of zeros (not isolated points) of the real part and the imaginary part are the same. This is a strong constraint.

For a non-centrosymmetric non-negative nonperiodic object, use constraints (i) – (v) (Yuen (2011b), (C)).

The Fourier transform of $f(x_1, x_2, x_3)$,

$$F(\omega_1,\omega_2,\omega_3)=\iiint f(x_1,x_2,x_3)\exp[-i(\omega_1 x_1+\omega_2 x_2+\omega_3 x_3)]dx_1 dx_2 dx_3$$

where the triple integral is over the volume of the object.

According to Yuen (2011b, 2011c), (C), the solution of the phase-retrieval method is very probably unique.

2.2. Iterative algorithms with oversampling.

The phase-retrieval method in Sect. 2.1 provides the theoretical foundation and gives the conditions that iterative algorithms with oversampling ratio = 2 will lead to the electron density which is probably unique. As the process in Sect. 2.1 is time-consuming, iterative algorithms with oversampling may be employed.

As we can see in Yuen (2011d), Sect. 2.2 the basic idea is to use a unit cell to contain the nonperiodic object. Then mathematically add replica of the unit cell successively to give a crystal. In this crystal, we choose a basic unit cell which has six sharp boundary surfaces. To achieve this, it is necessary to know the shape and dimensions of the nonperiodic object, and we use the smallest unit cell to enclose the object. For general shape of the object, we may not be able to choose a basic unit cell which has six sharp boundary surfaces. In some cases, for example, a sphere with a concentric cavity, there is no basic unit cell which has six sharp boundary surfaces. For thin spherical shell, most regions of the six boundary surfaces of any unit cell have zero values.

If we can choose a basic unit cell which has six sharp boundary surfaces, then we can use oversampling and iterative algorithms. Oversampling ratio of exactly 2 is needed.

2.3. Use a container

Use an orthogonal container to contain the nonperiodic object, for example, a macromolecule, in solution. The container plus solution and the nonperiodic object is considered as the basic unit cell and a new object, the structure of which will be determined. The shape and dimensions of the container walls form a very strong container constraint. The container provides six sharp boundary surfaces. The region of constant electron density of the solvent provides a solvent constraint. Mathematically add replica of the unit cell successively to give a crystal. Yuen (2011d) can then be applied.

According to phase-retrieval theory, all the | Fourier coefficients | and in-between | Fourier coefficients | are not independent (Yuen (2011a), Sect. 2.2). They need to be adjusted within their experimental uncertainties so that the electron densities obtained for each index are identical. This consistent condition must be satisfied.

Use

1. the experimentally determined dimensions of the container, and | Fourier coefficients | and in-between | Fourier coefficients | obtained from the experimentally measured diffraction intensity of the container plus the nonperiodic object.
2. Add a reference object of comparable size and known structure, for example, a pyramid, in the container,
3. random initial phases,
4. oversampling ratio $=2^3 = 8$; use | Fourier coefficients | and in-between | Fourier coefficients | for all (l,m,n),

   and all ($l+\frac{1}{2}$, and/or $m+\frac{1}{2}$, and/or $n+\frac{1}{2}$).
5. the five constraints for non-centrosymmetric non-negative nonperiodic object or eleven constraints for complex nonperiodic object (Sect. 2.1),
6. the container constraint and solvent constraint
7. in the final iterations, the | Fourier coefficients | in-between | Fourier coefficients | are relaxed or adjusted, within their uncertainties, to satisfy the consistent condition. Therefore we have **improved the experimental | Fourier coefficients | and in-between | Fourier coefficients |** reported in the literature.

Subjected to the strong constraint of reproducing the structure of the reference object, the iterative algorithms will most probably converge to a unique *ab initio* non-negative or complex electron density with R -index = 0.

Note that we have used the constraint of sharp boundary for both the unit cell and the nonperiodic object. The known dimensions of the container is a tight support, and a very strong and powerful constraint, in particular for three-dimensional crystal. Random initial phases are used. To compromise that proper initial phases are not used, we use the reference object and large oversampling ratio =8. In the process of iterations, the reference object has to be reproduced. Because of these, the iterative algorithms most probably converge to a unique non-negative or complex electron density. This convergence can be investigated empirically and theoretically.

If a reference object is not used, the shape and dimensions of the convex hull (envelope) of the nonperiodic object can be determined (Yuen (2011e), Sect. 2.4), and used as a loose support. The electron density obtained may be unique.

To avoid radiation damage, the solution can be frozen. The macromolecules have structure features, while the solvent is more homogeneous. These can be used in the iterative algorithms. The atomic coordinates of the macromolecule, the container and the solution of the basic unit cell will be determined.

Triclinic container may be used if the iterations can converge more efficiently.

2.4. Embedded in a solidified liquid

As in Sect. 2.3, the nonperiodic object, for example a macromolecule, can be embedded in a liquid in a rectangular container, which is then solidified. A reference object may or may not be used. The rectangular block containing the nonperiodic object forms the basic unit cell, of which the structure will be determined as in Sect. 2.3. If a reference object is used, the iterative algorithms most probably converge to a unique *ab initio* non-negative or complex electron density. The macromolecules have structure features, while the solidified liquid is more homogeneous. Radiation damage to the macromolecule is prevented by the solid block. The atomic coordinates of the macromolecule and the solidified liquid of the basic unit cell will be determined.

Parallelpiped of the solid may be used if the iterations can converge more efficiently.

2.5. Just a nonperiodic object

For some objects, for example an astronomical object, a container or solidified liquid cannot be employed. We just use the nonperiodic object.

2.5.1. Iterative algorithms with oversampling. Refinement of electron density of object reported in the literature

The electron density of an object reported in the literature has been determined by various methods. Use this electron density. Calculate the Fourier coefficients and in-between Fourier coefficients from the electron density in the basic unit cell. Use their phases as initial phases.

Use

1. the shape and dimensions of the convex hull (envelope) of the nonperiodic object (Yuen (2011e), Sect. 2.4). If the object is convex, this is a tight support. If the object is concave, this is a loose support.
2. Use a unit cell of smallest volume (or area for 2-dimensional object) to contain the convex hull of the object.
3. known unit cell constants.

   Use the | Fourier coefficients | and in-between | Fourier coefficients | obtained from the experimentally measured diffraction intensity of the nonperiodic object.
4. the initial phases,
5. oversampling ratio $=2^3 = 8$; use | Fourier coefficients | and in-between | Fourier coefficients | for all (l, m, n),

   and all ($l+\frac{1}{2}$, and/or $m+\frac{1}{2}$, and/or $n+\frac{1}{2}$).
6. the five constraints for non-centrosymmetric non-negative nonperiodic object or eleven constraints for complex nonperiodic object (Sect. 2.1).
8. in the final iterations, the | Fourier coefficients | and in-between | Fourier coefficients | are relaxed or adjusted, within their uncertainties, to satisfy the consistent condition.

We have convex hull and five constraints for a non-centrosymmetric non-negative nonperiodic object, and eleven constraints for a complex nonperiodic object. We use large oversampling ratio=8. T**he iterative algorithms will converge to a unique *ab initio* non-negative or complex electron density with R-index = 0.** The electron density of an object reported in the literature should not be very different from the true electron density. When we use the electron density reported in the literature as the initial electron density, it is reasonable to expect that the iterations will converge to a unique and improved electron density. This convergence can be investigated empirically and theoretically.

2.5.2. Iterative algorithms with oversampling. Determination of electron density of object which has not been obtained by other methods

Follow Sect. 2.5.1. We use random initial phases which are commonly employed. The phase-retrieval method in Sect. 2.1 shows that oversampling ratio = 2 very probably leads to a unique electron density. To compromise that proper initial phases are not used, more oversampling is employed. We use | Fourier coefficients | and in-between | Fourier coefficients | for all (l,m,n), and all ($l+\frac{1}{2}$, and/or $m+\frac{1}{2}$, and/or $n+\frac{1}{2}$). The oversampling ratio is $2^3=8$. The iterative algorithms may converge to an *ab initio* non-negative or complex electron density with R -index = 0. The convergence is determined by the boundary value, i.e. the boundary height of the object. This convergence can be investigated empirically and theoretically. The electron density obtained may not be unique.

2.6. Over-determined condition for complex object

It is mentioned in Yuen (2011c) that for complex object, the system may be over-determined. If the constraint of sharp boundary can be disregarded, container, solution or solidified liquid are not needed. Replicas of the object can be added mathematically to form a three-dimensional crystal. Procedures similar to Yuen (2011a), Sect. 2.2, 2.3 can be applied.

3. Discussions and conclusion

The phase-retrieval method in Sect. 2.1 provides the theoretical foundation and gives the conditions that iterative algorithms with oversampling will give a probably unique solution.

To apply iterative algorithms with oversampling, the basic idea is to use a unit cell to contain the nonperiodic object, and the unit cell has six sharp boundary surface. Then mathematically add replica of the unit cell successively to give a crystal. To achieve this purpose, we can use a rectangular container containing the object, or embed the object in a liquid which is then solidified. The container or the solidified liquid plus the object form the basic unit cell, the structure of which will be determined.

The shape and dimensions of the container walls form a very strong container constraint. For a nonperiodic object, for example, a macromolecule, in a solvent, the region of constant electron density of the solvent provides a solvent constraint. By means of iterative algorithms, the atomic coordinates of the macromolecule and the container of the basic unit cell will be determined.

For a macromolecule, protection against radiation damage is provided by employing frozen solution or solidified liquid. The macromolecules have structure features, while the solvent is more homogeneous. The atomic coordinates of the macromolecule, the container and the solution of the basic unit cell will be determined. Researches can be done in seeking for solidified liquids which provide same environments as the solutions for macromolecules.

As the electron density of the macromolecule and the solvent, frozen or not, are determined, this helps in the studies of the interaction of the macromolecule and the solvent molecules, in understanding the biological activity of the macromolecule, and is important in medicine.

For objects such as an astronomical object, a container or solidified liquid cannot be employed. We just use the nonperiodic object. The electron density of an object reported in the literature can be refined by iterative algorithms with oversampling. We urge that researchers will apply iterative algorithm with oversampling to refine the electron density of the many electron densities of nonperiodic objects reported in the literature, in particular, those structures solved by themselves (Sect. 2.5.1). Improved electron density with R-index = 0 will be obtained. The *ab initio* electron density obtained is unique. It is determined by the experimental diffraction intensities alone; no other chemical or physical information (for example, atomicity, chemical formula, bond length, bond angle, etc) is used. It can be regarded as a fundamental property of the object. It is just a mathematical transformation of the diffraction intensities to an internal property of the object.

For electron density of object which has not been solved, its electron density with R -index = 0 can also be determined by means of iterative algorithms with oversampling. A container (Sect. 2.3), or solidified liquid (Sect. 2.4) or just the nonperiodic object (Sect. 2.5.1) can be used. Random initial phases may be adopted. To compromise that proper initial phases are not used, more oversampling is employed by using all the in-between | Fourier coefficients | . The oversampling ratio is $2^3 = 8$. The iterative algorithms may converge to an *ab initio* non-negative or complex electron density with R -index = 0. The convergence is determined by the boundary value, i.e. the boundary height of the object. This convergence can be investigated empirically and theoretically. The electron density obtained may not be unique.

If a reference object is used, because of the very strong constraint of reproducing the structure of the reference object, it is most probable that the iterations will converge to the unique electron density. From the *ab initio* electron density, the atoms, their species and co-ordinates can be determined. The chemical formula can then be obtained. This serves as an experimental method of obtaining the atomic species and the chemical formula.

In all cases, The experimental | Fourier coefficients | and in-between | Fourier coefficients | are improved by means of the consistent condition.

For complex object, the system may be over-determined. If the constraint of sharp boundary can be disregarded, procedures similar to Yuen (2011a), Sect. 2.2, 2.3 can be applied.

By employing radiations of various wavelengths, cellular architecture at various resolutions can be imaged (Yuen (2011b), Sect. 2.3, Larabell & Nugent (2010)).

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16

Ab initio determination of electron densities of non-centrosymmetric non-negative or complex fibres from diffraction intensities. Iterative algorithms with oversampling

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Abstract

For a non-centrosymmetric non-negative or complex fibre (freely rotating or fixed), phase-retrieval method can be applied to obtain the phase from the diffraction magnitudes. With the constraints employed, the solution of the phase problem is very probable unique. The phase-retrieval method provides the theoretical foundation and serves as a theoretical basis for iterative algorithms with oversampling so that the iterative algorithms very probably converge to a unique non-negative or complex electron density with R-index = 0. A container may be used to contain the fibre and solution. The shape and dimensions, and constant electron density of the container walls form a very strong container constraint. The region of constant electron density of the solvent provides a solvent constraint. The solution may be frozen to prevent radiation damage to the fibre, for example, a macromolecule. The fibre may also be embedded in a liquid which is then solidified. Iterative algorithms with oversampling may also be applied to a freely rotating or fixed fibre; a container or solidified liquid is not employed.

Keywords: Fiber diffraction; freely rotating fibre; fixed fibre; one-dimensional periodic object; phase retrieval; unique solution; exact solution; oversampling; container constraint; solvent constraint; macromolecules; solidified liquid; radiation damage; protection.

1. Introduction

This is a sequel of Yuen (2011b, 2011c, 2011d. 2011e, 2011h).

For fibre diffraction, see Makowski (1981, 1982), Namba & Stubbs (1985), Millane (1988), and Kendall et al. (2008). Anomalous scattering on fibres is presented in Nave

(1987). Likelihood function is presented in Mu & Makowski (2000). Wang & Stubbs (1993) presents molecular dynamics in refinement of fibre diffraction data. Fiber drawing is presented in Ran et al. (2000). Reviews of fibre diffraction are presented in Stubbs (1999, 2001).

Diaz et al. (2005), Bunk et al. (2007b), Bunk et al. (2008), Satapathy et al. (2008), Nygård et al. (2009) and Satapathy et al. (2009) present a very useful and important instrument, microcavity arrays, and articles of diffractive imaging for one-dimensional periodic samples: retrieving concentration profiles across microfluidic channels. For iterative algorithms and oversampling, see Bunk et al. (2007a). Minkevich et al. (2011) presents selective coherent x-ray diffractive imaging of displacement fields in periodic wires.

For non-centrosymmetric non-negative or complex freely rotating or fixed fibres, phase-retrieval method can be applied to obtain the phase from the diffraction magnitudes (Sect. 2.1, 3.1). With the constraints employed, the solution of the phase problem is very probable unique.

The phase-retrieval method in Sect. 2.1, 3.1 is time-consuming. However, it provides the theoretical foundation and serves as a theoretical basis for iterative algorithms with oversampling so that the iterative algorithms very probably converge to a unique non-negative or complex electron density with R-index = 0. In Sect. 2.2, 3.2, a container is used to contain the fibre and solution. The solution may be frozen to prevent radiation damage to the fibre, for example a macromolecule. The shape and dimensions, and constant electron density of the container walls form a very strong container constraint. The region of constant electron density of the solvent provides a solvent constraint. The fibre may also be embedded in a liquid which is then solidified (Sect. 2.3, 3.3). Iterative algorithms with oversampling may also be applied to a freely rotating or fixed fibre; a container or solidified liquid is not employed (Sect. 4).

Over-determined condition for complex object is discussed in Sect. 5.

2.1. Fibre (freely rotating). Phase-retrieval method.

Fibre has circular symmetry in the x_1x_2 plane.

For the real part and the imaginary part together, we have a constraint:

(i) the size and the regions of zeros (not isolated points) of the real part and the imaginary part are the same. This is a very strong constraint.

The fibre has radius $R(x_3)$, and is periodic in x_3 with period c for both the real and imaginary parts.

For the real part, constraints used:

(ii) For each x_3, the fibre has finite radius $R(x_3)$ (value will be determined) in the x_1x_2 plane. $f_{r0}(x_1, x_2, x_3) = 0$ for $x_3 < 0$ or $x_3 > c$. Value of c is known.

(iii) $f_{r0}(x_1, x_2, x_3)$ is non-negative,

(iv) $f_{r0}(x_1, x_2, x_3)$ is bounded

(v) sharp boundary. $f_{r0}(x_1, x_2, x_3) > 0$ almost everywhere at the two boundary circles at $x_3 = 0$ and c, and at the boundary surface of revolution about the x_3 axis

(vi) $f_{r0}(x_1, x_2, x_3)$ is continuous with

$$f_{r0}(x_1, x_2, 0) = f_{r0}(x_1, x_2, c) \quad \text{for all } x_1,\ x_2,$$

(vii) For each x_3, the object has circular symmetry in the x_1x_2 plane.

with similar constraints for the imaginary part $f_{i0}(x_1, x_2, x_3)$.

For a non-centrosymmetric non-negative fibre, use constraints (ii) – (vi) (Yuen (2011c), (E)).

Because of the one-dimensional periodic structure, the diffracted intensity is greatly enhanced to form strong diffraction planes which can be measured accurately. Let $\omega_0 = 2\pi / c$. For all n, $n = 0, \pm1, \pm2., \ldots$, $|F_0(\omega_1, \omega_2, n\omega_0)|$ are measured experimentally. It has circular symmetry in the $\omega_1\omega_2$ plane.

$F_0(\omega_1, \omega_2, n\omega_0)$

$$=\int\int\left\{\int_0^c f_0(x_1,x_2,x_3)\exp(-jn\omega_0 x_3)dx_3\right\}\exp[-j(\omega_1 x_1+\omega_2 x_2)]dx_1 dx_2 \tag{1}$$

where the triple integral is over one period of the fibre.

For a fixed n', the term in curly brackets, $q(x_1,x_2,n'\omega_0)$, is a two-dimensional centrosymmetric complex object of x_1 and x_2. $q(x_1,x_2,n'\omega_0)$ is circularly symmetric in the x_1x_2 plane with radius = the largest $R(x_3)$. This becomes a two-dimensional problem. Constraints used:

(ii) $q(x_1,x_2,n'\omega_0)$ is circularly symmetric in the x_1x_2 plane with radius = the largest $R(x_3)$ which will be determined

(iv) $q(x_1,x_2,n'\omega_0)$ is bounded

(vi) $q(x_1,x_2,n'\omega_0)$ is continuous

Apply the method in Yuen (2011c, 2011d), (C). Use $|F_0(\omega_1,\omega_2,n'\omega_0)|$ for all ω_1, $\omega_2=0$.

Flipping a finite or denumerable number of zeros does not change the length of the object. In particular, the length of the object given by the inverse Fourier transform of the minimum-phase function is the length of the object. Value of the largest $R(x_3)$ is found.

Find the possible $q(x_1,x_2,n'\omega_0)$ satisfying these three constraints. $q(x_1,x_2,n'\omega_0)=0$ at its boundary in the x_1x_2 plane. Since it does not have the strong constraints of known size and sharp boundary, the number of possible $q(x_1,x_2,n'\omega_0)$ satisfying these three constraints, N is very large. Use the Fourier transform $F_0(\omega_1,\omega_2,n'\omega_0)$ in (1) of each possible $q(x_1,x_2,n'\omega_0)$. Denote these by ${}^1F_0(\omega_1,\omega_2,n'\omega_0)$,…, ${}^NF_0(\omega_1,\omega_2,n'\omega_0)$. The Fourier transform

$F_0(\omega_1,\omega_2,\omega_3)$ of $f_0(x_1,x_2,x_3)$ belongs to an entire function. For all n, associate the N functions ${}^1F_0(\omega_1,\omega_2,n'\omega_0)$,…, ${}^NF_0(\omega_1,\omega_2,n'\omega_0)$ from individual n' so that they belong to N continuous functions ${}^1F_0(\omega_1,\omega_2,\omega_3)$,…, ${}^NF_0(\omega_1,\omega_2,\omega_3)$. (If c is large, ω_0 is small. This association will be easier. If ω_0 is large, there may be ambiguity, resulting in more than N functions). From these, get ${}^1q(x_1,x_2,n\omega_0)$,…, ${}^Nq(x_1,x_2,n\omega_0)$.

Now $f(x_1,x_2,x_3)$ is periodic in x_3 with period c. For fixed x_1 and x_2, express $f(x_1,x_2,x_3)$ as a Fourier series of x_3.
The Fourier coefficient

$$=\frac{1}{c}\int_0^c f_0(x_1,x_2,x_3)\exp\left(-jn\omega_0x_3\right)dx_3=\frac{1}{c}q(x_1,x_2,n\omega_0)$$

Hence $$f(x_1,x_2,x_3)=\frac{1}{c}\sum_{n=-\infty}^{\infty}q(x_1,x_2,n\omega_0)\exp\left(jn\omega_0x_3\right) \tag{2}$$

For each $q(x_1,x_2,n\omega_0)$ and all x_1 and x_2, obtain a possible structure $f(x_1,x_2,x_3)$. As shown in the Appendix of Yuen (2011c), it is very probable that only one of the N possible $f_0(x_1,x_2,x_3)$ satisfies constraint (i), and its real and imaginary parts each satisfying the constraints (ii) to (vi). Thus the structure $f(x_1,x_2,x_3)$ including $R(x_3)$ is determined.

Note that this is a case of determining unique periodic structures without using interpolation. The structure obtained is exact.

2.2. Fibre (freely rotating). Use a container. Iterative algorithms with oversampling.

The phase-retrieval method (Sect. 2.1) is time-consuming. However, it provides the theoretical foundation and serves as a theoretical basis for iterative algorithms with oversampling.

Consider a non-crystalline fibre specimen (Millane (1988), p. 171). The molecules are approximately parallel to each other, have random rotations about the long axis, and not laterally organized, such that we have, in effect, a fibre (a one-dimensional periodic object) freely rotating about the long axis.

Use (Yuen (2011f), (H)), to determine the maximum diameter of a molecule of the fibre. As in Yuen (2011h), Sect. 2.3, use an orthogonal long container to contain the fibre and solution. The fibre axis is along the longest dimension of the container. The solution is frozen at very low temperature. The molecules of the solution have random locations and orientations. It constitutes a background of constant electron density in comparison to the strong diffraction planes of the fibre. This provides a solvent constraint. Likewise, the walls of the container have constant electron density. The shape and dimensions, and constant electron density of the container walls form a very strong container constraint (Bunk et al. (2007b), p. 311). The basic unit cell consists of one period of a molecule of the fibre plus the frozen solution and the top, bottom and two sides of the container. The frozen solution and the container provide the six sharp boundary surfaces. The atomic coordinates of the fibre molecule in the basic unit cell will be determined. Mathematically add replica of the basic unit cell successively to give a crystal. Yuen (2011e) can then be applied.

The | Fourier coefficients | for all (l, m, n) and in-between | Fourier coefficients | for ($l+\frac{1}{2}$, and/or $m+\frac{1}{2}$, n) are obtained from the experimentally measured intensity of the container plus the fibre and solution. The other in-between | Fourier coefficients | are obtained by interpolation in multidimensions.

If we use:

1. the known value of the maximum diamenter of the fibre molecule. This is a loose support.
2. the experimentally determined dimensions of the basic unit cell, and | Fourier

coefficients| for (l,m,n) and in-between |Fourier coefficients| for ($l+\frac{1}{2}$, and/or $m+\frac{1}{2}$, n) obtained from the experimentally measured diffraction intensity of the container plus the fibre and solution,

3. random initial phases,
4. oversampling ratio = $2^3 = 8$; use |Fourier coefficients| and in-between |Fourier coefficients| for all (l,m,n),

   and all ($l+\frac{1}{2}$, and/or $m+\frac{1}{2}$, and/or $n+\frac{1}{2}$).

   Some of them are experimental data (see 2.), the rest are obtained by interpolation in multidimensions.
5. the six constraints for non-centrosymmetric non-negative fibre or thirteen constraints for complex fibre in Sect. 2.1,
6. the container constraint and solvent constraint,
7. in the final iterations, the interpolated in-between |Fourier coefficients| are relaxed or adjusted, within their uncertainties, to satisfy the consistent condition (Yuen (2011a), Sect. 2.1); the interpolated in-between |Fourier coefficients| are improved.,

the iterative algorithms will converge to a non-negative or complex electron density with R-index = 0. The electron density is very probably unique for this set of |Fourier coefficients| and in-between |Fourier coefficients| .

Note that we have used the constraint of sharp boundary for both the unit cell and the surface of revolution of the fibre. To compromise that proper initial phases are not used, we use known value of the maximum diameter of the fibre, and larger oversampling ratio = $2^3 = 8$; use |Fourier coefficients| and in-between |Fourier coefficients| for all (l,m,n), and all ($l+\frac{1}{2}$, and/or $m+\frac{1}{2}$, and/or $n+\frac{1}{2}$). We hope that these will help the iterative algorithms to converge to a unique non-negative or complex electron density with R -index = 0. This convergence can be investigated empirically and theoretically.

As shown by the phase-retrieval method in Sect. 2.1, the interpolated in-between ∣Fourier coefficients∣ are, in fact, not needed. After the iterations converge to a electron density, we stop using all the interpolated in-between ∣Fourier coefficients∣ . The iterative algorithms will converge to a **unique** non-negative or complex electron density.

Radiation damage to the fibre, for example a macromolecule, may be prevented by the frozen solution.

2.3. A fibre embedded in a solidified liquid. Iterative algorithms with oversampling

The fibre, for example a macromolecule, can be embedded in a liquid in a rectangular container, which is then solidified. The rectangular block containing one period of a molecule of the fibre plus the solidified liquid forms a unit cell, of which the structure will be determined as in Sect. 2.2. Radiation damage to the macromolecule is prevented by the solid block. The atomic coordinates of the fibre molecule will be determined.

Parallelpiped of the solid may be used if the iterations can converge more efficiently.

3.1. Fixed fibre (a one-dimensional periodic object). Phase-retrieval method.

For the real part and the imaginary part together, we have a constraint:

(i) the size and the regions of zeros (not isolated points) of the real part and the imaginary part are the same. This is a very strong constraint.

The fibre is periodic in x_3 with period c for both the real and imaginary parts.

For the real part, constraints used:

(ii) finite size. $f_{r0}(x_1, x_2, x_3) = 0$ for $x_3 < 0$ or $x_3 > c$. Value of c is known. Shape and dimensions of the fibre will be determined.

(iii) $f_{r0}(x_1, x_2, x_3)$ is non-negative,

(iv) $f_{r0}(x_1, x_2, x_3)$ is bounded

(v) sharp boundary. $f_{r0}(x_1, x_2, x_3) > 0$ almost everywhere at the two boundary planes at $x_3 = 0$ and c, and at the boundary surface of the fibre

(vi) $f_{r0}(x_1, x_2, x_3)$ is continuous with

$$f_{r0}(x_1, x_2, 0) = f_{r0}(x_1, x_2, c) \quad \text{for all } x_1,\ x_2$$

(vii) symmetry elements, if applicable,

with similar constraints for the imaginary part $f_{i0}(x_1, x_2, x_3)$.

For a non-centrosymmetric non-negative fibre, use constraints (ii) – (vii) (Yuen (2011c), (F)).

Because of the one-dimensional periodic structure, the diffracted intensity is greatly enhanced to form strong diffraction planes which can be measured accurately. Let $\omega_0 = 2\pi / c$. For all n, $n = 0, \pm 1, \pm 2., \ldots$, $\left|F_0(\omega_1, \omega_2, n\omega_0)\right|$ are measured experimentally.

$$F_0(\omega_1, \omega_2, n\omega_0)$$
$$= \int\int \left\{ \int_0^c f_0(x_1, x_2, x_3) \exp(-jn\omega_0 x_3) dx_3 \right\} \exp[-j(\omega_1 x_1 + \omega_2 x_2)] dx_1 dx_2 \tag{3}$$

where the triple integral is over one period of the fibre.

For a fixed n', the term in curly brackets, $q(x_1, x_2, n'\omega_0)$, is a two-dimensional complex object of x_1 and x_2. This becomes a two-dimensional problem. Constraints used:

(ii) $q(x_1, x_2, n'\omega_0)$ has finite size which will be determined

(iv) $q(x_1, x_2, n'\omega_0)$ is bounded

(vi) $q(x_1, x_2, n'\omega_0)$ is continuous

Apply the method in Yuen (2011d), (C). Use $\left|F_0(\omega_1, \omega_2, n'\omega_0)\right|$ for all ω_1, ω_2. Find the possible $q(x_1, x_2, n'\omega_0)$ satisfying these three constraints. $q(x_1, x_2, n'\omega_0) = 0$ at its boundary in the $x_1 x_2$ plane. Since it does not have the strong constraints of known size and sharp boundary, the number of possible $q(x_1, x_2, n'\omega_0)$ satisfying these three constraints, N is very large. Use the Fourier transform $F_0(\omega_1, \omega_2, n'\omega_0)$ in (3) of each possible $q(x_1, x_2, n'\omega_0)$. Denote these by ${}^1F_0(\omega_1, \omega_2, n'\omega_0)$,…, ${}^NF_0(\omega_1, \omega_2, n'\omega_0)$. The Fourier transform $F_0(\omega_1, \omega_2, \omega_3)$ of $f_0(x_1, x_2, x_3)$ belongs to an entire function. For all n, associate the N functions ${}^1F_0(\omega_1, \omega_2, n'\omega_0)$,…, ${}^NF_0(\omega_1, \omega_2, n'\omega_0)$ from individual n' so that they belong to N continuous functions ${}^1F_0(\omega_1, \omega_2, \omega_3)$,…, ${}^NF_0(\omega_1, \omega_2, \omega_3)$. (If c is large, ω_0 is small. This association will be easier. If ω_0 is large, there may be ambiguity, resulting in more than N functions). From these, get ${}^1q(x_1, x_2, n\omega_0)$,…, ${}^Nq(x_1, x_2, n\omega_0)$. For each $q(x_1, x_2, l\omega_0)$ and all x_1 and x_2, obtain a possible structure $f(x_1, x_2, x_3)$ from (2). As shown in the Appendix of Yuen (2011c), it is very probable that only one of the N possible $f_0(x_1, x_2, x_3)$ satisfies constraint (i), and its real and imaginary parts each satisfying the constraints (ii) to (vii). Thus the structure $f(x_1, x_2, x_3)$ including shape and dimensions of the fibre is determined.

Note that this is a case of determining unique periodic structures without using interpolation. The structure obtained is exact.

3.2. A fixed fibre. Use a container. Iterative algorithms with oversampling.

The phase-retrieval method (Sect. 3.1) is time-consuming. However, it provides the theoretical foundation and serves as a theoretical basis for iterative algorithms with oversampling.

For a fixed fibre (a one-dimensional periodic object), use Yuen (2011g), Sect. 2.2 to determine the maximum length of the fibre along any direction normal to the fibre axis.

As in Yuen (2011h), Sect. 2.3, use an orthogonal long container to contain the fibre and solution. The fibre axis is along the longest dimension of the container. The solution is frozen at very low temperature. The region of constant electron density of the solvent provides a solvent constraint. The shape and dimensions, and constant electron density of the container walls form a very strong container constraint. The basic unit cell consists of one period of the fibre plus the frozen solution and the top, bottom and two sides of the container. The frozen solution and the container provide the six sharp boundary surfaces. The atomic coordinates of the fibre in the basic unit cell will be determined. Mathematically add replica of the basic unit cell successively to give a crystal. Yuen (2011e) can then be applied.

The | Fourier coefficients | for all (l,m,n) and in-between | Fourier coefficients | for ($l+\frac{1}{2}$, and/or $m+\frac{1}{2}$, n) are obtained from the experimentally measured intensity of the container plus the fibre and solution. The other in-between | Fourier coefficients | are obtained by interpolation in multidimensions.

If we use:

1. the known value of the maximum length of a fixed fibre along any direction normal to the fibre axis. This is a loose support.
2. the experimentally determined dimensions of the basic unit cell, and | Fourier coefficients | for (l,m,n) and in-between | Fourier coefficients | for ($l+\frac{1}{2}$, and/or $m+\frac{1}{2}$, n) obtained from the experimentally measured diffraction intensity of the container plus the fibre and solution,
3. random initial phases,
4. oversampling ratio = $2^3 = 8$; use | Fourier coefficients | and in-between | Fourier coefficients | for all (l,m,n),

and all ($l+\frac{1}{2}$, and/or $m+\frac{1}{2}$, and/or $n+\frac{1}{2}$).

Some of them are experimental data (see 2.), the rest are obtained by interpolation in multidimensions.

5. the six constraints for non-centrosymmetric non-negative fibre or thirteen constraints for complex fibre in Sect. 3.1,
6. the container constraint and the solvent constraint
7. in the final iterations, the interpolated in-between $|$ Fourier coefficients $|$ are relaxed or adjusted, within their uncertainties, to satisfy the consistent condition (Yuen (2011a), Sect. 2.1); the interpolated in-between $|$ Fourier coefficients $|$ are improved,

the iterative algorithms will converge to a non-negative or complex electron density with R -index = 0. The electron density is very probably unique for this set of $|$ Fourier coefficients $|$ and in-between $|$ Fourier coefficients $|$.

Note that we have used the constraint of sharp boundary for both the unit cell and the fibre. To compromise that proper initial phases are not used, we use known value of the maximum length of the fibre along any direction normal to the fibre axis, and larger oversampling ratio = $2^3 = 8$; use $|$ Fourier coefficients $|$ and in-between $|$ Fourier coefficients $|$ for all (l,m,n), and all ($l+\frac{1}{2}$, and/or $m+\frac{1}{2}$, and/or $n+\frac{1}{2}$). We hope that these will help the iterative algorithms to converge to a unique non-negative or complex electron density with R -index = 0. This convergence can be investigated empirically and theoretically.

As shown by the phase-retrieval method in Sect. 3.1, the interpolated in-between $|$ Fourier coefficients $|$ are, in fact, not needed. After the iterations converge to a electron density, we stop using all the interpolated in-between $|$ Fourier coefficients $|$. The iterative algorithms will converge to a **unique** non-negative or complex electron density.

3.3. A fixed fibre. Embedded in a solidified liquid. Iterative algorithms with oversampling

Similar to Sect. 2.3.

4. Fibre (freely rotating) and fixed fibre. Iterative algorithms with oversampling

We may just use a fibre (freely rotating) or fixed fibre; a container or solidified liquid is not employed.

Use

1. the maximum diameter of a freely rotating fibre (Yuen (2011f), (H)), or the maximum length of a fixed fibre along any direction normal to the fibre axis (Yuen (2011g), Sect. 2.2) as a loose support,
2. the known unit cell constants, the ∣ Fourier coefficients∣ and in-between ∣ Fourier coefficients∣ obtained from the experimentally measured diffraction intensity of the freely rotating or fixed fibre,
3. random initial phases,
4. oversampling ratio $= 2^3 = 8$; use ∣ Fourier coefficients∣ and in-between ∣ Fourier coefficients∣ for all (l, m, n),

   and all ($l+\frac{1}{2}$, and/or $m+\frac{1}{2}$, and/or $n+\frac{1}{2}$).

   Some of them are experimental data (see 2.), the rest are obtained by interpolation in multidimensions.

7. the six constraints for non-centrosymmetric non-negative freely rotating fiber or thirteen constraints for complex freely rotating fibre in Sect. 2.1.

   The six constraints for non-centrosymmetric non-negative fixed fiber or thirteen constraints for complex fixed fibre in Sect. 3.1.

6. in the final iterations, the interpolated in-between ∣ Fourier coefficients∣ are relaxed or adjusted, within their uncertainties, to satisfy the consistent condition (Yuen (2011a), Sect. 2.1); the interpolated in-between ∣ Fourier coefficients∣ are improved.

We use known maximum diameter of a freely rotating fibre, or the maximum length of a fixed fibre along any direction normal to the fibre axis. For a non-centrosymmetric non-negative freely rotating or fixed fibre, we use six constraints including the powerful sharp boundary at the two boundary planes at $x_3 = 0$ and c and the surface of revolution of the fibre. For a complex freely rotating or fixed fibre, we use thirteen constraints including the sharp boundary at the two boundary planes. In both cases, we use the larger oversampling ratio=8. **The iterative algorithms will converge to a non-negative or complex electron density with R-index = 0. The electron density is very probably unique for this set of | Fourier coefficients | and in-between | Fourier coefficients | .** This convergence can be investigated empirically and theoretically.

As shown by the phase-retrieval methods in Sect. 2.1 and 3.1, the interpolated in-between | Fourier coefficients | are, in fact, not needed. After the iterations converge to an non-negative electron density, we stop using all the interpolated in-between | Fourier coefficients | . The iterative algorithms may converge to a **unique** non-negative or complex electron density.

5. Over-determined condition for complex object

It is mentioned in Yuen (2011d) that for complex object, the system may be over-determined. If the constraint of sharp boundary can be disregarded, container, solution or solidified liquid are not needed. Replicas of a freely rotating or fixed fibre can be added mathematically to form a three-dimensional crystal of parallel fibres. Procedures similar to Yuen (2011a), Sect. 2.2, 2.3 can then be applied.

6. Discussions and conclusion

The phase-retrieval methods in Sect. 2.1 and 3.1 provide the theoretical foundation and give the conditions that iterative algorithms with oversampling will very probably converge to a unique solution. A container may be used to contain the fibre and solution. The solution may be frozen to prevent radiation damage to the fibre, for example, a macromolecule. The fibre may also be embedded in a liquid which is then solidified. The shape and dimensions, and constant electron density of the container walls form a very strong container constraint. The region of constant electron density of the solvent provides a solvent constraint.

Because of the one-dimensional periodic structure, the diffracted intensity is greatly enhanced to form strong diffraction planes. As the oversampling ratio is exactly 2, for a periodic fibre, no interpolation is needed. The intensity in the diffraction planes can be measured experimentally. The electron density obtained is unique and exact. This is an important case of determining unique periodic structures without using interpolation. By means of iterative algorithms with oversampling, electron density with R -index = 0 can be obtained. The *ab initio* electron density is determined by the experimental diffraction intensities alone; no other chemical or physical information (for example, atomicity, chemical formula, bond length, bond angle, etc) is used. It can be regarded as a fundamental property of the fibre. It is just a mathematical transformation of the diffraction intensities to an internal property of the fibre. Theories of molecular structure and properties (for example, quantum theory) should agree with this internal property. It forms a base for interpretation of experimental data. From this electron density, atomic coordinates are obtained. The atomic coordinates can be compared with those obtained from spectroscopy.

The one-dimensional periodic structure in Diaz et al. (2005) and Bunk et al. (2007b) is a good example of complex fixed fibre. Each channel is a rectangular container. A rectangular container plus the solution is treated as the object (the basic unit cell). Because of the one-dimensional periodic structure, the diffracted intensity is greatly enhanced to form strong diffraction planes. Iterative algorithms with oversampling in Sect. 3.2 for three-dimensional phase-retrieval can be applied. The constant electron density and known shape and dimensions of a channel provide a very strong container constraint. The region of constant electron density of the solvent provides a solvent constraint. The six surfaces of a channel provide the strong sharp-boundary constraint. The iterative algorithms will very probably converge to a unique complex electron density with R -index = 0.

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17

Ab initio determination of electron densities of non-centrosymmetric non-negative or complex membranes from diffraction intensities. Iterative algorithms with oversampling

November 29, 2011

Abstract

For a non-centrosymmetric non-negative or complex planar periodic membrane, phase-retrieval method can be applied to obtain the phase from the diffraction magnitudes. With the constraints employed, the solution of the phase problem is very probable unique. The phase-retrieval method provides the theoretical foundation and serves as a theoretical basis for iterative algorithms with oversampling so that the iterative algorithms very probably converge to a unique non-negative or complex electron density with R -index = 0. A container may be used to contain the membrane and solution. The solution may be frozen to prevent radiation damage to the membrane, for example, a macromolecule. The membrane may also be embedded in a liquid which is then solidified. Iterative algorithms with oversampling may also be applied to a membrane; a container or solidified liquid is not employed.

Keywords: Membrane diffraction; two-dimensional periodic object; diffraction rods; Bragg rods; phase retrieval; very probably unique solution.

1. Introduction

This is a sequel of Yuen (2011b, 2011c, 2011d. 2011e, 2011g).

For structures of two-dimensional crystals, see Li (1985), Dorset (1991), Vainshtein & Klechkovskaya (1993), Dorset (1995), Leslie et al. (1999), Marks (1999), Stevens et al. (2002), Subramaniam et al. (2002), Chen et al. (2005). Electron diffraction structure analysis of phospholipids is reviewed in Dorset (1987). For a review of direct methods for surface crystallography see Saldin & Shneerson (2008). Robinson & Tweet (1992) presents a review of surface X-ray diffracton.

For use of Bragg rod analysis in structure analysis, see Yacoby et al. (2000), Sowwan et al. (2002), Yacoby et al. (2002), Wakabayashi et al. (2010), Cohen et al. (2011), Kumah et al. (2011), and Yamamoto et al. (2011).

For iterative algorithms and oversampling, see Saldin et al. (2001), Fenter & Zhang (2005), Lyman et al. (2005), Lyman et al. (2006), Fung et al. (2007), Björck et al. (2008), Mancuso et al. (2009), Vartanyants et al. (2010). Mancuso et al. (2010) presents a review.

For non-centrosymmetric non-negative or complex planar periodic membrane, phase-retrieval method can be applied to obtain the phase from the diffraction magnitudes (Sect. 2.1). With the constraints employed, the solution of the phase problem is very probable unique.

The phase-retrieval method in Sect. 2.1 is time-consuming. However, it provides the theoretical foundation and serves as a theoretical basis for iterative algorithms with oversampling so that the iterative algorithms very probably converge to a unique non-negative or complex electron density with R -index = 0. In Sect. 2.2, a container is used to contain the membrane and solution. The solution may be frozen to prevent radiation damage to the membrane, for example a macromolecule. The membrane may also be embedded in a liquid which is then solidified (Sect. 2.3). Iterative algorithms with oversampling may also be applied to a membrane; a container or solidified liquid is not employed (Sect. 2.4).

Over-determined condition for complex object is discussed in Sect. 3.

2.1. Phase-retrieval method.

This is Yuen (2011c), (2011d), (H) for planar periodic membrane (a two-dimensional periodic object).

For the real part and the imaginary part together, we have a constraint:

(i) the size and the regions of zeros (not isolated points) of the real part and the imaginary part are the same. This is a very strong constraint.

The membrane is periodic in x_1 and x_2 with periods a and b respectively for both the real and imaginary parts.

For the real part, constraints used:

(ii) finite size with known unit cell constants, a, b, α . $f_{r0}(x_1, x_2, x_3) = 0$ for $x_1 < 0$ or $x_1 > a$, $x_2 < 0$ or $x_2 > b$. Shape and dimensions of the top and bottom boundary surfaces of the membrane will be determined.

(iii) $f_{r0}(x_1, x_2, x_3)$ is non-negative

(iv) $f_{r0}(x_1, x_2, x_3)$ is bounded

(v) sharp boundary. $f_{r0}(x_1, x_2, x_3) > 0$ almost everywhere at the six boundary surfaces of one period of the membrane

(vi) $f_{r0}(x_1, x_2, x_3)$ is continuous with

$f_{r0}(0, x_2, x_3) = f_{r0}(a, x_2, x_3)$ for all x_2, x_3

$f_{r0}(x_1, 0, x_3) = f_{r0}(x_1, b, x_3)$ for all x_1, x_3

(vii) symmetry elements, if applicable

with similar constraints for the imaginary part $f_{i0}(x_1, x_2, x_3)$.

For a non-centrosymmetric non-negative membrane, use constraints (ii) – (vii) (Yuen (2011c), (H)).

Because of the two-dimensional periodic structure, the diffracted intensity is greatly enhanced to form strong diffraction rods (Bragg rods) which can be measured accurately. Let $\omega_{10} = 2\pi / a$, $\omega_{20} = 2\pi / b$. For all l and m, $\left|F_0(l\omega_{10}, m\omega_{20}, \omega_3)\right|$ are measured experimentally.

$$F_0(l\omega_{10}, m\omega_{20}, \omega_3)$$

$$= \int \left\{ \int_0^a \int_0^b f_0(x_1, x_2, x_3) \exp[-j(l\omega_{10}x_1 + m\omega_{20}x_2)] dx_1 dx_2 \right\} \exp(-j\omega_3 x_3) dx_3 \tag{1}$$

where the triple integral is over one period of the membrane.

For fixed l' and m', the term in the curly brackets, $q(l'\omega_{10}, m'\omega_{20}, x_3)$, is a one-dimensional complex object of x_3. This is a one-dimensional problem. Constraints used:

(ii) $q(l'\omega_{10}, m'\omega_{20}, x_3)$ has finite size which will be determined

(iv) $q(l'\omega_{10}, m'\omega_{20}, x_3)$ is bounded

(vi) $q(l'\omega_{10}, m'\omega_{20}, x_3)$ is continuous

Apply the method in Yuen (2011d), (A). Use $|F_0(l'\omega_{10}, m'\omega_{20}, \omega_3)|$ for all ω_3. Find the possible $q(l'\omega_{10}, m'\omega_{20}, x_3)$ satisfying these three constraints. $q(l'\omega_{10}, m'\omega_{20}, x_3) = 0$ at its two boundary points along the x_3 axis. Since it does not have the strong constraints of known size and sharp boundary, the number of possible $q(l'\omega_{10}, m'\omega_{20}, x_3)$ satisfying these three constraints, N is very large. Use the Fourier transform $F_0(l'\omega_{10}, m'\omega_{20}, \omega_3)$ in (1) for each possible $q(l'\omega_{10}, m'\omega_{20}, x_3)$. Denote these by ${}^1F_0(l'\omega_{10}, m'\omega_{20}, \omega_3)$,…, ${}^NF_0(l'\omega_{10}, m'\omega_{20}, \omega_3)$. The Fourier transform $F_0(\omega_1, \omega_2, \omega_3)$ of $f_0(x_1, x_2, x_3)$ belongs to an entire function. For all l and m, associate the N functions ${}^1F_0(l'\omega_{10}, m'\omega_{20}, \omega_3)$,…, ${}^NF_0(l'\omega_{10}, m'\omega_{20}, \omega_3)$ from individual l' and m' so that they belong to N continuous functions ${}^1F_0(\omega_1, \omega_2, \omega_3)$,…, ${}^NF_0(\omega_1, \omega_2, \omega_3)$. (If a and b are large, ω_{10} and ω_{20} are small. This association will be easier. If ω_{10} or ω_{20} is large, there may be ambiguity, resulting in more than N functions). From these, get ${}^1q(l\omega_{10}, m\omega_{20}, x_3)$,…,

${}^{N}q(l\omega_{10}, m\omega_{20}, x_3)$.

Now $f(x_1, x_2, x_3)$ is periodic in x_1 and x_2. For fixed x_3, express $f(x_1, x_2, x_3)$ as a Fourier series of x_1 and x_2.

The Fourier coefficient

$$= \frac{1}{ab} \int_0^a \int_0^b f_0(x_1, x_2, x_3) \exp[-j(l\omega_{10}x_1 + m\omega_{20}x_2)] dx_1 dx_2$$

$$= \frac{1}{ab} q(l\omega_{10}, \omega_{20}, x_3)$$

Hence

$$f(x_1, x_2, x_3) = \frac{1}{ab} \sum_{l=-\infty}^{\infty} \sum_{m=-\infty}^{\infty} q(l\omega_{10}, m\omega_{20}, x_3) \exp[j(l\omega_{10}x_1 + m\omega_{20}x_2)]$$

For each $q(l\omega_{10}, m\omega_{20}, x_3)$ and all x_3, obtain a possible structure $f(x_1, x_2, x_3)$.

As shown in the Appendix of Yuen (2011c), it is very probable that only one of the N possible $f_0(x_1, x_2, x_3)$ satisfies constraint (i), and its real and imaginary parts each satisfying the constraints (ii) to (vii). Thus the structure $f(x_1, x_2, x_3)$ including its shape and dimensions is determined.

Note that this is a case of determining unique periodic structures without using interpolation. The structure obtained is exact.

2.2. Iterative algorithms with oversampling. Use a container

The phase-retrieval method (Sect. 2.1) is time-consuming. However, it provides the theoretical foundation and serves as a theoretical basis for iterative algorithms with oversampling.

Use Yuen (2011f), (J) to determine the maximum thickness of the membrane.

As in Yuen (2011g), Sect. 2.3, use an orthogonal container to contain the membrane and solution. The solution is frozen at very low temperature. The molecules of the solution have random locations and orientations. It constitutes a background of constant electron density in comparison to the strong diffraction rods of the membrane. Likewise, the walls of the container have constant electron density. The basic unit cell consists of one period of the membrane plus the frozen solution and the top and bottom of the container. The frozen solution and the container provide the six sharp boundary surfaces. The atomic coordinates of the membrane in the basic unit cell will be determined. Mathematically add replica of the basic unit cell successively to give a crystal. Yuen (2011e) can then be applied.

The | Fourier coefficients | for all (l,m,n) and in-between | Fourier coefficients | for $(l,\ m,\ n+\frac{1}{2})$ are obtained from the experimentally measured intensity of the container plus the membrane and solution. The other in-between | Fourier coefficients | are obtained by interpolation in multidimensions.

If we use:

1. the known value of the maximum thickness of a membrane. This is a loose support.
2. the experimentally determined dimensions of the basic unit cell, and | Fourier coefficients | for (l,m,n) and in-between | Fourier coefficients | for $(l,\ m,\ n+\frac{1}{2})$ obtained from the experimentally measured diffraction intensity of the container plus the membrane and solution,
3. random initial phases,
4. oversampling ratio $=2^3=8$; use | Fourier coefficients | and in-between | Fourier coefficients | for all (l,m,n),

   and all $(l+\frac{1}{2}$, and/or $m+\frac{1}{2}$, and/or $n+\frac{1}{2})$.

   Some of them are experimental data (see 2.), the rest are obtained by interpolation in multidimensions.
5. the six constraints for non-centrosymmetric non-negative membrane or thirteen

constraints for complex membrane in Sect. 2.1,

6. in the final iterations, the interpolated in-between | Fourier coefficients | are relaxed or adjusted, within their uncertainties, to satisfy the consistent condition (Yuen (2011a), Sect. 2.1); the interpolated in-between | Fourier coefficients | are improved,

the iterative algorithms will converge to a non-negative or complex electron density with R -index = 0. The electron density is very probably unique for this set of | Fourier coefficients | and in-between | Fourier coefficients | .

Note that we have used the constraint of sharp boundary for both the unit cell and the membrane. To compromise that proper initial phases are not used, we use known value of the maximum thickness of the membrane, and larger oversampling ratio $= 2^3 = 8$; use | Fourier coefficients | and in-between | Fourier coefficients | for all (l, m, n), and all ($l + \frac{1}{2}$, and/or $m + \frac{1}{2}$, and/or $n + \frac{1}{2}$). We hope that these will help the iterative algorithms to converge to a unique non-negative or complex electron density with R -index = 0. This convergence can be investigated empirically and theoretically.

As shown by the phase-retrieval method in Sect. 2.1, the interpolated in-between | Fourier coefficients | are, in fact, not needed. After the iterations converge to a unique non-negative electron density, we stop using all the interpolated in- between | Fourier coefficients | . The iterative algorithms will converge to a **unique** non-negative or complex electron density.

Radiation damage to the membrane, for example a macromolecule, may be prevented by the frozen solution.

2.3. Iterative algorithms with oversampling. Embedded in a solidified liquid

The membrane, for example a macromolecule, can be embedded in a liquid in a rectangular container, which is then solidified. The rectangular block containing one period of the membrane plus the solidified liquid forms a basic unit cell. The atomic coordinates of the membrane in the basic unit cell will be determined as in Sect. 2.2. Radiation damage to the macromolecule is prevented by the solid block.

Parallelpiped of the solid may be used if the iterations can converge more efficiently.

2.4. Iterative algorithms with oversampling. Membrane

We may just use a membrane; a container or solidified liquid is not employed.

Use

1. the maximum thickness of the membrane (Yuen (2011f), (J)), as a loose support,
2. the known unit cell constants, the | Fourier coefficients | and in-between | Fourier coefficients | obtained from the experimentally measured diffraction intensity of the membrane,
3. random initial phases,
4. oversampling ratio $=2^3=8$; use | Fourier coefficients | and in-between | Fourier coefficients | for all (l,m,n),

   and all ($l+\frac{1}{2}$, and/or $m+\frac{1}{2}$, and/or $n+\frac{1}{2}$).

   Some of them are experimental data (see 2.), the rest are obtained by interpolation in multidimensions.
5. the six constraints for non-centrosymmetric non-negative membrane or thirteen constraints for complex membrane in Sect. 2.1.
6. in the final iterations, the interpolated in-between | Fourier coefficients | are relaxed or adjusted, within their uncertainties, to satisfy the consistent condition (Yuen (2011a), Sect. 2.1); the interpolated in-between | Fourier coefficients | are improved,

the iterative algorithms will converge to a non-negative or complex electron density with R -index = 0. The electron density is very probably unique for this set of | Fourier coefficients | and in-between | Fourier coefficients | .

To compromise that proper initial phases are not used, we use known value of the maximum thickness of the membrane, and larger oversampling ratio $=2^3=8$; use | Fourier coefficients | and in-between | Fourier coefficients | for all (l,m,n), and all ($l+\frac{1}{2}$, and/or $m+\frac{1}{2}$, and/or $n+\frac{1}{2}$). We hope that these will help the iterative algorithms to converge to a unique non-negative or complex electron density with R -index = 0. This convergence can be investigated empirically and theoretically.

As shown by the phase-retrieval method in Sect. 2.1, the interpolated in-between ｜Fourier coefficients｜ are, in fact, not needed. After the iterations converge to an electron density, we stop using all the interpolated in-between ｜Fourier coefficients｜. The iterative algorithms may converge to a unique non-negative or complex electron density.

3. Over-determined condition for complex object

It is mentioned in Yuen (2011d) that for complex object, the system may be over-determined. If the constraint of sharp boundary may be disregarded, container, solution or solidified liquid are not needed. Replicas of the membrane can be added mathematically to form a three-dimensional crystal of parallel membranes. Procedures similar to Yuen (2011a), Sect. 2.2, 2.3 can then be applied.

4. Discussions and conclusion

The phase-retrieval method in Sect. 2.1 provides the theoretical foundation and gives the conditions that iterative algorithms with oversampling will very probably converge to a unique solution. A container may be used to contain the membrane and solution. The solution may be frozen to prevent radiation damage to the membrane, for example, a macromolecule. The membrane may also be embedded in a liquid which is then solidified.

Because of the two-dimensional periodic structure, the diffracted intensity is greatly enhanced to form strong diffraction rods. As the oversampling ratio is exactly 2, for a planar periodic membrane, no interpolation is needed. The intensity along the diffraction rods can be measured experimentally. The electron density obtained is unique and exact. This is an important case of determining unique periodic structures without using interpolation. By means of iterative algorithms with oversampling, electron density with R -index = 0 can be obtained. The *ab initio* electron density is determined by the experimental diffraction intensities alone; no other chemical or physical information (for example, atomicity, chemical formula, bond length, bond angle, etc) is used. It can be regarded as a fundamental property of the membrane. It is just a mathematical transformation of the diffraction intensities to an internal property of the membrane. Theories of molecular structure and properties (for example, quantum theory) should agree with this internal property. It forms a base for interpretation of experimental data. From this electron density, atomic coordinates are obtained. The atomic coordinates can be compared with those obtained from spectroscopy.

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18

Experimental measurement of the non-Bragg intensities of a thin periodic membrane and a crystal

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Abstract
An attempt is presented for measuring the non-Bragg intensities by employing a thin periodic membrane, or layers of a crystal. These are two-dimensional crystals. The weak diffracted waves are collected into strong diffraction rods, and can be measured accurately.

Keywords: In-between ｜Fourier coefficients｜ ; mid-Bragg intensities; thin periodic membrane; layers of a crystal; two-dimensional crystal; diffraction rods; Bragg rods; phase-retrieval method; consistent condition.

1. Introduction

By means of phase-retrieval method, electron density is obtained directly from diffraction intensities in X-ray crystallography. The in-between ｜Fourier coefficients｜ are employed (Yuen (2011a, 2011c, 2011d)). Instead of obtaining the in-between ｜Fourier coefficients｜ by interpolation in multidimensions (Bates, 1984, p. 216; Kincaid & Cheney, 1996, Sect. 6.10; Cheney & Light, 2000; Anita, 2002, Sect. 4.7, 4.8, Hau-Riege et al., 2004), a better way is to measure the non-Bragg intensities experimentally. Kaufmann (1985), p. 155 proposes a method of focusing incident X-ray on a few unit cells in a thin monocrystalline film, with a diameter of about 100 times the X-ray wavelength. Inspired by this proposal, this article presents an attempt to measure the non-Bragg intensities by means of a thin periodic membrane (Sect. 2.1), or layers of crystal (Sect. 2.2). These are two-dimensional crystals. The weak diffracted waves are collected into strong diffraction rods (Bragg rods), and can be measured accurately.

2.1. Non-Bragg intensities of a thin periodic membrane (a two-dimensional crystal)

This is Yuen (2011c, 2011d), (H). The thickness of the membrane consists of one or more layers. This thickness can be measured as in Yuen (2011f), (J). A parallel beam incident on the film or membrane is needed. The diffracted waves are collected into strong diffraction rods. They can be measured accurately.

2.2. Non-Bragg intensities of layers of a crystal

Consider a crystal with the longest unit cell edge along the, say, c axis. The X-ray source is collimated such that a parallel beam is incident on N layers of the crystal, parallel to the ab plane. N is a small number. Hence the object is a two-dimensional crystal. The diffracted wave is given by

$$F(h,k,\omega_3)$$

$$=\int_0^a\int_0^b\left\{\int f(x_1,x_2,x_3)\exp(-i\omega_3 x_3)dx_3\right\}\exp[-i(h\omega_{10}x_1+k\omega_{20}x_2)]dx_1dx_2,$$

where $\omega_{10}=2\pi/a$, $\omega_{20}=2\pi/b$.

The integrals are over the volume of N unit cells.

The diffracted waves are collected into strong diffraction rods, and can be measured accurately. For each reciprocal lattice point h,k, $|F_0(h,k,\omega_3)|$ can be measured.

These include the Bragg intensities and non-Bragg intensities. The in-between | Fourier coefficients | or mid-Bragg intensities occur at odd multiples of $\omega_3=2\pi/(2c)$. The contributions from adjacent layers to the in-between | Fourier coefficients | cancel each other. When N is even, all the in-between | Fourier coefficients | are equal to zero. When N is odd, the net contribution of the N layers to the in-between | Fourier coefficients | is one layer. From two slit-widths of the collimator where all the in-between | Fourier coefficients | equal to zero, a slit-width such that the X-ray is incident on an odd number of layers can be found. Hence the in-between | Fourier coefficients | can be measured. In adjusting the slit-width, ultimately it may be achieved that the X-ray is incident on only one layer, and the diffraction from this layer is measured. Hence the non-Bragg intensities for a monolayer two-dimensional crystal is found.

Diffraction from this monolayer,

$$F(h,k,\omega_3)$$

$$= \int \left\{ \int_0^a \int_0^b f(x_1,x_2,x_3) \exp[-i(l\omega_{10_1} x + m\omega_{20} x_2)] dx_1 dx_2 \right\} \exp(-i\omega_3 x_3) dx_3$$

where the integrals are over the volume of one unit cells.

The term in curly brackets is a one dimensional function of x_3 in one period.

Different layers of thickness of one layer of the three-dimensional crystal, but displaced from each other, will have different diffraction curves. This is because, in a one-dimensional periodic function, we can choose an arbitrary interval with length equal to the period as a unit cell. Hence for different displacement, there are different curves of non-Bragg intensities (including in-between | Fourier coefficients | or mid-Bragg intensities), while the Bragg intensities (or | Fourier coefficients |) are the same. For each displacement, there is a set of in-between | Fourier coefficients | . Therefore, for a three-dimensional crystal, there is not a single curve of non-Bragg intensities, and there are sets of in-between | Fourier coefficients | .

In one-, two- and three-dimensions (Yuen (2011a), (B), (D), Yuen (2011c, 2011d), (B), (D), (E), (F) and (H)), the oversampling ratio is exactly 2. Hence only the in-between

| Fourier coefficients | for ω_3 are needed for phase retrieval. Bragg intensities are also measured. They can be compared to those when the whole three-dimensional crystal is irradiated by X-ray. These are used for scaling purpose, and for reference when a crystal is damaged by radiation and is replaced by another crystal.

All the other in-between | Fourier coefficients | can be measured by using various orientations of the crystal planes. These will be used in the iterative algorithms with oversampling in Yuen (2011b, 2011e). Moreover, as only the in-between | Fourier coefficients | for, say, ω_3 are needed for phase retrieval, values of the | Fourier coefficients | and in-between | Fourier coefficients | for the three indices l, m and n are not independent. They must be consistent so that the electron densities obtained for each index are identical. This consistent condition must be satisfied (Yuen (2011b), Sect. 2.1). Hence all the | Fourier coefficients | and in-between | Fourier coefficients | are adjusted within their experimental uncertainties so that the electron densities obtained for each index are identical. These adjustments can be done by means of iterative algorithms with oversampling (Yuen (2011b), Sect. 2.3, 2.4 and Yuen (2011e), Sect. 2.2, 2.3).

To develop the technology, we may first apply this method to crystals with large unit cells. Examples:

cowpea mosaic virus (CPMV) (Lin et al., 1999)
  cubic $a = 317$ Å
  hexagonal $a = 451$ Å, $c = 1038$ Å
HK97 mature empty capsids (Wikoff et al., 2000)
  monoclinic $a = 580$ Å, $b = 625$ Å, $c = 790$ Å, $\beta = 90^\circ$
liver aldolase (B isozyme) (Dalby et al., 2001)
  monoclinic $a = 291.1$ Å, $b = 489.8$ Å, $c = 103.4$ Å, $\beta = 103.6^\circ$

2.3. Technology

To adjust the two edges of the collimator so that they are parallel to each other should not be difficult as it involves adjustment of orientations only; the smallness of the slit-width has no effect. Similarly, the orientation of the crystal can be adjusted so that its layers are parallel to the laser beam. When the parallel laser beam is incident on a two-dimensional crystal, as in Sect. 2.2, the diffracted waves are collected into strong diffraction rods. The thickness of the layers can be measured as in Yuen (2011f), (J).

$\boldsymbol{\omega}$ = wavevector of scattered wave – wavevector of incident wave

Since $\boldsymbol{\omega}$ is normal to the layers of the crystal and the incident wave is along the layers, the wavevector of scattered wave should be close to the layers. According to the resolution needed, we use all $\boldsymbol{\omega}$, $|\boldsymbol{\omega}| \le \omega_{\max}$ (Yuen (2011c), Sect. 2.3). In order to achieve this, laser beams of all wavelengths $\ge 2\pi / \omega_{\max}$ should be used.

At present it may be difficult to adjust the slit width by angstroms. This measuring technology needs to be developed. Before this technology is developed, we can use perhaps random positions of one side of the collimator, and a very weak laser beam. The thickness of one layer of the crystal can be obtained from the unit cell constants determined in three-dimensional X-ray crystallography. From the thickness of the laser beam, for each position of one side of the collimator, we know whether an even or odd number of the layers are irradiated by the laser beam. When an odd number of the layers are irradiated, the intensity of the laser beam can be increased, and the in-between | Fourier coefficients | (mid-Bragg intensities) measured. When the laser beam is incident on only one layer, the non-Bragg intensities for a monolayer two-dimensional crystal is obtained.

3. Discussions and conclusion

The in-between | Fourier coefficients | are very important quantities. In the future, they may be included in articles in X-ray crystallography. One purpose of this article is to stimulate interest in seeking for methods for measuring the in-between | Fourier coefficients | or mid-Bragg intensities. Kaufmann (1985), p. 155 proposes a method of focusing incident X-ray on a few unit cells of a thin monocrystalline film. But no efficient focusing devices for X-rays seem to exist. Miao & Sayre (2000), p. 597 suggests that non-Bragg intensities of small crystal can be measured (mid-Bragg intensities observed in Chapman et al. (2011), p. 74). As the intensities are weak, a strong source needs to be used. This method suffers from radiation damage of biological specimens.

In Sect. 2.1, it is suggested a thin membrane (a two-dimensional crystal) is used. The weak diffracted waves are collected into strong diffraction rods, and can be measured accurately. No focusing is used; the problem of focusing at a few unit cells in Kaufmann (1985), p. 155 is avoided.

In Sect. 2.2, it is suggested that small number of layers of a crystal can be used. This is a two-dimentional crystal. The weak diffracted waves are collected into strong diffraction rods. The number of unit cells in the two-dimensional crystal is large. The diffraction intensity is proportional to the square of the number of unit cells and is strong relative to the background. Strong X-ray source is not needed. The beam of X-ray is adjusted to have small width. The beam passing through the crystal is just like passing through a flat tunnel. The radiation damage to the specimen is, in fact, smaller than that when the whole three-dimensional crystal is irradiated. This is because the remaining portion of the crystal which is not irradiated, tends to maintain the crystallinity. This is different from imaging of biomolecules with femtosecond X-ray pulses (Neutze et al. (2000)). The number of unit cells irradiated needs to be known as this affects the intensity of the diffraction rods. Hence the shape of the crystal is measured. As the laser beam or the crystal is rotated so that all the diffraction rods are measured, the number of unit cells irradiated may vary. This can be avoided if whole layers of the crystal are irradiated.

If a crystal is damaged, it can be replaced by another crystal. As some Bragg intensities are measured for reference when a crystal is replaced by another crystal, we do not have the problem of random orientations (Huldt et al. (2003)). In general, another set of in-between｜Fourier coefficients｜ are measured. (For diffraction from a single layer, a different curve of non-Bragg intensities is measured). Displace the X-ray beam or the crystal so that the in-between｜Fourier coefficients｜ or mid-Bragg intensities for strong reflections are the same as those from the damaged crystal in order to have diffraction from the same layers.

According to the phase-retrieval theory, values of the in-between｜Fourier coefficients｜ for the three indices l, m and n are not independent. They must be consistent so that the electron densities obtained for each index are identical. This consistent condition must be satisfied (Yuen (2011b), Sect. 2.1). All the in-between ｜Fourier coefficients｜ need to be adjusted within their experimental uncertainties so that the electron densities obtained for each index are identical. The adjustments can be done by means of iterative algorithms with oversampling (Yuen (2011b), Sect. 2.3, 2.4

and Yuen (2011e), Sect. 2.2, 2.3).

The treatment in this article is related to that in Szöke (2001).

The measurement of the in-between | Fourier coefficients | or mid-Bragg intensitiesis very important. Researchers are welcome to develop the technology of the method proposed.

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19

Initial phases in determination of electron density of crystals and nonperiodic objects by means of iterative algorithms with oversampling

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Abstract

An attempt is presented for obtaining more realistic initial phases for iterative algorithms with oversampling. We employ atoms of various radii or electron density functions uniformly distributed in a unit cell or in sub-cells of the unit cell (initial phases obtained = 0 or π for real crystal), or use crystals of approximately same unit-cell shapes and dimensions, same space group and approximately same properties (initial phases obtained need not = 0 or π for real crystal). For nonperiodic object or crystal, phases of the minimum-phase function, obtained by Hilbert transform or discrete Hilbert transform, can also be used as initial phases. These initial phases are consistent with each other, and with the magnitudes $\left|F(\omega_1,\omega_2,\omega_3)\right|$. They may be better than the random initial phases.

Keywords: Phase retrieval; iterative algorithms; oversampling; realistic initial phases; macromolecular crystals; uniform distributions; unit-cell shapes; unit-cell dimensions; space groups; properties; Hilbert transform; discrete Hilbert transform; Shannon's sampling.

1. Introduction

To determine the electron density from the diffraction intensities, much work has been done by means of iterative algorithms and oversampling, and has gained much success empirically (see the references in the articles in Yuen (2011a, 2011c, 2011d, 2011e, 2011f, 2011g)). It is stated in the review of Thibault & Elser (2010), p. 244 that ‘It is probably fair to say that for every successful reconstruction, tens of diffraction patterns have failed to yield any results whatsoever’. Hence it is important to seek for procedures that will lead to the unique electron density.

The phase-retrieval method shows that the result is unique for centrosymmetric real crystal (Yuen (2011a), (D)), and very probably unique for non-centrosymmetric non-negative crystal (Yuen (2011d), (D)) or complex object with non-negative real and imaginary parts (Yuen (2011e), (D)). When iterative algorithms with oversampling are applied for determination of electron density of crystal structure which has not been solved, random initial phases are commonly employed. The iterations will converge to an electron density with R-index = 0. The electron density may be unique (Yuen (2011b), Sect. 2.4, Yuen (2011f), Sect. 2.3). But this is not guaranteed. No chemical information has been used used. In order to be more certain that we can obtain the correct electron density, a compromise is to use chemical informations.

To help the iterations to converge to the unique electron density, an important step is to use more realistic initial phases, instead of the random phases. A crude phase estimation for a nonperiodic object is presented and used in Bates (1982), Fright & Bates (1982), Garden & Bates (1982), Bates & Fright (1983), Bates (1984), Won et al. (1985), Bates & McDonnell (1986), p. 118, Gardenier et al. (1986) and Millane et al. (1986). Pérez-Ilzarbe (1992) uses an initial structure by filling the unit cell with uniformly distributed random numbers. In Helliwell et al. (2005), initial phases are derived from anomalous scattering. Initial phases may be derived from isomorphous replacement or from a partial structure (Rossmann (1990), Baker et al. (1993), Lilien et al. (2004), Qian et al. (2007)).

This article presents an attempt of obtaining initial phases for Fourier coefficients and in-between Fourier coefficients more realistic than random phases, via chemical informations. Analogous to the idea of atomicity constraint in crystallography or contrast constraints in diffraction microscopy (Thibault & Elser (2010), p. 242), in Sect. 2.1, we employ atoms of various radii or electron density functions uniformly distributed in a unit cell or in sub-cells of the unit cell. The initial phases obtained = 0 or π for real crystal. In Sect. 2.2, we use crystals of approximately same unit-cell shapes and dimensions, same space group, and approximately same properties. The initial phases obtained need not = 0 or π for real crystal.

For nonperiodic object or crystal, phases of the minimum-phase function, obtained by Hilbert transform or discrete Hilbert transform, can also be used as initial phases (Sect. 2.3). These initial phases are consistent with each other, and with the magnitudes $\left|F(\omega_1,\omega_2,\omega_3)\right|$. They may be better than the random initial phases.

2. Method.

2.1. Use uniformly distributed atoms

Choose a macromolecular crystal the structure of which has been solved by conventional method. Calculate the Fourier coefficients and in-between Fourier coefficients from the electron density in the basic unit cell. Use their phases as initial phases. Follow Yuen (2011f), Sect. 2.2, and apply iterative algorithms with oversampling. The iterations will converge to the electron density of this crystal.

Use the in-between | Fourier coefficients | calculated in above. We then proceed to develop a procedure of finding realistic initial phases, assuming that the crystal structure is not known. The total number of non-hydrogen atoms N in a unit cell is known. Distribute uniformly the atoms in the unit cell.

(A) Use spherical atoms of constant electron density and radius R. Obtain the phases of the Fourier coefficients and in-between Fourier coefficients for this crystal. This artificial crystal is centrosymmetric. The phases = 0 or π for real crystal. Use these as the initial phases for iterative algorithms with oversampling. Apply this to the crystal. See whether the iterations will converge to the known electron density. The value of the radius R can be adjusted for achieving such convergence, and achieving it efficiently. The iterations can be compared with those in above.

(B) The number of non-hydrogen atoms for each atomic species is known. Use the electron density functions corresponding to the atomic scattering factors in the International Tables for Crystallography (Brown et al. (2004)). Calculate the average electron density function for the N non-hydrogen atoms. Use atoms of this average electron density function. Obtain the phases of the Fourier coefficients and in-between Fourier coefficients for this crystal. This artificial crystal is centrosymmetric. The phases = 0 or π for real crystal. Use these as the initial phases for iterative algorithms with oversampling. Apply this to a crystal. See whether the iterations will converge to the known electron density. The shape of the electron density function can be adjusted for achieving such convergence, and achieving it efficiently. The iterations can be compared with those in above.

From (A) and (B), find the better way of obtaining initial phases for efficient convergence of the iterations.

Alternatively, divide the unit cell into M identical parallelpiped sub-cells. The three edge-lengths of a sub-cell are approximately the same. M is an integer closest to N. $M \leq$ or $> N$. For macromolecular crystals, N is very large. Although M may not be equal to N, the effect can be neglected. If $M \leq N$, all the sub-cells are filled. If $M > N$, some of the sub-cells are unfilled. Repeat the process in above. Find the better way of obtaining initial phases.

Repeat these for many macromolecular crystals.

2.2 Use crystals of approximately same unit-cell shapes and dimensions, same space group, and approximately same properties

To find initial phases of a macromolecular crystal, use another macromolecular crystal of approximately same unit-cell shapes and dimensions, the structure of which has been solved. It would be better if it has the same space group, and approximately same chemical constituents, density, etc. For examples, they have the same amount of heavy atoms. Calculate the Fourier coefficients and in-between Fourier coefficients of this second crystal. The phases of these Fourier coefficients and in-between Fourier coefficients form a consistent set in a sense that they come from an actual distribution of atoms. These phases need not 0 or π for a real crystal. Use these phases as the initial phases of the Fourier coefficients and in-between Fourier coefficients of the first crystal. Because we use a consistent set of initial phases to start the iterations, we hope that the iterations will lead to the correct electron density.

Define a *M*-index $= \dfrac{\sum_{hkl} w\left||F_{o1}| - |F_{o2}|\right|}{\sum_{hkl} w|F_{o1}|}$

where w is a weighting factor with more weights for stronger reflections (larger

$|F_{o1}|$ or $|F_{o2}|$),

$|F_o|$ are the observed | Fourier coefficients | (structure factors).

If the *M*-index is small, we expect that the two crystal structures closely resemble each other. The phases calculated from the second crystal will be good initial phases for the first crystal. If there is more than one crystal which has approximately same unit-cell shapes and dimensions, and same space group and approximately same properties, use the phases calculated from all these crystals as initial phases for the first crystal.

Apply this method to a large number of macromolecular crystals. We hope to establish a correlation of *M*-index and the goodness of the initial phases. Then for an unknown crystal structure, if we can find another crystal of approximately same unit-cell shapes and dimensions, same space group, approximately same properties, and an *M*-index below certain value, we know that this second crystal may provide a set of good initial phases.

In point-atom model of crystals, an atom is represented by a point atom $\delta(\mathbf{r}')$. It has bounded height H centered at a circle with radius R. The value of the atom in this circle, except the origin, is equal to zero.

$$\int \delta(\mathbf{r}')d\mathbf{r}' = 1$$

To obtain initial phases for point-atom models of crystals in Yuen (2011h, 2011i), the atoms in the second macromolecular crystal are replaced by point-atoms centered at the same atomic coordinates. The Fourier coefficients and in-between Fourier coefficients of this point-atom crystal are then calculated. Their phases are used as a consistent set of initial phases for the first macromolecular crystal.

2.3. Use the phase of the minimum phase function for a nonperiodic object or a crystal

Consider a three-dimensional nonperiodic object. As in Yuen (2011d), (10),

$$F(\omega_1,\omega_2,\omega_3)$$
$$=\int\left\{\iint f(x_1,x_2,x_3)\exp[-i(\omega_2x_3+\omega_3x_3)]dx_2dx_3\right\}\exp(-i\omega_1x_1)dx_1$$

where the integrals are over the volume of the object.

Denote the term in curly brackets by $q(x_1,\omega_2,\omega_3)$.

$$F(\omega_1,\omega_2,\omega_3)=\int q(x_1,\omega_2,\omega_3)\exp(-i\omega_1x_1)dx_1$$

For a fixed $\omega_2=\omega_2'$, $\omega_3=\omega_3'$,

$$F(\omega_1,\omega_2',\omega_3')=\int q(x_1,\omega_2',\omega_3')\exp(-i\omega_1x_1)dx_1$$

$q(x_1,\omega_2',\omega_3')$ is a one-dimensional complex object.

Use the Hilbert transform in Yuen (2011d), (5) to obtain the phase $\phi_M(\omega)$ of the minimum-phase function, from $|F(\omega_1,\omega_2',\omega_3')|$. This is used as the initial phase for $F(\omega_1,\omega_2',\omega_3')$. For all ω_2, ω_3, obtain the initial phases for $F(\omega_1,\omega_2,\omega_3)$, from $|F(\omega_1,\omega_2,\omega_3)|$. These initial phases are consistent with each other, and with the magnitudes $|F(\omega_1,\omega_2,\omega_3)|$. They may be better than the random initial phases. But this has to be checked by many tests.

For a crystal, obtain the in-between | Fourier coefficients | by interpolation in multidimensions or by experimental measurement. Use the discrete Hilbert transform and Shannon's sampling in Yuen (2011d), (8) and (9) respectively, to obtain the phase of the minimum-phase function.

Replicas of a nonperiodic object of finite size can be added successively to form a crystal. Discrete Hilbert transform can then be employed.

3. Discussions and conclusion

Employment of atoms in the unit cell may be more realistic than using random initial phases. In Sect. 2.1, the initial phases (= 0 or π for real crystal) are obtained from a uniform distribution of atoms. In Sect. 2.2, the initial phases (need not = 0 or π for real crystal) are obtained from a crystal in reality. If the initial distribution of atoms corresponding to the initial phases is the same as the actual distribution, the correct initial phases are indeed used. Crystals in reality are regarded as perturbations. If the perturbation is small, the process in Sect. 2 works. In general, the perturbation is not small. However, the initial phases are consistent in a sense that they come from a uniform or actual distribution of atoms. Crystals in reality have atoms distributed roughly uniformly; there are no large voids in a crystal, which contain no atoms. The initial distribution of the atoms may not be close to the actual distribution. But because we use a consistent set of initial phases to start the iterations, we hope that the iterations will lead to the correct electron density. This remains to be investigated empirically and theoretically.

For nonperiodic object or crystal, phases of the minimum-phase function, obtained by Hilbert transform or discrete Hilbert transform can also be used as initial phases. These initial phases are consistent with each other, and with the magnitudes $|F(\omega_1,\omega_2,\omega_3)|$. They may be better than the random initial phases. This needs to be checked by many tests.

It is important to find a routine and efficient method of determining the electron densities of macromolecular crystals or nonperiodic objects. The choice of proper initial phases is very important. It directly affects the convergence of the iterations to the correct result. The primary goal of this article is to obtain a set of initial phases better than the random initial phases. This is the first step. If this is achieved, we can proceed to find a consistent set of initial phases so that the iterations will converge to the correct result. Various methods of obtaining more realistic or consistent set of initial phases may be investigated. All the chemical and physical knowledge of the crystal, for examples, atomicity, non-negativity, interatomic distances, structural informations, bond lengths, bond angles, may be employed. May be a probabilistic approach like the direct method can provide a consistent set of initial phases.

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20

Phases of reflections normal to a mirror plane of a real object can be determined uniquely

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For real nonperiodic object or crystal with a mirror plane, phases of reflections normal to the plane mirror can be obtained uniquely from the intensities by means of phase-retrieval method.

Keywords: Phase-retrieval method; initial phases; nonperiodic object; crystal; fibre; membrane.

1. Introduction

For iterative algorithms with oversampling and the direct methods in X-ray crystallography, initial phases are needed (Ladd & Palmer, 1993, Ch. 7; Woolfson & Fan, 1995, Ch. 3). For real nonperiodic object or periodic object with a mirror plane, phases of reflections normal to the mirror plane can be obtained uniquely from the intensities by means of phase-retrieval method.

2. Phase-retrieval method

(A) Nonperiodic object with a mirror plane

Constraints used:

(i) finite size. $f(x_1, x_2) = 0$ for $|x_1| > a/2$ or $x_2 < H(x_1)$ or $x_2 > K(x_1)$. Values of a, $H(x_1)$ and $K(x_1)$ need not be known.

(ii) $f(x_1, x_2)$ is real

(iii) The plane $x_1 = 0$ is a mirror plane, $f(x_1, x_2) = f(-x_1, x_2)$ for all x_2,

(iv) $f(x_1, x_2)$ is bounded.

$$F(\omega_1, \omega_2) = \int_{-a/2}^{a/2} \left\{ \int f(x_1, x_2) \exp(-i\omega_2 x_2) dx_2 \right\} \exp(-i\omega_1 x_1) dx_1$$

$$= \int_{-a/2}^{a/2} q(x_1, \omega_2) \exp(-i\omega_1 x_1) dx_1$$

where the double integral is over the object.

For fixed $\omega_2 = 0$, the term in curly brackets, $q(x_1, 0)$ can be regarded as an object with size from $x_1 = -a/2$ to $x_1 = a/2$. $q(x_1, 0)$ is a real even function of x_1. This is a one-dimensional problem. Following Yuen (2011a), (A), phase of $F(\omega_1, 0)$ can be obtained uniquely from $|F(\omega_1, 0)|$ for all ω_1.

Consider a three-dimensional nonperiodic object with a mirror plane $x_1 = 0$. Constraints used:

(i) $f(x_1, x_2, x_3)$ has finite size $-a/2 \le x_1 \le a/2$, $H(x_1) \le x_2 \le K(x_1)$, $L(x_1, x_2) \le x_3 \le M(x_1, x_2)$. Values of a, $H(x_1)$, $K(x_1)$, $L(x_2, x_3)$ and $M(x_2, x_3)$ need not be known.

(ii) $f(x_1, x_2, x_3)$ is real

(iii) The plane $x_1 = 0$ is a mirror plane $f(x_1, x_2, x_3) = f(-x_1, x_2, x_3)$ for all x_2, x_3

(iv) $f(x_1, x_2, x_3)$ is bounded.

$F(\omega_1, \omega_2, \omega_3)$

$$= \int_{-a/2}^{a/2} \left\{ \iint f(x_1, x_2, x_3) \exp\left[-i(\omega_2 x_2 + \omega_3 x_3)\right] dx_2 dx_3 \right\} \exp(-i\omega_1 x_1) dx_1$$

$$= \int_{-a/2}^{a/2} q(x_1, \omega_2, \omega_3) \exp(-i\omega_1 x_1) dx_1$$

where the triple integral is over the object.

For fixed $\omega_2 = \omega_3 = 0$, the term in curly brackets, $q(x_1, 0, 0)$ is a one-dimensional object with size from $x_1 = -a/2$ to $x_1 = a/2$. $q(x_1, 0, 0)$ is a real even function of x_1. Following Yuen (2011a), (A), phase of $F(\omega_1, 0, 0)$ can be obtained uniquely from $|F(\omega_1, 0, 0)|$ for all ω_1.

If, say, $x_2 = 0$ is also a mirror plane, phase of $F(0, \omega_2, 0)$ can also be obtained uniquely from $|F(0, \omega_2, 0)|$ for all ω_2.

(B) Periodic object with a mirror plane

Choose the cell, $-a/2 \le x_1 \le a/2$, $0 \le x_2 \le b$, as the basic unit cell.

Constraints used:

(i) $f_0(x_1, x_2)$ has finite size with known unit cell constants, $a, b, \alpha = \pi/2$.

(ii) $f_0(x_1, x_2)$ is real

(iii) The plane $x_1 = 0$ is a mirror plane, $f_0(x_1, x_2) = f_0(-x_1, x_2)$ for all x_2

(iv) $f_0(x_1, x_2)$ is bounded

Let $\omega_{10} = 2\pi/a$, $\omega_{20} = 2\pi/b$.

$$F_0(h\omega_{10}, k\omega_{20}) = \int_{-a/2}^{a/2} \left\{ \int_0^b f_0(x_1, x_2) \exp(-ik\omega_{20} x_2) dx_2 \right\} \exp(-ih\omega_{10} x_1) dx_1$$

$$= \int_{-a/2}^{a/2} q(x_1, k\omega_{20}) \exp(-ih\omega_{10}x_1)dx_1$$

For fixed $k = 0$, $q(x_1,0)$ is a real even function of x_1. $\left|F_0(h\omega_{10},0)\right|$, $h = 0,\pm1,\pm2.,...$, are measured experimentally. Measure experimentally $\left|F_0((h+1/2)\omega_{10},0)\right|$, $h = 0,\pm1,\pm2.,...$, (Yuen, 2011b) or obtain these in-between | Fourier coefficients | by interpolation in multidimensions (Bates, 1984, p. 216; Kincaid & Cheney, 1996, Sect. 6.10; Cheney & Light, 2000; Anita, 2002, Sect. 4.7, 4.8, Hau-Riege et al., 2004). Following Yuen (2011a), (B), phase of $F_0(h\omega_{10},0)$ can be obtained uniquely from $\left|F_0(h\omega_{10},0)\right|$, $\left|F_0((h+1/2)\omega_{10},0)\right|$ for all h.

Consider a three-dimensional periodic object with a mirror plane $x_1 = 0$. Choose the cell, $-a/2 \le x_1 \le a/2$, $0 \le x_2 \le b$, $0 \le x_3 \le c$, as the basic unit cell.

Constraints used:

(i) $f_0(x_1, x_2, x_3)$ has finite size $-a/2 \le x_1 \le a/2, 0 \le x_2 \le b, 0 \le x_3 \le c$ with known unit cell constants, $a, b, c, \alpha, \beta = \pi/2, \gamma = \pi/2$.

(ii) $f_0(x_1, x_2, x_3)$ is real

(iii) The plane $x_1 = 0$ is a mirror plane

(iv) $f_0(x_1, x_2, x_3)$ is bounded

$$F_0(h\omega_{10}, k\omega_{20}, l\omega_{30})$$

$$= \int_{-a/2}^{a/2} \left\{ \int_0^b \int_0^c f_0(x_1, x_2, x_3) \exp[-i(k\omega_{20}x_2 + l\omega_{30}x_3)]dx_2 dx_3 \right\} \exp(-ih\omega_{10}x_1)dx_1$$

$$= \int_{-a/2}^{a/2} q(x_1, k\omega_{20}, l\omega_{30}) \exp(-ih\omega_{10} x_1) dx_1$$

For fixed $k = l = 0$, $q(x_1, 0, 0)$ can be regarded as an object with size from $x_1 = -a/2$ to $x_1 = a/2$. $q(x_1, 0, 0)$ is a real even function of x_1. This is a one-dimensional problem. $|F_0(h\omega_{10}, 0, 0)|$, $h = 0, \pm 1, \pm 2., ...$, are measured experimentally. Measure experimentally $|F_0((h + 1/2)\omega_{10}, 0, 0)|$, $h = 0, \pm 1, \pm 2., ...$, or obtain these in-between | Fourier coefficients | by interpolation in multidimensions. Following Yuen (2011a), (B), phase of $F_0(h\omega_{10}, 0, 0)$ can be obtained uniquely from $|F_0(h\omega_{10}, 0, 0)|$, $|F_0((h + 1/2)\omega_{10}, 0)|$ for all h. That is, phases of reflections $h00$ for all h are obtained.

If, say, $x_2 = 0$ is also a mirror plane, phase of $F_0(0, k\omega_{20}, 0)$ can also be obtained uniquely from $|F_0(0, k\omega_{20}, 0)|$, $|F_0(0, (k + 1/2)\omega_{20}, 0)|$ for all k. That is, phases of reflections $0k0$ for all k are obtained.

(C) General fibre

Fibre has circular symmetry in the $x_1 x_2$ plane with radius $R(x_3)$, and is periodic in x_3 with period c. Choose the cell $(x_1{}^2 + x_2{}^2)^{1/2} \le R(x_3)$, $0 \le x_3 \le c$, as the basic unit cell.

Constraints used:

(i) For each x_3, $f_0(x_1, x_2, x_3)$ has finite radius $R(x_3)$ in the x_1x_2 plane. $f_0(x_1, x_2, x_3) = 0$ for $x_3 < 0$ or $x_3 > c$. Value of c is known. Value of $R(x_3)$ needs not be known

(ii) $f_0(x_1, x_2, x_3)$ is real

(iii) $f_0(x_1, x_2, x_3)$ is circularly symmetric in the x_1x_2 plane. Hence $x_1 = x_2 = 0$ are mirror planes.

(iv) $f_0(x_1, x_2, x_3)$ is bounded

Let $\omega_0 = 2\pi / c$.

$$F_0(\omega_1, \omega_2, l\omega_0)$$

$$= \int \left\{ \int \int_o^c f_0(x_1, x_2, x_3) \exp[-i(\omega_2 x_2 + l\omega_0 x_3)] dx_2 dx_3 \right\} \exp(-i\omega_1 x_1) dx_1$$

$$= \int q(x_1, \omega_2, l\omega_0) \exp(-i\omega_1 x_1) dx_1$$

where the triple integral is over the basic unit cell.

Phase of $F_0(\omega_1, 0, 0)$ can be obtained uniquely from $|F_0(\omega_1, 0, 0)|$ for all ω_1. By rotating the coordinate system, $F_0(\omega_1, \omega_2, 0)$ for all ω_1, ω_2 are determined uniquely.

If, in addition, there is an additional constraint

(iii) $x_3 = 0$ is a mirror plane,

choose the cell $(x_1^2 + x_2^2)^{1/2} \le R(x_3)$, $-c/2 \le x_3 \le c/2$, as the basic unit cell.

Phase of $F_0(0, 0, l\omega_0)$ can also be obtained uniquely from $|F_0(0, 0, l\omega_0)|$ for all

ω_3.

(D) Fixed fibre molecule (that is, not freely rotating).

If it has one or more mirror planes, phase of $F_0(\omega_1,0,0)$, $F_0(0,\omega_2,0)$ or $F_0(0,0,l\omega_0)$ can be obtained uniquely from the magnitude.

(E) Planar periodic membrane

The object is periodic in x_1 and x_2 with periods a and b respectively. Let $\omega_{10}=2\pi/a$, $\omega_{20}=2\pi/b$. If it has one or more mirror planes, phase of $F_0(h\omega_{10},0,0)$, $F_0(0,k\omega_{20},0)$ or $F_0(0,0,\omega_3)$ can be obtained uniquely from the magnitude.

3. Discussions and conclusion

As the phase can only be 0 or π radians, the correct phase can be obtained even if there is experimental uncertainty or computational approximation (Berg & Goffeney, 1997, p. 622). For nonperiodic object, crystal, fibre or membrane with one or more mirror planes, the phases of the reflections $h00$, $0k0$ or $00l$, determined in this article, can be used as initial phases for iterative algorithms with oversampling or direct methods. These will be useful in the determination of structures of non-centrosymmetric crystals by means of iterative algorithms with oversampling or direct methods. For a solved crystal structure in the literature, the author may use the known | Fourier coefficients | and in-between | Fourier coefficients | , and apply the phase-retrieval method to obtain the phases of the reflections $h00$, $0k0$ or $00l$. The result can be compared with the phases reported.

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21

Equal point-atom model of crystals. Iterative algorithms with oversampling

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Abstract

This article develops the technology of iterative algorithms with oversampling that will lead to the unique electron density. It is achieved by means of a systematic study of the equal point-atom crystals containing only one atomic species. A powerful tool of determining crystal electron density and atomic coordinates via equal point-atom model in combination with iterative algorithms and oversampling is presented. The point-atom coordinates are first determined by large number of｜Fourier coefficients｜and initial in-between｜Fourier coefficients｜ with eleven to sixteen strong constraints. *Ab initio* electron density with R-index = 0 is then obtained. The in- between｜Fourier coefficients｜ are refined because of the consistent condition. The in-between｜Fourier coefficients｜ and｜Fourier coefficients｜ are adjusted within their uncertainties to obtain all the missed atoms and reasonable bond lengths and bond angles. This tool can be applied to macromolecular crystals.

Keywords: Crystallography; equal point atom; iterative algorithms; oversampling; excessively over-determined system; powerful point-atom constraint; low-density elimination; charge-flipping algorithms; electron density; atomic coordinates; consistent condition; partial structures; macromolecular crystals; missed atoms.

1. Introduction

To determine the electron density from the diffraction intensities, much work has been done by means of iterative algorithms and oversampling, and has gained much success empirically. However, ‘It is probably fair to say that for every successful reconstruction, tens of diffraction patterns have failed to yield any results whatsoever’ (Thibault & Elser (2010), p. 244). In Yuen (2011a), Sect. 2.4, Yuen (2011b), Sect. 2.3, Yuen (2011c), Sect. 2.3, Yuen (2011d), Sect. 2.3, and Yuen (2011e), Sect. 2.5.2, for centrosymmetric or non-centrosymmetric crystal or nonperiodic object, the electron density obtained by means of iterative algorithms and oversampling, may or may not be unique. But we are not sure about the result. Hence it is important to seek for procedures that will lead to the unique electron density. One approach is to use all chemical and physical informations.

There are many chemical and physical informations for crystals. Hence it is natural to first deal with crystals. In Sect. 2.1, we state that the many crystal structures solved by conventional methods are good candidates for developing the technology of iterative algorithms with oversampling that will lead to the unique electron density, and sixteen advantages are presented. After effective iterative algorithms with oversampling are developed for determining crystal electron densities, they can then be extended to diffractive microscopy or diffractive imaging of nonperiodic objects where many of these advantages are absent.

For a crystal, the structure factor

$$F(\mathbf{h}) = \sum f_j(\mathbf{h}) \exp[2\pi i(\mathbf{h}\cdot\mathbf{r}_j)] \tag{1}$$

where $f_j(\mathbf{h})$ is the atomic scattering factor of the j -th atom

$\mathbf{h}$ is a reciprocal space vector
$\mathbf{r}$ is position vector of the j -th atom

From (1), the structure factor

$$F(\mathbf{h}) = \sum f_j(\mathbf{h})\delta(\mathbf{r}'-\mathbf{r}_j) \exp[2\pi i(\mathbf{h}\cdot\mathbf{r}_j)] \tag{2}$$

where $\delta(\mathbf{r}')$ represents a point atom. It is bounded (Yuen (2011a), Sect. 2.1, constraint (iv), Yuen (2011d), Sect. 2.1, constraint (iii)), and

$$\int \delta(\mathbf{r}')d\mathbf{r}' = 1 \tag{3}$$

The atomic scattering factors have approximately same shapes (Harker & Kasper (1948)).

Let $\hat{f}(\mathbf{h}) = (\sum f_j(\mathbf{h}))/(\sum Z_j)$ (4)

where Z_j is the atomic number of the j -th atom.

$\hat{f}(\mathbf{h})$ is the atomic scattering factor of a hypothetical atom with $Z = 1$.

$$f_j(\mathbf{h}) \approx Z_j \hat{f}(\mathbf{h}) \tag{5}$$

Divide the structure factors (2) by $\hat{f}(\mathbf{h})$.

$$F(\mathbf{h}) / \hat{f}(\mathbf{h}) \approx \sum Z_j \delta(\mathbf{r}' - \mathbf{r}_j) \exp[2\pi i(\mathbf{h} \cdot \mathbf{r}_j)] \tag{6}$$

The crystal is represented by an approximate point-atom crystal.

For crystal with only one atomic species with atomic scattering factor $f(\mathbf{h})$, (6) becomes

$$F(\mathbf{h}) / f(\mathbf{h}) = \sum \delta(\mathbf{r}' - \mathbf{r}_j) \exp[2\pi i(\mathbf{h} \cdot \mathbf{r}_j)] \tag{7}$$

The right-hand side of (7) is an equal point-atom crystal.

Hence to study the application of iterative algorithms with oversampling to the many crystal structures solved by conventional methods, the first step is to study the equal point-atom crystals containing only one atomic species. This article presents a systematic study.

The point-atom constraint is extremely strong and powerful because besides the N points for the point atoms, in the whole region of a unit cell, the electron density is zero. This is much stronger than the non-negativity constraint. It is reasonable to expect that with the point-atom constraint and other constraints, the phases of the structure factors can be determined. The number of structure factors is finite, according to the resolution chosen.

The constraints of equal point-atom model of crystals are:

number constraint

equal-height constraint
atomic radius constraint
point-atom constraint.
interatomic distance constraint
structure constraint.

This article develops the technology of iterative algorithms with oversampling that will lead to the unique electron density. It is achieved by means of a systematic study of the equal point-atom crystals containing only one atomic species. A powerful tool of determining crystal electron density and atomic coordinates via equal point-atom model in combination with iterative algorithms and oversampling is developed. The point-atom coordinates of known equal point-atom crystals are first determined by large number of ∣ Fourier coefficients∣ and initial in-between ∣ Fourier coefficients∣ with eleven to sixteen strong constraints. For many crystal structures, centrosymmetric or non-centrosymmetric, and various symmetries and unit-cell shapes and sizes, we find the largest number of point-atoms that can be determined by this method (Sect. 2.2 and 2.3). *Ab initio* electron density with R -index = 0 is then obtained. The atomic coordinates obtained from this refined electron density is an improvement of the atomic coordinates reported in the literature. The in-between ∣ Fourier coefficients∣ are refined because of the consistent condition (Sect. 2.4). For an unknown structure of a crystal containing one atom species, the atomic coordinates of the point atoms can be determined (Sect. 2.5). *Ab initio* electron density with R -index = 0 is then obtained. The atomic coordinates obtained from this *ab initio* electron density will be close to, and are more accurate than the point-atom coordinates. The in-between ∣ Fourier coefficients∣ are refined because of the consistent condition (Sect. 2.6). The in-between ∣ Fourier coefficients∣ and ∣ Fourier coefficients∣ are adjusted within their uncertainties to obtain all the missed atoms and reasonable bond lengths and bond angles (Sect. 2.7). Molecule and solvent boundary can be obtained in Sect. 2.8. This tool can be applied to macromolecular crystals.

Sayre (1952) presents a squaring method for phase determination, which employs like atoms.

Avrami (1938), Buerger (1960), p. 553, Cervellino, A. & Ciccariello, S. (1996, 1999, 2001, 2005) present an algebraic approach to the phase problem. Zimmermann & Fischer (2009) employs the parameter space.

Direct methods use point atoms (Langs (1993)).

Low-density elimination is presented in Barrett & Zwick (1971), Shiono & Woolfson (1992).

Charge-flipping algorithms are presented in Oszlányi & Sütő (2004), Oszlányi & Sütő (2005), Milanesio & Viterbo (2011), Sect. 6.3.2.2, Appendix 6D, and Zanotti (2011), p. 732.

2.1. The many crystal structures solved by conventional methods are good candidates for developing the technology of iterative algorithms with oversampling that will lead to the unique electron density

The many crystal structures solved by conventional methods are good candidates for developing the technology of iterative algorithms with oversampling that will lead to the unique electron density. The advantages are:

(i) There are many solved crystal structures, small, moderate or large unit-cell sizes, different types of atoms, molecules, symmetries, with or without heavy atoms. The magnitudes of the structure factors have been measured.

(ii) The very regular shape of the unit cell with sharp corners and edges, and known unit cell constant. This is a very strong tight support.

(ii) Centrosymmetric or non-centrosymmetric

(iv) The in-between Fourier coefficients can be calculated from the structure solved. They are reasonably good (Yuen (2011a), Sect. 2.2).

(v) Non-negativity

(vi) Atomicity.

(vii) If needed, values of atomic radii can be included explicitly in the iterations.

(viii) Knowledge of values of interatomic distances. The distance between two neighboring atoms $\geq 2R$. The shortest interatomic distance d between an atom and its nearest neighbor is also known.

(ix) Knowledge of the environment of an atom is known, for examples, number of nearest neighbors, the interatomic distances between an atom and its nearest neighbors, the shortest nearest neighbor distance d , etc.

(x) Other symmetry constraints

(xi) Continuity constraint.

The electron density in a unit cell $f_0(x_1, x_2, x_3)$ is continuous with

$$f_0(0, x_2, x_3) = f_0(a, x_2, x_3) \quad \text{for all } x_2,\ x_3$$

$$f_0(x_1, 0, x_3) = f_0(x_1, b, x_3) \quad \text{for all } x_1,\ x_3$$

$$f_0(x_1, x_2, 0) = f_0(x_1, x_2, c) \quad \text{for all } x_1,\ x_2$$

(xii) Sharp-boundary constraint. The boundary values at the six boundary surfaces have the same order of magnitude as the value $f_0(x_1, x_2, x_3)$ inside a unit cell.

(xiii) By employing all the chemical and physical informations of a crystal (the advantages listed), more realistic or consistent sets of initial phases may be developed (Yuen (2011g), Sect. 3).

(xiv) The iterations (Yuen (2011h), Sect. 2.1, 2.2) can be compared with the process in determination of electron density of crystal structure which has been solved by conventional method. We can then seek for improvements.

(xv) Many methods have be developed for solving crystal structures, for example, the physical and non-physical methods of solving crystal structure in Woolfson & Fan (1995), the multiple-beam scattering method (Chang (2004)) and the direct methods in Giacovazzo (1998). To find a routine and efficient method of solving the structures of macromolecular crystals, may be we can incorporate these methods with iterative algorithms and oversampling. At least, we can get some realistic initial phases from these methods. To have a routine and efficient method of solving the structures of macromolecular crystals is very important. All methods and all chemical and physical informations may be employed. An image reconstruction package for coherent X-ray diffractive imaging, *Hawk* is presented in Maia et al. (2010).

(xvi) Atomicity (vi) can be employed in depth. An approximate point-atom crystal is given by (6). Hence the very powerful point-atom constraint can be applied (Sect. 2.2). We may be able to incorporate the direct method developed by Hauptman & Karle (Thibault & Elser (2010), pp. 241-242) with iterative algorithms and oversampling.

2.2. Determination of the atomic coordinate of an artificial equal point-atom crystal

The constraints of equal point-atom model of crystals are:

(i) number constraint. The number of atoms N in a unit cell is known.

(ii) equal-height constraint. The atoms have equal height H. H is bounded.

(iii) atomic radius constraint and point-atom constraint. All atoms have same radius R. A point atom is represented by a point with height H centered at a circle with radius R. The value of the atom in this circle, except the origin, is equal to zero (point-atom constraint). To help the visualization, we use two- dimensional description and the word 'circle'. In fact, it is three-dimensional, and should be 'sphere'.

(iiia) The value of R is not specified.

(iiib) The value of R is given.

(iv) interatomic distance constraint. The distance between two neighboring atoms $\geq 2R$. The shortest interatomic distance d between an atom and its nearest neighbor is also known.

(v) structure constraint. The environment of an atom is known, for examples, number of nearest neighbors, the interatomic distances between an atom and its nearest neighbors, the shortest nearest neighbor distance d, etc.

We distribute N equal point-atoms in a unit cell according to the structure constraint (v) to construct an artificial centrosymmetric equal point-atom crystal. Calculate the Fourier coefficients and in-between Fourier coefficients for this crystal.

Use large oversampling ratio $2^3 = 8$. Then we use these | Fourier coefficients | and in-between | Fourier coefficients | . These in-between | Fourier coefficients | will be fixed in the iterations. Our purpose is to see whether we can recover the atomic coordinates of this artificial centrosymmetric equal point-atom crystal from these | Fourier coefficients | and in-between | Fourier coefficients | .

Use more realistic initial phases of these | Fourier coefficients | and in-between | Fourier coefficients | obtained in Yuen (2011g), Sect. 2.1, (A) or Sect. 2.3 for equal point atoms. Apply iterative algorithms with oversampling in Yuen (2011a), Sect. 2.4 to calculate the electron density of this crystal. Eleven constraints are used (the five constraints for equal point-atom model in above, and six constraints in Yuen (2011a), Sect. 2.1). In the electron density obtained after the first iteration, use a small value of atomic radius r. Start from the highest atom, set the electron density in the circle centered at this atom and radius r, to zero. That is, we have a point atom. A point atom has a sharp boundary at its circumference of radius r. Apply the same process to the next highest atom, and so on. Probably, the resultant electron density has more than N atoms. Then increase the value r so that the resultant electron density has N atoms. The number constraint (i) and the atomic radius constraint (iiia) are satisfied. The negative electron density outside the circles of the point atoms is set equal to zero. This is low-density elimination (Barrett & Zwick (1971), p. 8, Shiono & Woolfson (1992)). The positive electron density is then flipped. This is charge flipping in Oszlányi & Sütő (2004). The boundary of a point atom becomes sharper and has its height above its surrounding increased by the density flipped. 'Sharper' and 'higher' accelerate the convergence of the iterations. After a number of iterations, the electron density between the circles should be iteratively forced to negligible value. An alternative is just set the electron density of the whole crystal except the center peaks of the point atoms to zero. Experience will show which is more effective.

Use

(A) Set the height of each point atom to H ; the equal-height constraint (ii) is satisfied. Proceed to the second iteration, and follow the same procedure. So on. If the iterations converge, see whether the result is the correct centrosymmetric equal point-atom crystal.

or

(B) Set the height of the highest point atom to H. Adjust the heights of the remaining $N-1$ atoms accordingly. Set the height of the 1% N highest atoms to H. It is reasonable to believe that these atoms are correct. Hence we have a list of 1% N atoms which are reasonably correct. Proceed to the second iteration. Follow the same procedure and set the height of the 2% N highest atoms to H. So on. Here we use a 'slow but sure' process. After each iteration, we increase 1% N of the list of atoms which are reasonably correct. After 100 iterations, all the atoms have height H; the equal-height constraint (ii) is satisfied. Then proceed as in (A). If the iterations converge, see whether the atomic coordinates of the centrosymmetric equal point-atom crystal are recovered.

As experience grows, we will learn whether we use steps smaller or larger than 1%. In fact, we may choose different size of these steps as the iterations progress.

We expect that this method will provide a unique result for unit cell with small number of atoms because the point-atom constraint is very powerful. This has been shown in the success of the charge-flipping algorithms. We then increase the value of N. As the number of atoms N is increased, the unit cell will become larger. We will keep the same shape. For example, for a triclinic unit cell, the unit cell angles (α,β,γ), and the ratios of the unit cell edge-lengths (a,b,c) remain the same. Find the maximum value of N, say $N_{\max 1}$ for this shape of the unit cell, so that we can recover the atomic coordinates of the artificial centrosymmetric equal point-atom crystal.

After we find the maximum value of $N_{\max 1}$, we increase the number of atoms to $N_{\max 1}+\delta N$, where δN is a small integer. Apply iterative algorithms with oversampling in Yuen (2011a), Sect. 2.4. The iterations converge to atomic coordinates which are different from the artificial centrosymmetric equal point-atom crystal. This is because, during the iterations, the convergence is shifted to a local minimum. In general, we expect that the difference comes from:

> There are two atoms whose interatomic distance is too small, giving too small atomic radius.
>
> The shortest nearest neighbor distance is very much different from the value d determined from chemical knowledge.
>
> The atomic coordinates obtained violates chemical knowledge.

(If these three conditions are not violated, may be crystal with these atomic coordinates also exists).

Therefore, start from the electron density obtained from the first iteration, we add constraint (iiib) value of R from chemical knowledge, is given. The atoms are adjusted so that all interatomic distances in the crystal $\geq 2R$. If now the iterations converge to the correct atomic coordinates, we will increase the value of δN, until we find the maximum value of N, say $N_{\max 2}$ for which we can recover the correct atomic coordinates.

We further increase the value of δN. Now we add constraint (iv) the value of the shortest interatomic distance d from chemical knowledge is given. Repeat the process. Find the maximum value of N, say $N_{\max 3}$ for which we can recover the correct atomic coordinates.

We further increase the value of δN. Now we add constraint (v) the structure constraint from chemical knowledge is given. In the iterations, when some **partial structures** are identified, we can increase the heights of these point atoms to, say, $1.2H$. The fine structure and the locations of these partial structures may be refined in the iterations that follow. The heights of the point atoms of the partial structures are always adjusted to $1.2H$. In this way, we hope to force the convergence of the remaining point atoms to the correct positions. Find the maximum value of N, say $N_{\max 4}$ for which we can recover the correct atomic coordinates. When the iterations converge, all point atoms have height $1.2H$. As experience grows, depending on the reliability of the partial structure, we will learn whether we use height smaller or larger than $1.2H$. In fact, we may choose different heights for different partial structures as the iterations progress.

Then for this unit-cell shape, we obtain a set of $\{N_{\max 1}, N_{\max 2}, N_{\max 3}, N_{\max 4}\}$.

Repeat this for all shapes of a triclinic unit cell:

Use the reasonably smallest α, several intermediate values of α, and reasonably largest α,

the reasonably smallest β, several intermediate values of β, and reasonably largest β,

the reasonably smallest γ, several intermediate values of γ, and reasonably largest γ,

the reasonably smallest a, several intermediate values of a, and reasonably largest a,

the reasonably smallest b, several intermediate values of b, and reasonably largest b,

the reasonably smallest c, several intermediate values of c, and reasonably largest c.

Hence, for triclinic unit cells, we have a finite set of $\{N_{\max 1}, N_{\max 2}, N_{\max 3}, N_{\max 4}\}$.

Then we repeat this for all centrosymmetric space groups. For all centrosymmetric space groups, we have a finite set of $\{N_{\max 1}, N_{\max 2}, N_{\max 3}, N_{\max 4}\}$.

Repeat this for all non-centrosymmetric space groups. Apply iterative algorithms with oversampling in Yuen (2011d), Sect. 2.3. Sixteen constraints are used (the five constraints for equal point-atom model in above, and eleven constraints in Yuen (2011d), Sect. 2.1, except (iv)). We have a finite set of $\{N_{\max 1}, N_{\max 2}, N_{\max 3}, N_{\max 4}\}$.

Repeat the calculations with sets of random initial phases.

By means of this systematic study, we have obtained a finite set of $\{N_{\max 1}, N_{\max 2}, N_{\max 3}, N_{\max 4}\}$ for all space groups and unit-cell shapes and sizes, for which iterative algorithms with oversampling will lead to the unique and correct atomic coordinates.

In the iterative algorithms with oversampling, large oversampling ratio $2^3 = 8$ is used. The total number of | Fourier coefficients| and initial in-between | Fourier coefficients｜ is eight times that of the｜Fourier coefficients｜ (observed structure factors). This total number is much larger than the number of atomic coordinates of the point atoms. The system is excessively over-determined. Seven constraints are used in Yuen (2011a), Sect. 2.1, 2.3, or six or thirteen constraints in Yuen (2011d), Sect. 2.1, plus the five constraints for equal point-atom crystal in above. The constraints are strong. We expect that $N_{\max 4}$ will be very large, that is, the method applies to macromolecular crystals.

2.3. Determination of the atomic coordinates of the equal point-atom crystal from crystal structure reported in the literature

Choose a crystal structure containing only one atomic species. Divide the magnitudes of the structure factors by the atomic scattering factor. The result represents an equal point-atom crystal (see (7)). Replace the N atoms per unit cell by point-atoms. This equal point-atom crystal is more realistic than the artificial equal point-atom crystal in Sect. 2.2. Calculate the Fourier coefficients and in-between Fourier coefficients for this crystal. Use large oversampling ratio $2^3 = 8$. Then we use these｜Fourier coefficients｜ and in-between｜Fourier coefficients｜. Our purpose is to see whether we can recover the unique atomic coordinates of this equal point-atom crystal from these｜Fourier coefficients｜ and in-between｜Fourier coefficients｜.

Apply Sect. 2.2 with constraints (i), (ii) and (iiia). Use more realistic or consistent set of initial phases in Yuen (2011g), Sect. 2.1, (A), Sect. 2.2, or Sect. 2.3, for point atoms. If the correct atomic coordinates are recovered, then we know that iterative algorithms with oversampling work for this crystal. If not, apply constraints (iiib), (iv) and (v) successfully, until the correct atomic coordinates are recovered, then we know that for this crystal, iterative algorithms with oversampling and these five constraints work. If even though we use all five constraints, iterative algorithms with oversampling fail to recover the correct atomic coordinates, we may seek for help from other methods in crystallography.

Repeat this for other crystal structures containing only one atomic species. For each crystal, we find the number of constraints which can or cannot recover the atomic coordinates.

2.4. Refinement of the electron density and atomic coordinates

After the atomic coordinates of the point atoms are determined in Sect. 2.3, use (1) to calculate the structure factors $F(\mathbf{h})$, from which the approximate electron density of the crystal is determined. Even though the crystal contains only one atom species, the electron density may be different for different atoms. Eq. (1) is always an approximation. Calculate the in-between Fourier coefficients for this approximate electron density. The calculated in-between Fourier coefficients are reasonably good (Yuen (2011a), Sect. 2.2). Use their magnitudes as the initial in-between | Fourier coefficients | . Use the experimental | Fourier coefficients | . Use the phases of the calculated Fourier coefficients and in-between Fourier coefficients as initial phases. Using Yuen (2011a), Sect. 2.3 or Yuen (2011d), Sect. 2.2 (Sect. 2.1, constraint (iv) is now used), iterative algorithms with oversampling will converge to a unique and refined electron density. This refined *ab initio* electron density has R -index = 0. The atomic coordinates obtained from this refined electron density is an improvement of the atomic coordinates reported in the literature. The in-between | Fourier coefficients | are refined because of the consistent condition (Yuen (2011a), Sect. 2.1).

2.5. Determination of the atomic coordinates of the equal point-atom crystal for an unknown structure of a crystal containing one atom species.

The procedures presented in Sect. 2.2 and 2.3 can be applied for determining the atomic coordinates of the point atoms of an unknown structure of a crystal containing one atom species. Obtain the initial in-between | Fourier coefficients | (or mid-Bragg intensity) by interpolation in multidimensions or by experimental measurement (See Yuen (2011a), Sect. 2.1). Use large oversampling ratio $2^3 = 8$. Divide the | Fourier coefficients | and in-between | Fourier coefficients | by the atomic scattering factor of the atoms. From (7), the crystal is an equal point-atom crystal. Use more realistic or consistent set of initial phases in Yuen (2011g), Sect. 2.1, (A), Sect. 2.2, or Sect. 2.3, for point atoms. Apply iterative algorithms with oversampling in Sect. 2.2 and 2.3. The coordinates of the point atoms are determined. From the many results in Sect. 2.2 and 2.3, we have more confidence whether the procedure can or cannot recover the atomic coordinates of the point-atom crystal. As mentioned in Sect. 2.2, with the large number of ｜Fourier coefficients｜ and initial in-between ｜Fourier coefficients｜, and eleven to sixteen strong constraints, it is reasonable to expect that the method applies to macromolecular crystals.

2.6. Determination of the *ab initio* electron density and atomic coordinates

After the atomic coordinates of the point atoms are determined in Sect. 2.5, follow Sect. 2.4 to obtain the crystal electron density with R-index = 0. The atomic coordinates obtained from this *ab initio* electron density will be close to, and are more accurate than the point-atom coordinates. The in-between ｜Fourier coefficients｜ are refined because of the consistent condition.

2.7. Recovery of missing atoms and improvement of bond lengths and bond angles

Following Yuen (2011a), Sect. 2.5 and Yuen (2011d), Sect. 2.4, the in-between ｜Fourier coefficients｜ and ｜Fourier coefficients｜ are adjusted within their uncertainties to obtain all the missed atoms, and reasonable bond lengths and bond angles.

2.8. Molecular and solvent boundary

Consider a crystal of macromolecules in a solvent, all of the same species of atoms. The volume contents of the macromolecule and the solvent are known. The macromolecules have structure features, while the solvent is more homogeneous. During the iterations in Sect. 2.3 and 2.5, a gross molecular structure may be recognized. From the volume contents of the macromolecules and the solvent, a gross molecule and solvent boundary can be obtained. This can be used as a loose support for further iterations.

3. Discussions and conclusion

This article develops the technology of iterative algorithms with oversampling that will lead to the unique crystal electron density. It is achieved by means of a natural and systematic study of the equal point-atom crystals containing only one atomic species. Sect. 2.2 and 2.3 apply to ideal conditions (Cervellino & Ciccariello (1996), p. 8).

If we find the answer to the question in Sect. 2.2, that is, we find the maximum value of N , $N_{\max 1}$, so that we can recover the electron density of the artificial centrosymmetric equal point-atom crystal, this is a very important step in the determination of structures of macromolecular crystals.

In Sect 2.2, for crystals of all centrosymmetric and non-centrosymmetric space groups, all shapes and dimensions of the unit cells, we can determine the set of $\{N_{\max 1}, N_{\max 2}, N_{\max 3}, N_{\max 4}\}$. This is a systemic investigation of solving the phase problem for equal point-atom crystals. The purposes of this systemic investigation are:

(a) To verify that there exists solution to the phase problem in X-ray crystallography; the phases can be determined uniquely from the | Fourier coefficients| and in-between | Fourier coefficients| .

(b) To show that iterative algorithms with oversampling can be a routine and efficient tool to obtain the unique solution.

(c) To determine the shapes and size of the unit cells, and the number of atoms per unit cell, such that iterative algorithms with oversampling is a routine and efficient tool to obtain the unique solution, that is to determine the set of $\{N_{\max 1}, N_{\max 2}, N_{\max 3}, N_{\max 4}\}$. For an unknown crystal structure containing only one atomic species, we know immediately whether it can be solved by applying iterative algorithms with oversampling applied to an equal point-atom crystal (Oszlányi & Sütő (2005), p. 148).

(d) To identify the important steps in the algorithms, so that we can do more researches about these steps:

  (d1) Initial phases. The starting point of the iterations, the initial phases, is very important. It may decide whether or not the iterations will converge to the unique solution. The commonly employed random phases are not very ideal. It would be better if we can find some more realistic initial phases so that we can have more confidence that the iterations will converge to the solution. Yuen (2011g) presents an attempt to find more realistic or consistent set of initial phases. Hope that other researchers will offer more and better methods. May be a probabilistic approach like the direct method can provide a consistent set of initial phases.

(d2) In-between | Fourier coefficients | . These are very important quantities. It would be better it they can be measured experimentally. Yuen (2011f) presents an attempt. Hope that other researchers will develop the technology of this method, or offer more and better methods.

(d3) The iterative algorithms can be improved and specialized to include all chemical and physical knowledge of crystals, for examples, the sixteen advantages in Sect. 2.1 and the five constraints in Sect. 2.2 for equal point-atom model.

(d4) Other important steps to be discovered.

(e) For a crystal which is out of the range $\{N_{\max 1}, N_{\max 2}, N_{\max 3}, N_{\max 4}\}$, and cannot be solved by means of iterative algorithms with oversampling, we can then seek for co-operations with other conventional methods in solving crystal structures.

Note that we have divided $F(\mathbf{h})$ by $f(\mathbf{h})$ to obtain the equal point-atom crystal in (7). We can then apply the powerful point-atom constraint: A point atom is represented by a point with height H centered at a circle with radius R. The value of the atom in this circle, except the origin, is equal to zero. Iterations of low-density elimination and charge flipping can then be applied to force the electron density between the atoms to zero. This helps to determine the crystal structure in terms of point atoms. After the atomic coordinates of the point atoms are determined, the electron density of the crystal in reality can be obtained. The crystal electron density is determined with the | Fourier coefficients | and initial in-between | Fourier coefficients | (Sect. 2.4, 2.6). The electron density obtained has R-index = 0. Note that we have employed chemical and physical informations in determination of the point-atom coordinates (Sect 2.3, 2.5). In fact, the employment of point atoms is itself a chemical information. However the electron density obtained is *ab initio*. It is determined by the experimental lattice constants, diffraction intensities and initial in-between | Fourier coefficients | ; no other chemical or physical information (for example, atomicity, chemical formula, bond length, bond angle, etc) is used. Moreover, the in-between | Fourier coefficients | are also refined because of the consistent condition.

From the electron density obtained, atomic coordinates are obtained. For a crystal structure reported in the literature, these atomic coordinates is an improvement of the atomic coordinates reported in the literature. For an unknown crystal structure, the atomic coordinates obtained from this refined electron density will be close to, and is more accurate than the point-atom coordinates.

That is, crystal electron density and atomic coordinates are determined via the point-atom model (Eq. (7)). This is because of atomicity; the atomic scattering factors have approximately same shapes. This powerful tool cannot be applied directly to $F(\mathbf{h})$ in (1).

Intuitively, we ignore the bulk electron distribution of an atom in reality, and replace it by a point atom. All the | Fourier coefficients | and initial in-between | Fourier coefficients | are used to determine the atomic coordinates of the point atoms. This number is much larger than the number of atomic coordinates of the point atoms. The system is excessively over-determined. With eleven to sixteen strong constraints employed (see Sect. 2.2), it is reasonable to expect that atomic coordinates for macromolecules can be determined by means of iterative algorithms and oversampling.

As we have obtained an electron density with R -index = 0, we have made full use of the | Fourier coefficients | . Hence it is meaningful to have Sect. 2.7: Recovery of missing atoms and improvement of bond lengths and bond angles.

To solve the phase problem in X-ray crystallography, a systematic study of the simplest model is necessary. The equal point-atom model is the simplest model. All the | Fourier coefficients | and in-between | Fourier coefficients | are assumed to be known, and there are no experimental uncertainties. Studies of such ideal situations are important and necessary because the result will not be masked by the bulk electron densities of the atoms, or atomic scattering factors, the experimental details and experimental uncertainties. It shows the true power of the method studied.

As in Sect. 2.2, we may also perform a systematic study of direct methods applied to the equal point-atom model with known | Fourier coefficients | and in-between | Fourier coefficients | .

For charge-flipping algorithms, Oszlányi & Sütő (2005), p. 148 states that 'All-light-atom structures are more difficult to solve, especially with more than 100 atoms in the unit cell'. We expect that iterative algorithms with oversampling applied to equal point-atom crystal may be an improvement.

With large oversampling ratio $2^3 = 8$, the number of | Fourier coefficients | and in-between | Fourier coefficients | employed is eight times the number of | Fourier coefficients | employed by Avrami (1938), Buerger (1960), p. 553, Cervellino, A. & Ciccariello, S. (1996, 2001, 2005) and Zimmermann & Fischer (2009). It will be very interesting if algebraic or parameter space approach can be extended to include all | Fourier coefficients | and in-between | Fourier coefficients | . Moreover, it will be useful if this can be combined with iterative algorithms and oversampling so that the amount of computation can be reduced.

As a conclusion, a powerful tool of determining crystal electron density and atomic coordinates via equal point-atom model in combination with iterative algorithms and oversampling is developed. It can be applied to macromolecular crystals. The next step is to extend this article to crystals containing atoms of different species, in particular, heavy atoms (see Yuen (2011h)).

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22

Ab initio determination of crystal electron density and atomic coordinates via point-atom model. Iterative algorithms with oversampling

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Abstract

Yuen (2011f) and this article develop the technology of iterative algorithms with oversampling that will lead to the unique electron density. It is achieved by means of a systematic study of point atoms of crystals. Crystal electron density and atomic coordinates are determined via a point-atom model in combination with iterative algorithms and oversampling. The point-atom coordinates are first determined by large number of｜Fourier coefficients｜ and initial in-between｜Fourier coefficients｜ with eleven to sixteen strong constraints. An iterative scheme is proposed. It concentrates on changing the **heaviest atoms** to point atoms, and have their coordinates determined, so on. *Ab initio* electron density with R -index = 0 is then obtained. The in-between ｜Fourier coefficients｜ are refined because of the consistent condition. The in-between｜Fourier coefficients｜ and｜Fourier coefficients｜ are adjusted within their uncertainties to obtain all the missed atoms and reasonable bond lengths and bond angles. This may be a powerful tool, and is applicable to macromolecular crystals.

Keywords: Crystallography; point atom; iterative algorithms; oversampling; excessively over-determined system; powerful point-atom constraint; low-density elimination; charge-flipping algorithms; iterative scheme; heaviest atoms; electron density; atomic coordinates; consistent condition; partial structures; macromolecular crystals; missed atoms.

1. Introduction

This is a sequel to Yuen (2011f). These two articles develop the technology of iterative algorithms with oversampling that will lead to the unique electron density. Crystal electron density and atomic coordinates are determined via a point-atom model in combination with iterative algorithms and oversampling.

The non-hydrogen point-atom coordinates of crystal structures reported in the literature are first determined by large number of | Fourier coefficients | and initial in-between | Fourier coefficients | with eleven to sixteen strong constraints. An iterative scheme concentrates on changing the **heaviest atoms** to point atoms, the locations of which are then determined, so on. This may be a very effective method. This is done for many crystal structures, centrosymmetric or non-centrosymmetric, and various symmetries and unit-cell shapes and sizes (Sect. 2.1 and 2.2). *Ab initio* electron density with R -index = 0 is then obtained. The hydrogen and non-hydrogen atomic coordinates obtained from this refined electron density is an improvement of the atomic coordinates reported in the literature. The in-between | Fourier coefficients | are refined because of the consistent condition (Sect. 2.3). For the unknown structure of a crystal, the non-hydrogen atomic coordinates of the point atoms can be determined by the method presented (Sect. 2.4). *Ab initio* electron density with R -index = 0 is then obtained. Hydrogen and non-hydrogen atomic coordinates can be obtained from this *ab initio* electron density. The non-hydrogen atomic coordinates will be close to, and are more accurate than the point-atom coordinates. The in-between | Fourier coefficients | are refined because of the consistent condition (Sect. 2.5). The in-between | Fourier coefficients | and | Fourier coefficients | are adjusted within their uncertainties to obtain all the missed atoms and reasonable bond lengths and bond angles (Sect. 2.6). Molecule and solvent boundary can be obtained in Sect. 2.7. This tool can be applied to macromolecular crystals.

Low-density elimination is presented in Barrett & Zwick (1971), Shiono & Woolfson (1992), Refaat & Woolfson (1993), Matsugaki & Shiono (2001) and Takakura et al. (2006). It is used for powder diffraction in Altomare et al. (2002).

Charge-flipping algorithms are presented in Oszlányi & Sütő (2004), Palatinus (2004), Wu et al. (2004), Oszlányi & Sütő (2005, 2006, 2007), Zhou & Harris (2008), Eggeman et al. (2009), Fleischer et al. (2010) and Palatinus et al. (2011). Incorporation with the tangent formula is presented in Coelho (2007).

Application of charge-flipping algorithms to X-ray powder diffraction is presented in

Wu et al. (2006), McCusker & Baerlocher (2008) and Robertson & Bish (2010). A combination of maximum entropy method and charge flipping is presented in Samy et al. (2010).

Dumas & van der Lee (2008) presents macromolecular structure solution by charge flipping.

Oszlányi & Sütő (2008) presents a very useful review.

Solvent flattening is presented in Wang (1981, 1985), Woolfson & Fan (1995), Sect. 5.2, 7.6.5, Gu et al. (1997), Terwilliger et al. (1999), Yonekura & Toyoshima (2000), and Roversi et al. (2010).

Density modification is presented in Schevitz et al. (1981), Bhat & Blow (1982), Cannillo et al. (1983), Navaza et al. (1983), Roberts & Brünger (1995), Cowtan & Main (1996, 1998), Cowtan & Zhang (1999), Read & Kleywegt (2001), Takakura et al. (2002), Terwilliger (2003), Turk (2007), Cowtan (2010), Milanesio & Viterbo (2011), Sect. 6.3.2, and Zanotti (2011), Sect. 9.4.13.

Iterative methods in protein crystallography is presented in Thumiger & Zanotti (2009).

Direct-space methods in phase extension and phase refinement are presented in Refaat et al. (1996).

Direct methods for macromolecules are presented in Sheldrick & Schneider (2001) and Chowdhury et al. (2005).

2.1. Determination of the non-hydrogen atomic coordinate of the point-atom crystal from crystal structure reported in the literature.

Consider a crystal structure reported in the literature. The structure factor

$$F(\mathbf{h}) = \sum f_j(\mathbf{h}) \exp[2\pi i(\mathbf{h}\cdot\mathbf{r}_j)] \qquad (1)$$

Neglect the hydrogen atoms.

$$F(\mathbf{h}) = \sum f_j(\mathbf{h})\delta(\mathbf{r}'-\mathbf{r}_j) \exp[2\pi i(\mathbf{h}\cdot\mathbf{r}_j)] \qquad (2)$$

where $\delta(\mathbf{r}')$ represents a point atom. It is bounded, and

$$\int \delta(\mathbf{r}')d\mathbf{r}' = 1 \tag{3}$$

The atomic scattering factors have approximately same shapes.

Let $\hat{f}(\mathbf{h}) = (\sum f_j(\mathbf{h}))/(\sum Z_j)$ (4)

where Z_j is the atomic number of the j -th atom.

$\hat{f}(\mathbf{h})$ is the atomic scattering factor of a hypothetical atom with $Z = 1$.

$$f_j(\mathbf{h}) \approx Z_j \hat{f}(\mathbf{h}) \tag{5}$$

Divide the structure factors (2) by $\hat{f}(\mathbf{h})$.

$$F(\mathbf{h})/\hat{f}(\mathbf{h}) \approx \sum Z_j \delta(\mathbf{r}'-\mathbf{r}_j)\exp[2\pi i(\mathbf{h}\cdot\mathbf{r}_j)] \tag{6}$$

The right-hand side of (6) represents a point-atom crystal.

For this point-atom crystal, use the atomic coordinates reported in the literature. Calculate the Fourier coefficients and in-between Fourier coefficients. Then we use these | Fourier coefficients | and in-between | Fourier coefficients | . Use large oversampling ratio $2^3 = 8$. Our purpose is to see whether we can recover the atomic coordinates of the point-atom crystal from these | Fourier coefficients | and in-between | Fourier coefficients | .

Follow Yuen (2011f), Sect. 2.2. Use constraints:

(i) number constraint. The number of non-hydrogen atoms N in a unit cell is known.

(ii) height constraint.

The height of a point atom = (atomic number of this atom) × H = ZH

H is the height of a point atom with $Z = 1$.

(iii) atomic radius constraint and point-atom constraint.

(iiia) The atomic radii are not specified.

(iiib) The values of atomic radii from chemical knowledge, are given. The atomic radius depends on the atomic number.

(vi) interatomic distance constraint. The distance between two neighboring atoms ≥ sum of the atomic radii of the two atoms. The shortest interatomic distance d between an atom and its nearest neighbor is also known.

(v) structure constraint. The environment of an atom is known, for examples, number of nearest neighbors, the interatomic distances between an atom and its nearest neighbors, the shortest nearest neighbor distance d, etc.

Use more realistic or consistent set of initial phases in Yuen (2011e), Sect. 2.1, (A), Sect. 2.2, or Sect. 2.3, of point atoms for the $|$ Fourier coefficients $|$ and in-between $|$ Fourier coefficients $|$. Apply iterative algorithms with oversampling in Yuen (2011a), Sect. 2.4 or Yuen (2011b), Sect. 2.3 to calculate the electron density of this crystal. In the electron density obtained after the first iteration, use a small value of atomic radius r. Start from the highest atom, set the electron density in the circle centered at this atom and radius r, to zero. We have a point atom. A point atom has a sharp boundary at its circumference of radius r. Apply the same process to the next highest atom, and so on. Probably, the resultant electron density has more than N atoms, the number of atoms in a unit cell. Then increase the value r so that the resultant electron density has N atoms. The number constraint (i) and the atomic radius constraint (iiia) are satisfied. The negative electron density outside the circles of the point atoms is set equal to zero. This is low-density elimination (Barrett & Zwick (1971), p. 8, Shiono & Woolfson (1992)). The positive electron density is then flipped. This is charge flipping in Oszlányi & Sütő (2004). The boundary of a point atom becomes sharper and has its height above its surrounding, increased by the density flipped. 'Sharper' and 'higher' accelerate the convergence of the iterations. After a number of iterations, the electron density between the circles should be iteratively forced to negligible value. An alternative is just set the electron density outside the circles of the point atoms to zero. Experience will show which is more effective.

Use the 'slow but sure' process in Yuen (2011f), Sect. 2.2, (B). Set the height of the highest point atom to (atomic number of the heaviest atom) × H. Adjust the heights of the remaining $N-1$ atoms accordingly. Set the height of the 1% N highest atoms to (atomic number of the heaviest atom) × H. If the number of heaviest atoms is less than 1% N, then set the heights of the remaining of the 1% N atoms to (atomic number of the next heaviest atom) × H. It is reasonable to believe that these 1% N atoms are correct. Hence we have a list of 1% atoms which are reasonably correct. Proceed to the second iteration, so on. After each iteration, we increase 1% N of the list of atoms which are reasonably correct. The heights of all the reasonably correct atoms are set equal to (atomic number of the atom) × H. After 100 iterations, all the atoms have height = (atomic number of the atom) × H; the height constraint (ii) is satisfied. Then proceed with more iterations. Heights of some atoms may be changed. Adjust the heights so that the height constraint (ii) is always satisfied. If the iterations converge, see whether the atomic coordinates of the point-atom crystal are recovered.

As experience grows, we will learn whether we use steps smaller or larger than 1%. In fact, we may choose different size of these steps as the iterations progress.

If the correct atomic coordinates are recovered with constraints, (i), (ii) and (iiia), then we know that iterative algorithms with oversampling with these constraints work for this crystal. If not, we follow Yuen (2011f), Sect. 2.2, and apply constraints (iiib), (iv) and (v) successfully. In the iterations, when some **partial structures** are identified, we can increase the heights of these point atoms to, say, 1.2 × (atomic number of the atom) × H. The fine structure and the locations of these partial structures may be refined in the iterations that follow. The heights of the point atoms of the partial structures are always adjusted to 1.2 × (atomic number of the atom) × H. In this way, we hope to force the convergence of the remaining point atoms to the correct positions. In the final stage of the iterations, the heights of all the atoms are set equal to (atomic number of the atom) × H. The height constraint (ii) is satisfied. If the correct point-atom coordinates are recovered, we know that for this crystal, iterative algorithms with oversampling and these constraints work.

If even though we use all five constraints, iterative algorithms with oversampling fail to recover the correct point-atom coordinates, we may seek for help from other methods in crystallography.

Repeat this for many crystal structures reported in the literature, centrosymmetric or non-centrosymmetric, and various symmetries and unit-cell shapes and sizes.

Repeat the calculations with sets of random initial phases.

In the iterative algorithms with oversampling, large oversampling ratio $2^3 = 8$ is used. The total number of | Fourier coefficients | and initial in-between | Fourier coefficients | is eight times that of the | Fourier coefficients | (observed structure factors). This total number is much larger than the number of atomic coordinates of the non-hydrogen point atoms. The system is excessively over-determined. Seven constraints are used in Yuen (2011a), Sect. 2.1, 2.3, or six or thirteen constraints in Yuen (2011b), Sect. 2.1, plus the five constraints for point-atom crystal in above. The constraints are strong. We expect that the method is applicable to macromolecular crystals.

2.2. Determination of the coordinates of non-hydrogen point-atoms of crystal structure reported in the literature.

Consider a crystal structure reported in the literature. Use the experimental | Fourier coefficients | . Calculate the Fourier coefficients and in-between Fourier coefficients for this crystal structure. The calculated in-between Fourier coefficients are reasonably good (Yuen (2011a), Sect. 2.2). Use their magnitudes as the initial in-between | Fourier coefficients | . Use large oversampling ratio $2^3 = 8$. Divide the experimental | Fourier coefficients | and the calculated in-between | Fourier coefficients | by $\hat{f}(\mathbf{h})$. From (6), the crystal is an approximate point-atom crystal. Our purpose is to see whether we can recover the unique atomic coordinates of this point-atom crystal from these | Fourier coefficients | and in-between | Fourier coefficients | . Use more realistic or consistent set of initial phases in Yuen (2011g), Sect. 2.1, (A), Sect. 2.2, or Sect. 2.3, for point atoms. Apply iterative algorithms with oversampling in Sect. 2.1 with constraints (i), (ii) and (iiia).

Instead of dividing the | Fourier coefficients | and in-between | Fourier coefficients| by $\hat{f}(\mathbf{h})$, we may divide by $[\sum f_j^2(\mathbf{h})]^{1/2}$ (the normalized structure factors), or by $[\sum f_j^n(\mathbf{h})]^{1/n}$ as $n \to \infty$, $= f_H(\mathbf{h})$. $f_H(\mathbf{h})$ is the atomic scattering factor of the heaviest atom in the crystal (Oszlányi & Sütő (2008), p. 130)). Various iterative schemes can be tested. One scheme is to first divide by the atomic scattering factor of the heaviest atom. That is, we concentrate on changing the **heaviest atoms** to point atoms. Iterations are then used until all the heaviest atoms are located. As mentioned in Sect. 2.1, with the large number of |Fourier coefficients| and initial in-between |Fourier coefficients| , and eleven to sixteen strong constraints, it is reasonable to expect that this iterative scheme will be able to determine the locations of the heaviest atoms. Then divide the |Fourier coefficients| and in-between |Fourier coefficients| by the atomic scattering factor of the second heaviest atom. Then all the second heaviest atoms are found, so on. In this way, the coordinates of the all non-hydrogen point-atoms are determined. This may be a very effective method. Determination of the heavier atoms helps the determination of all other atoms. The usefulness of these variants or iterative schemes can be investigated by applying them to a number of crystals.

If the correct non-hydrogen atomic coordinates are recovered, then we know that iterative algorithms with oversampling work for this crystal. If not, apply constraints (iiib), (iv) and (v) successfully, until the correct atomic coordinates are recovered, then we know that for this crystal, iterative algorithms with oversampling and these five constraints work. If even though we use all five constraints, iterative algorithms with oversampling fail to recover the correct non-hydrogen atomic coordinates, we may seek for help from other methods in crystallography.

Repeat this for many crystal structures reported in the literature, centrosymmetric or non-centrosymmetric, and various symmetries and unit-cell shapes and sizes.

2.3. Refinement of the electron density and the hydrogen and non-hydrogen atomic coordinates

After the coordinates of the non-hydrogen point-atoms are determined in Sect. 2.2, use (1) to calculate the structure factors $F(\mathbf{h})$, from which the approximate electron density of the crystal is determined. The electron density may be different for different atoms of the same species. Eq. (1) is always an approximation. Calculate the in-between Fourier coefficients for this approximate electron density. These calculated in-between Fourier coefficients are reasonably good (Yuen (2011a), Sect. 2.2). Use their magnitudes as the initial in-between | Fourier coefficients | . Use the experimental | Fourier coefficients | . Use the phases of the calculated Fourier coefficients and the calculated in-between Fourier coefficients as initial phases. Using Yuen (2011a), Sect. 2.3 or Yuen (2011b), Sect. 2.2, iterative algorithms with oversampling will converge to a unique and refined electron density. This refined *ab initio* electron density has R -index = 0. From this refined electron density, hydrogen and non-hydrogen atomic coordinates are obtained. These atomic coordinates is an improvement of the atomic coordinates reported in the literature. The in-between | Fourier coefficients | are refined because of the consistent condition (Yuen (2011a), Sect. 2.1).

2.4. Determination of the coordinates of non-hydrogen point-atoms of the unknown structure of a crystal

Consider an unknown crystal structure. Use the experimental | Fourier coefficients | . Obtain the initial in-between | Fourier coefficients | (or mid-Bragg intensity) by interpolation in multidimensions or by experimental measurement (See Yuen (2011a), Sect. 2.1). Use more realistic or consistent set of initial phases in Yuen (2011e), Sect. 2.1, (A), Sect. 2.2, or Sect. 2.3, for point atoms. Use large oversampling ratio $2^3 = 8$. Divide the experimental | Fourier coefficients | and in-between | Fourier coefficients | by $\hat{f}(\mathbf{h})$. From (6), the crystal is an approximate point-atom crystal. Apply iterative algorithms with oversampling in Sect. 2.1 and 2.2. The coordinates of all non-hydrogen point-atoms are determined. From the many results in Sect. 2.1 and 2.2, we have more confidence whether the procedure can or cannot recover the coordinates of the non-hydrogen atoms. With the large number of | Fourier coefficients | and initial in-between | Fourier coefficients | , and eleven to sixteen strong constraints (Sect. 2.1) and the iterative scheme in Sect. 2.2, it is reasonable to expect that the locations of the atoms can be determined, starting from the heaviest atoms, and that the method applies to macromolecular crystals. This may be a very effective method.

2.5. Determination of the *ab initio* electron density and hydrogen and non-hydrogen atomic coordinates

After the atomic coordinates of the non-hydrogen point atoms are determined, follow Sect. 2.3, and iterative algorithms with oversampling will converge to a unique electron density. with R -index = 0. From this *ab initio* electron density, hydrogen and non-hydrogen atomic coordinates are obtained. The non-hydrogen atomic coordinates obtained from this electron density will be close to, and are more accurate than the point-atom coordinates. The in-between | Fourier coefficients | are refined because of the consistent condition (Yuen (2011a), Sect. 2.1).

2.6. Recovery of missing atoms and improvement of bond lengths and bond angles

Following Yuen (2011a), Sect. 2.5 and Yuen (2011b), Sect. 2.4, the in-between | Fourier coefficients | and | Fourier coefficients | are adjusted within their uncertainties to obtain all the missed atoms, and reasonable bond lengths and bond angles.

2.7. Molecular and solvent boundary

Follow Yuen (2011f), Sect. 2.8. A gross molecular and solvent boundary is obtained. This can be used as a loose support for the iterative algorithms in Sect. 2.1, 2.2 and 2.4.

3. Discussions and conclusion

Yuen (2011f) and this article develop the technology of iterative algorithms with oversampling that will lead to the unique electron density. It is achieved by means of a natural and systematic study of structure determination of crystals by means of iterative algorithms with oversampling, via point-atom model. As atomic scattering factors have approximately the same shape (see (5)), the crystal is represented by approximate point atoms (see (6)). The locations of these point atoms are determined by means of iterative algorithms with oversampling. Large oversampling ratio $2^3 = 8$ is used. All the ｜Fourier coefficients｜ and initial in-between ｜Fourier coefficients｜ are used to determine the atomic coordinates of the point atoms. This number is much larger than the number of atomic coordinates of the point atoms. The system is excessively over-determined. With eleven to sixteen strong constraints employed (see Sect. 2.1), it is reasonable to expect that atomic coordinates for macromolecules can be determined by this method. Moreover, an iteration scheme is used. It concentrates on changing the **heaviest atoms** to point atoms, the atomic coordinates of which are then determined. The second heaviest atoms are then changed to point atoms, and their coordinates determined, so on. The atomic coordinates of all non-hydrogen point-atoms are determined. This may be a very effective method.

The electron density of the crystal with R -index = 0 is then obtained, using Yuen (2011a), Sect. 2.4 or Yuen (2011b), Sect. 2.3. The electron density obtained is *ab initio*. It is determined by the experimental lattice constants, diffraction intensities and initial in-between ｜Fourier coefficients｜ ; no other chemical or physical information (for example, atomicity, chemical formula, bond length, bond angle, etc) is used. Moreover, the in-between ｜Fourier coefficients｜ are also refined because of the consistent condition. From this electron density, hydrogen and non-hydrogen atomic coordinates are obtained. For a crystal structure reported in the literature, these atomic coordinates is an improvement of the atomic coordinates reported in the literature. For an unknown crystal structure, the non-hydrogen atomic coordinates obtained from this *ab initio* electron density will be close to, and is more accurate than the point-atom coordinates.

As we have obtained an electron density with R -index = 0, we have made full use of the ｜Fourier coefficients｜ . Hence it is meaningful to have Sect. 2.6: Recovery of missing atoms and improvement of bond lengths and bond angles.

Let $\delta>0$. In low-density elimination, after density $< \delta$ is set equal to zero, an atom has a sharp boundary of δ. It is sharper and has its height above its surrounding increased by δ. In charge-flipping algorithms, after charge $< \delta$ is flipped, an atom has a sharp boundary of 2δ. It is sharper and has its height above its surrounding increased by 2δ. These accelerate the convergence of the iterations. 'Sharper' and 'higher' are two main reasons that the charge-flipping algorithms and low-density elimination algorithms work. In this way, the atomicity is used directly and explicitly. In point-atom model, the density concentrated at the centre of the circle is 'sharp' and 'high'. After the negative density between the circles of the point atoms is set equal to zero, and the positive density flipped, the circles of the point atoms have 'sharper' boundaries, and the atoms are 'higher'. The point-atom constraint is extremely strong and powerful because besides the N points for the point atoms, in the whole region of a unit cell, the electron density is zero. In Oszlányi & Sütő (2005), p. 147, it is stated 'Large intermolecular voids are an advantage, they act as a natural support for the structure even if their position is not known in advance'. The point-atom constraint is much stronger than the non-negativity constraint, and point-atom model may be more effective than the charge-flipping method or low-density elimination method. It is reasonable to expect that with the point-atom constraint and other constraints, and the large number of $|$ Fourier coefficients $|$ and in-between $|$ Fourier coefficients $|$, and the iterative scheme in Sect. 2.2, the point-atom coordinates of macromolecular crystals can be determined.

In the application of iterative algorithms with oversampling to the point-atom crystal, the chemical informations are used directly and explicitly: (iiib) atomic radii, (iv) shortest interatomic distance, (v) structure constraint. If, during the iterations, some heavy atoms or **partial structures** are identified, they will be employed. In Oszlányi & Sütő (2008), p. 131, it is stated, 'A good part of the crystallographic literature deals with the question of how to use various pieces of known information in an optimal way'.

The solvent flattening method 'is based on the assumption that average density within the protein region should be higher than that of the solvent region' (Woolfson & Fan (1995), p. 147). Such assumption is not used in Sect. 2.7 for determination of a gross molecular and solvent boundary (see Yuen (2011f), Sect. 2.8). These two methods may be compared empirically.

It will be very interesting if we can use, for powder diffraction, iterative algorithms with oversampling applied to point-atom crystals.

It is stated in Oszlányi & Sütő (2008), p. 133, 'An algebraic extraction of the electron density from nothing but the known scattering intensities is hopeless for systems of this size; the only imaginable way is *via* an iterative algorithm'. Determination of crystal electron density and atomic coordinates of macromolecule via point-atom model and iterative algorithms with oversampling may be a very effective method. This remains to be investigated by applying to many crystals.

Point-atom model may be employed in the application of iterative algorithms with oversampling to fixed fibre or membrane (Yuen (2011c, 2011d).

The previous articles Yuen (2011e, 2011f) and this article propose many researches. Interested researchers are welcome to pursuit the investigations.

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23

Equal point-atom model of crystals. Iterative algorithms without oversampling

June 29, 2011

Yuen (2011) presents a systematic study of the equal point-atom crystals containing only one atomic species. A powerful tool of determining crystal electron density and atomic coordinates via equal point-atom model in combination with iterative algorithms and oversampling is presented. In-between | Fourier coefficients | and large oversampling ratio $2^3 = 8$ are employed.

If in-between | Fourier coefficients | , hence oversampling, are not employed, Yuen (2011) and iterative algorithms without oversampling can still be applied. The strong and powerful constraints of equal point-atom model of crystals:

number constraint
equal-height constraint
atomic radius constraint
point-atom constraint.
interatomic distance constraint
structure constraint,

and the powerful low-density elimination and charge flipping can be employed. The main purpose will be the determination of atomic coordinates.

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24

Determination of crystal atomic coordinates via point-atom model. Iterative algorithms without oversampling

June 29, 2011

In Yuen (2011), crystal electron density and atomic coordinates are determined via a point-atom model in combination with iterative algorithms and oversampling. In-between | Fourier coefficients | and large oversampling ratio $2^3 = 8$ are employed.

If in-between | Fourier coefficients | , hence oversampling, are not employed, Yuen (2011) and iterative algorithms without oversampling can still be applied. The strong and powerful constraints of point-atom model of crystals:

number constraint
height constraint
atomic radius constraint
point-atom constraint.
interatomic distance constraint
structure constraint,

and the powerful low-density elimination and charge flipping can be employed. The main purpose will be the determination of atomic coordinates. The electron density determined will not be as accurate.

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25

Phase-retrieval method in deconvolution

March 4, 2008

By means of phase-retrieval method, solution is presented for two deconvolution problems: autocorrelation and convolution, including periodic convolution. This is applied to determination of system function and blind deconvolution.

Keywords: Phase retrieval; autocorrelation; convolution; periodic convolution; empirical system function; blind deconvolution.

1. Introduction

Deconvolution is needed in many fields of sciences, optical, electrical, X-ray crystallography, etc., and methods have been devised for this purpose.

The problem of error in deconvolution is discussed in ref. 1.

Deconvolution employing zero sheets is presented in refs 2-6.

Deconvolution employing point zeros is presented in ref. 7.

Blind deconvolution is discussed in refs 8-13.

Multiple-image deconvolution is presented in refs 14-16.

Non-Gaussian deconvolution is discussed in ref. 17.

Application of deconvolution is presented in refs 18-29.

Maximum entropy is discussed in ref. 30.

Because the phase of the output function cannot be measured, deconvolution is difficult. By means of phase-retrieval method, the phase can be found from the magnitude of the output function. This article presents solutions of two deconvolution problems with unknown phase by means of phase-retrieval method: autocorrelation and convolution, including periodic convolution. The input function may be nonperiodic or periodic, centrosymmetric or non-centrosymmetric, real or complex. These solutions are applied to determination of system function and blind deconvolution,. Refs 31 and 32 present solutions of the phase problem for non-centrosymmetric and complex objects with sharp boundary. Deconvolution method can be used to provide solution for objects without sharp boundary.

In this article, $f(\mathbf{x})$ is the input function, $k(\mathbf{x})$ the output function.

2.1. Autocorrelation

(A) Nonperiodic object $f(\mathbf{x})$

Let $k(\mathbf{x}) = f(\mathbf{x}) \otimes f^*(-\mathbf{x})$

$\otimes$ denotes convolution.

* denotes complex conjugate.

$k(\mathbf{x})$ is the autocorrelation of $f(\mathbf{x})$.

Let $K(\boldsymbol{\omega})$ and $F(\boldsymbol{\omega})$ be the Fourier transforms of $k(\mathbf{x})$ and $f(\mathbf{x})$ respectively.

$$K(\boldsymbol{\omega}) = F(\boldsymbol{\omega})F^*(\boldsymbol{\omega}) = \left|F(\boldsymbol{\omega})\right|^2 \quad (1)$$

Given $K(\boldsymbol{\omega})$ for all $\boldsymbol{\omega}$. $\left|F(\boldsymbol{\omega})\right|$ is found. The problem is to find $f(\mathbf{x})$.

If the object satisfies constraints:

(i) finite size. Size will be determined

(ii) real

(iii) centrosymmetric

(iv) bounded,

this is ref. 33, case (C). Phase of $F(\boldsymbol{\omega})$ is found from $\left|F(\boldsymbol{\omega})\right|$ for all $\boldsymbol{\omega}$. The solution is unique.

If the object satisfies constraints:

(i) finite size. Size will be determined.

(ii) non-negative

(iii) bounded

(iv) sectionally continuous

(v) sharp boundary,

or, the object is complex with the real part satisfying constraints (i) – (v), and, for the imaginary part, similar constraints, and

(vi) the supports of the real part and the imaginary part are the same

These are cases (C) of refs 31 and 32 respectively. The solution is very probably unique.

(B) Periodic object $f(\mathbf{x})$ with known unit cell constants

As in (A), while periodic or circular convolution is used (ref. 34, pp. 133, 217, 263):

$$k(\mathbf{x}) = \int f(\mathbf{y}) f^*(-(\mathbf{x}-\mathbf{y}))d\mathbf{y}$$

The integration is over one unit cell.

$k(\mathbf{x})$ is also a periodic function with same unit cell constants.

Let $U = abc$, where a, b, c are the unit cell edges, c_{klmn} and c_{flmn}, $l, m, n = 0, \pm 1, \pm 2, ...$, be the Fourier coefficients of $k(\mathbf{x})$ and $f(\mathbf{x})$ respectively.

$$c_{klmn} = Uc_{flmn}c_{flmn}{}^* = U\left|c_{flmn}\right|^2$$

corresponding to (1) (ref. 34, (4.133)). $k(\mathbf{x})$ is the Patterson function in X-ray crystallography.

Given c_{klmn}, $l, m, n = 0, \pm 1, \pm 2, ...$ $\left|c_{flmn}\right|$ is found. The problem is to find $f(\mathbf{x})$, solution of the phase problem in X-ray crystallography. This is case (D) in refs 31, 32 or 33. The solution is unique for centrosymmetric object and very probably unique for non-centrosymmetric or complex object.

2.2. Convolution

Let $k(\mathbf{x}) = f(\mathbf{x}) \otimes h(\mathbf{x})$

For a linear system, $h(\mathbf{x})$ is the unit impulse response.

The Fourier transform of $h(\mathbf{x})$ is the system function $H(\boldsymbol{\omega})$.

$$K(\boldsymbol{\omega}) = F(\boldsymbol{\omega})H(\boldsymbol{\omega}) \tag{2}$$

Given $|K(\boldsymbol{\omega})|$, for all $\boldsymbol{\omega}$, and $h(\mathbf{x})$, for all $\mathbf{x}$. The problem is to find the phase of $K(\boldsymbol{\omega})$ and the object $f(\mathbf{x})$.

(C) Nonperiodic $f(\mathbf{x})$ and $h(\mathbf{x})$

Let $f(\mathbf{x})$ and $h(\mathbf{x})$ satisfy the constraints:

(i) finite size. Size (shape and dimensions) of $f(\mathbf{x})$ will be determined.

(ii) real

(iii) centrosymmetric

(iv) bounded,

Then $k(\mathbf{x})$ also satisfies similar constraints.

From $|K(\boldsymbol{\omega})|$, phase of $K(\boldsymbol{\omega})$ is found. The solution is unique (ref. 33, case (C)).

$$F(\boldsymbol{\omega}) = K(\boldsymbol{\omega})/H(\boldsymbol{\omega}) \tag{3}$$

The object $f(\mathbf{x})$ is found.

Let $f(\mathbf{x})$ satisfies the constraints:

(i) finite size. Size will be determined.

(ii) non-negative

(iii) bounded

(iv) sectionally continuous

or, $f(\mathbf{x})$ is complex with the real part satisfying constraints (i) – (iv), and, for the imaginary part, similar constraints, and

(vi) the supports of the real part and the imaginary part are the same

Let $h(\mathbf{x})$ satisfies the constraints:

(i) infinite size

(ii) non-negative

(iii) bounded

(iv) sectionally continuous

$k(\mathbf{x}) = f(\mathbf{x}) \otimes h(\mathbf{x})$

We deal with only a finite portion $k_t(\mathbf{x})$ of $k(\mathbf{x})$, with sharp boundary. t stands for truncated. Let $K_t(\boldsymbol{\omega})$ be the Fourier transform of $k_t(\mathbf{x})$. From $|K_t(\boldsymbol{\omega})|$, phase of $K_t(\boldsymbol{\omega})$ is found. The solution is very probably unique (refs 31, 32, cases (C)).

$$F(\boldsymbol{\omega}) \approx K_t(\boldsymbol{\omega}) / H(\boldsymbol{\omega}) \tag{4}$$

Approximate solution of the object $f(\mathbf{x})$ is found.

(D) Nonperiodic $f(\mathbf{x})$ and periodic $h(\mathbf{x})$ with known unit cell constants. $f(\mathbf{x})$ can be contained in a unit cell of $h(\mathbf{x})$.

If $f(\mathbf{x})$ satisfies the constraints:

(i) finite size. The object $f(\mathbf{x})$ can be contained in a unit cell of $h(\mathbf{x})$. Size of $f(\mathbf{x})$ will be determined.

(ii) real

(iii) centrosymmetric

(iv) bounded,

use a real, centrosymmetric and bounded $h(\mathbf{x})$.

$$k(\mathbf{x}) = \int f(\mathbf{y}) h(\mathbf{x} - \mathbf{y})) d\mathbf{y} = f(\mathbf{x}) \otimes h(\mathbf{x}) \tag{5}$$

$k(\mathbf{x})$ is a periodic function with the same unit cell constants as $h(\mathbf{x})$. This is case (D) of ref. 33. The in-between | Fourier coefficients | along the longest unit cell edge are obtained by interpolation. Find the phase of $K(\boldsymbol{\omega})$ from $|K(\boldsymbol{\omega})|$. The solution is unique. Add the replica of $f(\mathbf{x})$ successfully to give a periodic function. Let c_{klmn}, c_{flmn} and c_{hlmn} $l, m, n = 0, \pm 1, \pm 2, \ldots$, be the Fourier coefficients of $k(\mathbf{x})$, the extended $f(\mathbf{x})$, and $h(\mathbf{x})$ respectively.

$$c_{flmn} = \frac{c_{klmn}}{Uc_{hlmn}} \tag{6}$$

if $c_{hlmn} \neq 0$ (ref. 34, (4.133)).

$U = abc$, where a, b, c are the unit cell edges

If $c_{hlmn} = 0$, set this c_{flmn} as parameter to be determined. Use the Fourier series to obtain an extended $f(\mathbf{x})$. Adjust this Fourier coefficient c_{flmn} to satisfy the constraints for $f(\mathbf{x})$. $f(\mathbf{x})$ is then obtained. The solution is very probably not unique. Therefore, use $h(\mathbf{x})$ with non-zero Fourier coefficients.

From the Fourier coefficients determined in (6), $f(\mathbf{x})$ is obtained.

If the object satisfies constraints:

(i) finite size. The object $f(\mathbf{x})$ can be contained in a unit cell of $h(\mathbf{x})$. Size of $f(\mathbf{x})$ will be determined.

(ii) non-negative

(iii) bounded

(iv) sectionally continuous,

or, the object $f(\mathbf{x})$ is complex with the real part satisfying constraints (i) – (iv), and, for the imaginary part, similar constraints, and

(vi) the supports of the real part and the imaginary part are the same

Use a non-negative, bounded and sectionally continuous $h(\mathbf{x})$ with non-zero Fourier coefficients. Use (5). These are cases (D) of refs 31 and 32 respectively. Find the phase of $K(\boldsymbol{\omega})$ from $|K(\boldsymbol{\omega})|$. The solution is very probably unique.

From the Fourier coefficients determined in (6), $f(\mathbf{x})$ is obtained.

Note that the object $f(\mathbf{x})$ may or may not have sharp boundary. $f(\mathbf{x})$ can taper to zero. The periodicity of $h(\mathbf{x})$ is used to form a periodic function $k(\mathbf{x})$ with the same unit cell constants as $h(\mathbf{x})$. A unit cell of $k(\mathbf{x})$ can then be chosen to have sharp boundary. Hence solution of a non-centrosymmetric or complex object $f(\mathbf{x})$ without sharp boundary can be obtained by means of a periodic function $h(\mathbf{x})$. This is a supplement of refs 31 and 32.

(E) Periodic or circular convolution. Periodic $f(\mathbf{x})$ and $h(\mathbf{x})$ with same unit cell constants.

Let $f(\mathbf{x})$ and $h(\mathbf{x})$ satisfy the constraints:

(a) (i) periodic with same known unit cell constants
  (ii) real
  (iii) centrosymmetric
  (iv) bounded
  (v) the Fourier coefficients of $h(\mathbf{x})$ are non-zero,

or

(b) (i) periodic with same known unit cell constants
  (ii) non-negative
  (iii) bounded
  (iv) sectionally continuous,
  (v) the Fourier coefficients of $h(\mathbf{x})$ are non-zero,

or

(c) $f(\mathbf{x})$ is complex with the real part satisfying constraints (i) – (iv) of (b), and, for the imaginary part, similar constraints, and
  (vi) the supports of the real part and the imaginary part are the same
  $h(\mathbf{x})$ satisfies the constraints in (b).

Periodic or circular convolution is used (ref. 34, pp. 133, 217, 263):

$$k(\mathbf{x}) = \int f(\mathbf{y})h(\mathbf{x}-\mathbf{y})d\mathbf{y}$$

The integration is over one unit cell.

$k(\mathbf{x})$ is also a periodic function with same unit cell constants. Find the phase of $K(\boldsymbol{\omega})$ from $|K(\boldsymbol{\omega})|$. For (a), the solution is unique. For (b) and (c), the solution is very probably unique.

Let c_{klmn}, c_{flmn} and c_{hlmn} $l,m,n = 0,\pm1,\pm2,\ldots$, be the Fourier coefficients of

$k(\mathbf{x})$, $f(\mathbf{x})$, and $h(\mathbf{x})$ respectively. $U = abc$, where a, b, c are the unit cell edges

From the Fourier coefficients determined in (6), $f(\mathbf{x})$ is obtained.

2.3. Determination of system function $H(\boldsymbol{\omega})$

For a linear system, the theoretical system function may be difficult to calculate accurately, and in applications it may be affected by other factors, for example, optical blurring. Deconvolution method may be used to determine the system function empirically. For centrosymmetric or non-centrosymmetric, real or complex, nonperiodic or periodic $h(\mathbf{x})$, following the treatment in above, and use appropriate known objects $f(\mathbf{x})$, for example, a point source. From $|K(\boldsymbol{\omega})|$, find the phase of $K(\boldsymbol{\omega})$. From (2), the empirical system function $H(\boldsymbol{\omega})$ and hence the empirical unit impulse response $h(\mathbf{x})$ are found. Other factors in the system are included in $H(\boldsymbol{\omega})$ (see ref. 35, Sect.6.4, aberrations of lens). Hence, even if the system is not perfect, using this empirical system function, the structure of an unknown object can still be determined.

2.4. Blind deconvolution

$$k(\mathbf{x}) = f(\mathbf{x}) \otimes h(\mathbf{x})$$
$$K(\boldsymbol{\omega}) = F(\boldsymbol{\omega})H(\boldsymbol{\omega}) \tag{7}$$

Given $|K(\boldsymbol{\omega})|$ for all $\boldsymbol{\omega}$. The problem is to find the phase of $K(\boldsymbol{\omega})$, $f(\mathbf{x})$ and $h(\mathbf{x})$.

Consider general and reasonable constraints: centrosymmetric, real, non-negative, complex, nonperiodic, periodic, finite size, bounded, continuity, differentiability, analyticity, sharp boundary, etc. Choose appropriate constraints for $f(\mathbf{x})$ and $h(\mathbf{x})$ (see for example, ref. 21, Ch. 4). From $|K(\boldsymbol{\omega})|$, find the phase of $K(\boldsymbol{\omega})$. The constraints chosen limit the choice of $F(\boldsymbol{\omega})$ and $H(\boldsymbol{\omega})$ in (7).

3. Conclusion

By means of phase-retrieval method, solution is presented for two deconvolution problems with unknown phase of the output function: autocorrelation and convolution, including periodic convolution. These solutions are applied to determination of system function and blind deconvolution. The problem of noise is not dealt with. Solution of the phase problem for non-centrosymmetric and complex objects without sharp boundary is provided. Case (D) has an important application of transforming an nonperiodic object to a crystal.

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X-ray crystallography

(in-between Fourier coefficients not employed)

(In-between Fourier Coefficients Not Employed)

26

Sharpening of Patterson function of a crystal.

Lowest Patterson function. Iterative algorithms and superresolution by extrapolation

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Abstract

This article presents a general and effective method of sharpening the Patterson function so that the atomic coordinates of crystal containing large number of atoms can be determined. The effect of Fourier series truncation is also treated. Various sharpening of the Patterson function of a crystal is used, and the lowest Patterson function introduced. A general method of sharpening is then presented. We found that: The investigation of sharpening is for all crystals. The result can then be used for any specific crystal, as needed. We show an illustration employing Tchebycheff approximation. The effects of Fourier series truncation are due to truncation of the sharpened peak ${}^{s}G(\mathbf{r})$. The ripples are wiped out when all non-negative sharpening Patterson functions are used. To reduce the half-widths of the peaks, iterative algorithms and superresolution by extrapolation are employed. Using point atom-pairs, all the peaks of the Patterson function of a crystal may be resolved.

Keywords: Atomicity; periodic convolution; circular convolution; autocorrelation; ripples; Tchebycheff approximation; Fourier series truncation; series termination; half-widths; superresolution by extrapolation; point atom-pair.

1. Introduction

Making use of atomicity, Patterson introduces the Patterson function which has great value in solving crystal atomic coordinates. Because of the large number of the peaks, each corresponding to an interatomic pair, and the sizes of the atoms, there is much overlapping of peaks. Hence it is difficult to resolve the Patterson peaks for crystals containing large number of atoms. As the Patterson function is very useful and important in determination of crystal atomic coordinates, this article presents a general and effective method of sharpening the Patterson function so that the atomic coordinates of crystal containing large number of atoms can be determined. The effect of Fourier series truncation or termination is also treated.

After the peaks of the Patterson function are resolved, the crystal atomic coordinates can then be determined from these peaks (see, for example, Buerger (1959), Tokonami & Hosoya (1965), Raman & Katz (1967a), Ramachandran & Srinivasan (1970), Ch. 3, Soldatov et al. (1978), Rossmann & Arnold (2001), Burla et al. (2006, 2007b), Burla et al. (2007a), Caliandro et al. (2007a), Feng (2009), Milanesio & Viterbo (2011), Appendix 6.B, and, for powder Patterson map, Estermann & David (2006), Sect. 12.6-12.8, Burla et al. (2007c), Altomare et al. (2009)). Simonov et al. (2006) determinates atomic structure of nanosized particles using diffuse-scattering data.

Sharpening of the Patterson function is presented in Lipson & Cochran (1966), p. 165-169, Ramachandran, G. N. & Srinivasan, R. (1970), p. 20-23, Ladd & Palmer (1985), p. 237-239, 296, Stout, G. H. & Jensen, L. H. (1989), p. 281-283, Woolfson (1997), p. 249 and Milanesio & Viterbo (2011), p. 426. Gradient Patterson function is used in Jacobson et al. (1961), Wunderlich (1965). Raman & Katz (1967b) presents an analytical method of obtaining sharpened Patterson functions. It deals with crystal structures with rational atomic coordinates. The method is different from that in this article. Goldak (1969, 1971, 1974) presents extraction of the periodic set from the Patterson function by means of generalized polynomial and Tchebycheff approximation. Waser & Schomaker (1953) presents an extensive treatment of modification functions.

For correction of Fourier series truncation error, see Raman & Katz (1967b), Wilson (1970), Sect. 9-5, Caliandro et al. (2005a, 2005b), Yao et al. (2005), Caliandro et al. (2007b), Usón et al. (2007), Caliandro et al. (2008), Milanesio & Viterbo (2011), Sect. 6.3.2.1 and Zanotti (2011), p. 733.

This article studies the sharpening of the Patterson function of a crystal. In Sect. 2.1, various sharpening of the Patterson function is discussed, and the lowest Patterson function introduced (not to be confused with the minimum function (Buerger (1959), pp. 239-247, Woolfson (1997), p. 249). The problem of sharpening the Patterson function reduces to the problem of using all the sharpening functions such that the sharpened Patterson functions are non-negative.

In Sect. 2.2, a general method of sharpening is presented. We found that: The investigation of sharpening is for all crystals. The result can then be used for any specific crystal, as needed. We show an illustration employing Tchebycheff approximation. The effects of Fourier series truncation are due to truncation of the sharpened peak ${}^{s}G(\mathbf{r})$. When all sharpened Patterson functions are used, and the lowest Patterson function obtained, the ripples are wiped out.

To reduce the half-widths of the peaks, iterative algorithms and superresolution by extrapolation are employed in Sect. 2.3. Using point atom-pairs, all the peaks of the Patterson function of a crystal may be resolved.

The peak at the origin and all known Patterson peaks may be removed (Sect. 2.4).

2.1. Various sharpening of Patterson function of a crystal. Lowest Patterson function

Assume that a very large number of reflections are obtained; very high resolution is obtained and the errors due to the Fourier series truncation of the sharpened Patterson functions are negligible. Because of the atomicity, the Patterson function shows peaks at positions of the interatomic vectors with height approximately proportional to the product of the atomic numbers of the two atoms (Ladd & Palmer (1985), p. 233, Stout & Jensen (1989), p. 283, Woolfson & Fan (1995), p. 35). This is the reason why the Patterson function of a crystal is very useful. The maximum width of a peak in the Patterson function is equal to the sum of the maximum widths of the two atoms involved (Stout & Jensen (1989), p. 280-281).

Consider real atoms. We will use a sharpening function such that the sharpened Patterson function is non-negative.

For example, choose (atomic scattering factor of the heaviest atom)$^{-2}$. $F(\mathbf{h})$ is the structure factor. Multiply all the observed $|F_o(\mathbf{h})|^2$ by this sharpening function. In this sharpened Patterson function, the peaks involving two heaviest atoms are very high peaks with very small radius. Record the locations and heights of these peaks.

Then choose (atomic scattering factor of the heaviest atom × atomic factor of the second heaviest atom)$^{-1}$. Multiply all the observed $|F_o(\mathbf{h})|^2$ by this sharpening function. In this sharpened Patterson function, the peaks involving a heaviest atom and a second heaviest atom are very high peaks with very small radius. Record the locations and heights of these peaks.

Then use (atomic scattering factor of the second heaviest atom)$^{-2}$ as the sharpening function, so on.

Let Z_n and $f_n(\mathbf{h})$ be the atomic number and atomic scattering factor of the n-th atom respectively. The atomic scattering factors have approximately same shapes. Let

$$\hat{f}(\mathbf{h}) = (\sum f_n(\mathbf{h}))/(\sum Z_n) \tag{1}$$

$\hat{f}(\mathbf{h})$ is the atomic scattering factor of a hypothetical atom with $Z = 1$.

$$f_n(\mathbf{h}) \approx Z_n \hat{f}(\mathbf{h}) \tag{2}$$

After we have used all these product of atomic scattering factors, we use

$[\hat{f}(\mathbf{h})]^{-2}$ as a sharpening function, and obtain a sharpened Patterson function. Record the peaks.

Then use $[\sum f_n^2(\mathbf{h})]^{-1}$ as a sharpening function. Record the peaks of the sharpened Patterson function.

Then use $[\sum f_n^m(\mathbf{h})]^{-2/m}$ for any positive integer m >2, as a sharpening function.

Record the peaks of the sharpened Patterson function.

A sharpening function maintains the atomicity of the crystal and keeps the interatomic distances unchanged. The idea is to use all sharpening functions that we can think of. After we obtain all sharpened Patterson functions, multiply each sharpened Patterson function by a constant so that the height of the peak at the origin is the same as that of the Patterson function. Superimpose all these Patterson function and sharpened Patterson functions. Insert all the peaks that we have recorded in above. As the Fourier series truncation-error is negligible, and the sharpened Patterson functions are non-negative, the value of a sharpened Patterson functions is as valid as the values of the Patterson functions and other sharpened Patterson functions. Hence at the other points of the unit cell, we can use the lowest value of these functions. From this resultant function, see if there are any peaks not shown in the Patterson function or sharpened Patterson functions.

The problem of sharpening the Patterson function reduces to the problem of using all the sharpening functions such that the sharpened Patterson functions are non-negative. The resultant function obtained may be called the lowest Patterson function of a crystal. This is a sharpened Patterson function.

2.2. General method of sharpening

To find all the sharpening functions of a crystal, an analytical method is used.

Consider a crystal of real or complex electron density $\rho(\mathbf{r})$. Form the autocorrelation

$$p(\mathbf{r}) = \rho(\mathbf{r}) \,\overline{\otimes}\, \rho^*(-\mathbf{r}) \tag{3}$$

$$= \int \rho(\mathbf{s})\rho^*(-(\mathbf{r}-\mathbf{s}))d\mathbf{s} = \int \rho(\mathbf{r}+\mathbf{s})\rho^*(\mathbf{s})d\mathbf{s} \tag{4}$$

$\overline{\otimes}$ denotes periodic or circular convolution (Oppenheim et al (1983), pp. 133, 217, 263).

The integration is over one unit cell.

$p(\mathbf{r})$ is also a periodic function with same unit cell constants. $p(\mathbf{r})$ is the Patterson function of the crystal

For a crystal, the structure factor

$$F(\mathbf{h}) = \sum_n f_n(\mathbf{h}) \exp[2\pi i(\mathbf{h}\cdot\mathbf{r}_n)] \tag{5}$$

where $\mathbf{h}$ is a reciprocal lattice vector

$\mathbf{r}$ is position vector of the n-th atom

$$\rho(\mathbf{r}) = \frac{1}{V}\sum_{\mathbf{h}} F(\mathbf{h}) \exp[-2\pi i(\mathbf{h}\cdot\mathbf{r})] \tag{6}$$

Substitute (6) into (4). The Patterson function

$$p(\mathbf{r}) = \frac{1}{V}\sum_{\mathbf{h}} |F(\mathbf{h})|^2 \exp[-2\pi i(\mathbf{h}\cdot\mathbf{r})] \tag{7}$$

V is the volume of one unit cell. The factor V^{-1} is used as in Ladd & Palmer (1985), (6.54), (6.55).

Consider real atoms. The atomic scattering factors have approximately same shapes. Substitute (5) and (2) into (7),

$$p(\mathbf{r}) \approx \frac{1}{V}\sum_{n=1}^{N}\sum_{m=1}^{N} Z_n Z_m G[\mathbf{r} - (\mathbf{r}_m - \mathbf{r}_n)] \tag{8}$$

where $G(\mathbf{r}) = \sum_{\mathbf{h}} \left|\hat{f}(\mathbf{h})\right|^2 \exp[-2\pi i(\mathbf{h}\cdot\mathbf{r})]$ (9)

$G(\mathbf{r})$ has the form of a peak at the origin. From (8), this peak is shifted to each point given by interatomic vectors $\mathbf{r}_m - \mathbf{r}_n$. The magnitude of the peak is modified by $Z_n Z_m$. Thus, this displacement gives rise to the Patterson peaks.

If, in (5), the electrons not only form a cluster around the nuclei but also concentrate in an infinitesimal volume around each nucleus, this gives rise to a system of point atoms. (5) then becomes

$$^{s}F(\mathbf{h}) = \frac{F(\mathbf{h})}{\hat{f}(\mathbf{h})} = \sum_n Z_n \exp[2\pi i(\mathbf{h}\cdot\mathbf{r}_n)] \tag{10}$$

s stands for sharpened.

Replacing $|F(\mathbf{h})|^2$ in (7) by $|{}^{s}F(\mathbf{h})|^2$, we obtain

$$^{s}p(\mathbf{r}) = \frac{1}{V}\sum_{n=1}^{N}\sum_{m=1}^{N} Z_n Z_m G[\mathbf{r} - (\mathbf{r}_m - \mathbf{r}_n)] \tag{11}$$

with $G(\mathbf{r}) = \sum_{\mathbf{h}} \exp[-2\pi i(\mathbf{h} \cdot \mathbf{r})]$ (12)

which represents a point atom-pair.

We intend to transform (8) so that the shapes of the peaks are changed while the displacement property is maintained. In (8) and (9), $G(\mathbf{r})$ is a periodic function with a unit cell of the crystal as a period. To obtain the other sharpening functions, we use a periodic function $K(\mathbf{r})$ with the same period. Consider a transform of $p(\mathbf{r})$:

Let $^{s}p(\mathbf{r}) = p(\mathbf{r}) \overline{\otimes} K(\mathbf{r})$ (13)

$$= \int p(\mathbf{s}) K(\mathbf{r} - \mathbf{s}) d\mathbf{s} \tag{14}$$

The integration is over one unit cell. Substitute (8) into (14),

$$^{s}p(\mathbf{r}) \approx \frac{1}{V} \sum_{n=1}^{N} \sum_{m=1}^{N} Z_n Z_m \, ^{s}G[\mathbf{r} - (\mathbf{r}_m - \mathbf{r}_n)] \tag{15}$$

where $^{s}G(\mathbf{r}) = G(\mathbf{r}) \overline{\otimes} K(\mathbf{r})$ (16)

(15) has exactly the displacement property exhibited in (8). It has sharpened peaks $^{s}G(\mathbf{r})$ (see (16)) at interatomic vectors $\mathbf{r}_m - \mathbf{r}_n$. $^{s}p(\mathbf{r})$ is a sharpened Patterson function.

Use the Fourier series of $K(\mathbf{r})$,

$$K(\mathbf{r}) = \sum_{\mathbf{h}} q(\mathbf{h}) \exp[-2\pi i(\mathbf{h} \cdot \mathbf{r})] \tag{17}$$

Substitute (9) and (17) into (16),

$$^{s}G(\mathbf{r}) = \sum_{\mathbf{h}} \left| \hat{f}(\mathbf{h}) \right|^2 q(\mathbf{h}) \exp[-2\pi i(\mathbf{h} \cdot \mathbf{r})] \tag{18}$$

Compare (18) with (9), we see that the net result of the transformation (14) is the introduction of the coefficients $q(\mathbf{h})$ into (18). These coefficients can be adjusted so that $^{s}G(\mathbf{r})$ is a peak of satisfactory shape, while the displacement property is maintained (see (15)).

Conversely, if, from the beginning, we modify the Patterson function (7) by defining

$$^{s}p(\mathbf{r}) = \frac{1}{V}\sum_{\mathbf{h}} \left|F(\mathbf{h})\right|^{2} q(\mathbf{h}) \exp[-2\pi i(\mathbf{h}\cdot\mathbf{r})] \tag{19}$$

From (5) and (2), $F(\mathbf{h}) \approx \sum_{n} Z_n \hat{f}(\mathbf{h}) \exp[2\pi i(\mathbf{h}\cdot\mathbf{r}_n)]$

Substitute this into (19), we obtain (15) with $^{s}G(\mathbf{r})$ given by (18). Hence, the displacement property is maintained when coefficients $q(\mathbf{h})$ are introduced into the Patterson function (see (19)). This is pointed out by Lipson & Cochran (1966), p. 166. Moreover, (14) is obtained (Waser & Schomaker (1953), p. 686).

For the sharpened peak $^{s}G(\mathbf{r})$ in (18) and the sharpened Patterson function $^{s}p(\mathbf{r})$ in (15) to be real, the coefficients $q(\mathbf{h}) = [q(-\mathbf{h})]^{*}$.

From (19), the Fourier coefficient of the sharpened Patterson function $^{s}p(\mathbf{r})$,

$$= \frac{1}{V}\left|F(\mathbf{h})\right|^{2} q(\mathbf{h})$$

In general, $q(\mathbf{h})$ is complex. Hence the Fourier coefficients of the sharpened Patterson function are complex. This is different from the Fourier coefficients of the Patterson function, which are non-negative (see (7)). Hence we have the conclusion: For a sharpened Patterson function of a crystal to be the Patterson function of an electron density distribution of the crystal, $q(\mathbf{h})$ must be non-negative.

If $q(\mathbf{h}) = 1/[\hat{f}(\mathbf{h})]^{2}$, (19) becomes

$$^{s}p(\mathbf{r}) = \frac{1}{V}\sum_{\mathbf{h}} \frac{\left|F(\mathbf{h})\right|^{2}}{[\hat{f}(\mathbf{h})]^{2}} \exp[-2\pi i(\mathbf{h}\cdot\mathbf{r})] \tag{20}$$

This is the sharpened Patterson function of point-atom pairs obtained by substituting (10) into (7).

Choose $q(\mathbf{h}) = c(\mathbf{h})/[\hat{f}(\mathbf{h})]^{2}$. (21)

(18) becomes

$$^{s}G(\mathbf{r}) = \sum_{\mathbf{h}} c(\mathbf{h}) \exp[-2\pi i(\mathbf{h}\cdot\mathbf{r})] \tag{22}$$

$\mathbf{h}$ are all the reciprocal lattice vectors.

This Fourier series gives the most general sharpened peak ${}^{s}G(\mathbf{r})$, and hence the most general sharpening Patterson function ${}^{s}p(\mathbf{r})$ in (15). ${}^{s}G(\mathbf{r})$ is a periodic function with the unit cell as a period. It is for all crystals. Here we have an important conclusion of this article: **Investigation of sharpening is for all crystals. The result can then be used for any specific crystal, as needed.**

Substitute (21) into (19).

$$ {}^{s}p(\mathbf{r}) = \frac{1}{V}\sum_{\mathbf{h}} \frac{|F(\mathbf{h})|^2}{[\hat{f}(\mathbf{h})]^2} c(\mathbf{h}) \exp[-2\pi i(\mathbf{h}\cdot\mathbf{r})] \tag{23} $$

The coefficients $c(\mathbf{h})$ in (22) are chosen to give the desired sharpened peak ${}^{s}G(\mathbf{r})$. With these coefficients $c(\mathbf{h})$ and the experimentally determined $|F(\mathbf{h})|^2$, Eq. (23) is used to obtain the sharpened Patterson function ${}^{s}p(\mathbf{r})$. In (23), instead of the sharpening function $[\hat{f}(\mathbf{h})]^{-2}$, other sharpening functions in Sect. 2.1 can also be used. In particular, use (atomic scattering factor of the heaviest atom)$^{-2}$, (atomic scattering factor of the heaviest atom × atomic factor of the second heaviest atom)$^{-1}$ (atomic scattering factor of the second heaviest atom)$^{-2}$, so on. This will sharpen the peaks in order, so that they can be recognized, and the atomic species identified.

Now consider a non-negative electron density $\rho(\mathbf{r})$. From (4), the Patterson function is non-negative. Consider the case of finite number of diffraction intensities, limited by experimental measurement. There is Fourier series truncation error. There will be large negative and positive ripples in the Patterson function.

We will discuss the method of obtaining the coefficients $c(\mathbf{h})$ in (22), in one dimension. Let $Q(x)$ be a real periodic function with the unit cell length 1 as a period (fractional coordinate is used). $Q(x)$ has peaks at the lattice points. We would like to choose the coefficients $c(h)$ such that ${}^{s}G(x)$ in (22) is a good approximation to $Q(x)$. In order to define the degree of approximation, we define the L_p-norm of a function $g(x)$ defined in the closed interval [0,1] as

$$L_p(g) = \left[\int_0^1 |g(x)|^p \, dx \right]^{1/p} , \quad p \geq 1 \tag{24}$$

If the coefficients, $c(h)$ of ${}^sG(x)$ in (22) are varied such that

$$\left[\int_0^1 \left| {}^sG(x) - Q(x) \right|^p dx \right]^{1/p} \tag{25}$$

is a minimum, we call this ${}^sG(x)$ the best L_p-approximation to $Q(x)$. For example, $p = 2$. ${}^sG(x)$ is then the best L_2-approximation (i.e., least squares) to $Q(x)$. In this case, $c(h)$ are simply the Fourier coefficients of $Q(x)$.

In addition to the L_p-norm, there is the Tchebycheff (Chebyshev) norm, $\|g(x)\|_T$, which is defined as supremum of $|g(x)|$ for $0 \leq x \leq 1$. The Tchebycheff norm is sometimes called the L_∞-norm. If the coefficients, $c(h)$ are varied such that $\left\| {}^sG(x) - Q(x) \right\|_T$ = supremum of $\left| {}^sG(x) - Q(x) \right|$ for $0 \leq x \leq 1$, is a minimum, this ${}^sG(x)$ is called the Tchebycheff approximation, best approximation, or minimax approximation, to $Q(x)$. Discussion of approximation theory may be found in Rice (1964, 1969), Lorentz (1966), Watson (1980), Rivlin (1981) and Cheney (1998).

As an example, we will use the Tchebycheff approximation. A finite number of terms in (22) is used.

$${}^sG(x) = \sum_{h=-H}^{H} c(h) \exp(-2\pi i h x) \tag{26}$$

$Q(x)$ is real. Hence $c(h) = [c(-h)]^*$.

For centrosymmetric real $Q(x)$, $c(h)$ is real and $c(h) = c(-h)$, (27)

$${}^sG(x) = c(0) + \sum_{h=1}^{H} 2c(h) \cos(2\pi h x) \tag{28}$$

A necessary and sufficient condition that ${}^sG(x)$ be a Tchebycheff approximation of

$Q(x)$ is that the error curve $\left|{}^sG(x) - Q(x)\right|$ alternates at least $(H+1)$ times in [0,1], that is: there exists at least $(H+2)$ points,
$x_1 < x_2 < \ldots < x_{H+2}$ ε [0,1], such that

$$ {}^sG(x_i) - Q(x_i) = -[{}^sG(x_{i+1}) - Q(x_{i+1})] $$

$$ = \pm \text{ supremum of } \left|{}^sG(x) - Q(x)\right|, \text{ for } 0 \le x \le 1. $$

$i = 1,2,\ldots,H+1.$

This theorem also applies to a set of discrete points ε [0,1]. This is discussed in Rice (1964), pp. 54-56, Watson (1980), p. 55, Th. 3.3, p. 74, Th. 3.9, and is demonstrated in the following illustration.

For the Tchebycheff approximation of a given real function over a discrete range by a linear combination of M linearly independent continuous real functions, IBM has a subroutine for calculating the M coefficients.

We consider three choices concerning the shape of $Q(x)$:

(A) $Q(x)$ =100 at $x = 0,\pm1,\pm2,\ldots$

$Q(x) = 0$ at $x\varepsilon[\sigma, 1-\sigma]$. σ is a small positive number. $Q(x)$ is periodic with period = 1.

$Q(x)$ is undefined at other points of x. This is to give flexibility to the approximating function ${}^sG(x)$.

The purpose of this choice of $Q(x)$ is to have sharp peaks at the points $x = 0,\pm1,\pm2,\ldots$.

As an illustration, we apply the IBM subroutine to approximate $Q(x)$ by ${}^sG(x)$ in (28). The interval $[0,1]$ is divided into 101 or 501 points to see if there is large discrepancy between the approximating functions obtained. We give the result for $H = 7$, $\sigma = 0.06$:

c_0	c_1	c_2	c_3	c_4	c_5	c_6	c_7
6.7	6.6	6.3	5.9	5.3	4.6	3.9	7.9

$x_i =$	0	0.060	0.093	0.155	0.225	0.290	0.360	0.430	0.500
$Q(x_i)$ =	100	0	0	0	0	0	0	0	0
$^{s}G(x_i)$ =	88	12	-12	12	-12	12	-12	12	-12
$^{s}G(x_i) - Q(x_i)$ =	-12	12	-12	12	-12	12	-12	12	-12

In Sect. 2.1, the sharpened Patterson function used is non-negative. Hence we set all the negative value of $^{s}G(x)$ to zero, and obtain a non-negative sharpened peak $^{s}\overline{G}(x)$. $^{s}\overline{G}(x)$ has intervals of zeros.

For a specific crystal, the non-negative sharpened function $^{s}\overline{p}(x)$ is then obtained from (23).

For different values of σ, the ripple heights and positions change. For all the non-negative sharpened Patterson functions $^{s}\overline{p}(x)$, obtain the lowest Patterson function as in Sect. 2.1. **These shifted zeros of all $^{s}\overline{G}(x)$ wipe out all the ripples.**

(B) $Q(x)$ is a triangle at each lattice point, i.e., for $x\varepsilon[-\sigma,\sigma]$.
$Q(x)$ is undefined at other points of x.

(C) Unsymmetrical $Q(x)$.
$Q(x) = 0$ at $x\varepsilon[-0.2,0)$. $Q(x)$ is periodic with period = 1.
$Q(x) = 100$ at $x = 0,\pm1,\pm2,...$
$Q(x)$ is undefined at other points of x.

The purpose of this choice of $Q(x)$ is to force $^{s}G(x)$ to have peaks very sharp at one side, at the points $x = 0,\pm1,\pm2,...$. This may help in resolving the peaks.

Use all reasonable shapes of $Q(x)$, and obtain the best L_p-approximation to

$Q(x)$ for all $p \geq 1$ (see (25)) .

For three-dimensional crystals, approximation in three dimensions, or product of three one-dimensional approximating functions for the three axes may be used (Rice (1969), Cheney & Light (2000)). For all $Q(\mathbf{r})$, and, in (22), the reciprocal lattice vectors $\mathbf{h}$ used, determine the coefficients $c(\mathbf{h})$, and the non-negative sharpened peak ${}^{s}\overline{G}(\mathbf{r})$. ${}^{s}\overline{G}(\mathbf{r})$ has regions of zeros. As these are for all crystals (see (22)), the results may be tabulated. For a specific crystal, the non-negative sharpened Patterson function ${}^{s}\overline{p}(\mathbf{r})$ is obtained in (23), employing the experimentally determined $|F(\mathbf{h})|^2$, and various sharpening functions, and $c(\mathbf{h})$. For all the non-negative sharpened Patterson functions ${}^{s}\overline{p}(\mathbf{r})$, as in Sect. 2.1, multiply each sharpened Patterson function by a constant so that the height of the peak at the origin is the same as that of the Patterson function. Superimpose all these Patterson function and sharpened Patterson functions. Insert all the peaks in the Patterson function and each sharpened Patterson function. At the other points of the unit cell, use the lowest value of these functions. From this resultant function, see if there are any more peaks not shown in the Patterson function or sharpened Patterson functions. We obtain a lowest Patterson function. The ripples are wiped out by the zeros of all the non-negative sharpened Patterson functions.

If, available, compare the lowest Patterson function with the lowest Patterson function from the Patterson function of a unit cell (Yuen (2011e), Sect. 2.1, 2.2), to see if any more peaks are revealed.

2.3. Fourier series truncation. Iterative algorithms and superresolution by extrapolation

For a crystal structure of one kind of point atoms, the Fourier coefficients will not decline with $(\sin\theta)/\lambda$. Infinite number of terms of the Fourier series will be required (Buerger (1959), p. 61-63). There are two effects due to Fourier series truncation: the peaks have large half-widths and there are large negative and positive ripples.

In (23), only the experimentally determined $\left|F(\mathbf{h})\right|^2$ are used. It has a finite number of terms. Hence in (22), only these finite number of $\mathbf{h}$ is used. Because of this, we have the two effects due to Fourier series truncation: large peak half-widths and large negative and positive ripples. As shown in the illustration in Sect. 2.2, the ripples are due to the ripples of the sharpened peak ${}^{s}G(\mathbf{r})$. Thus, the effects of series truncation are due to truncation of the sharpened peak ${}^{s}G(\mathbf{r})$. Eq. (26) is an example.

When all the non-negative sharpened Patterson functions are used and the lowest Patterson function obtained, the ripples are wiped out (Sect. 2.1, 2.2). Hence the remaining problem is the large half-widths of the non-negative sharpened peak ${}^{s}\overline{G}(\mathbf{r})$. Overlapping of these peaks may cause resolution difficult. To obtain very sharp peaks, more structure factors are needed (see (23)). They can be obtained experimentally or by extrapolation (superresolution). To obtain experimentally a very large number of structure factors is difficult. Hence extrapolation may be used. By employing the lowest Patterson function, the ripples are wiped out. This is possible because, basically the number of observed structure factors is much larger than the number of atomic coordinates. Hence the number of atomic coordinates can be determined. By the same reason, we may be able to resolve the peaks of the Patterson function even though the half-width is large. That is, just the observed structure factors will be sufficient; no more experimental structure factors are needed. Iterative algorithms and superresolution by extrapolation (Yuen (2011a), Sect. 2.3) will be employed.

We intend to divide the lowest Patterson function into point-atom pairs. In the reciprocal space, the observed diffraction intensities multiplied by $[\ \hat{f}(\mathbf{h})\ ]^{-2}$ will be used as the constraint.

In the lowest Patterson function obtained in Sect 2.2, replace each peak by a point atom-pair with height equal to the product of the atomic numbers (see point atom in Yuen (2011c, 2011d). If the peak is due to more than one interatomic pairs, use the sum of their products of atomic numbers. Because a nearby Patterson peak may exist, we use a sphere of very small radius, for example 0.001Å for a peak. The point atom-pair is zero inside this sphere, except the sharp point at the centre. Calculate the Fourier coefficients. There is no initial phase problem. For the reciprocal lattice vectors $\mathbf{h}$ for which the diffraction intensities are measured, replace the Fourier coefficients by the observed diffraction intensities multiplied by $[\,\hat{f}(\mathbf{h})\,]^{-2}$. For the other reciprocal lattice vectors $\mathbf{h}$, use the magnitude of the calculated Fourier coefficients. Calculate the function from these non-negative Fourier coefficients. In this function, see whether we can obtain more peaks. Then replace these peaks in the lowest Patterson function by point-atom pairs. These peaks may involve light atoms. Repeat the iterations until no more peaks can be obtained. A sharpened Patterson function is obtained. Replace all the peaks by point-atom pairs.

To quantify the regions of non-zero values (for examples, plateaus) of this sharpened Patterson function, these regions are kept fixed. Divide these regions of a unit cell into closely packed spheres of same radius. The content of a sphere is the value of the function inside the sphere plus shares from the gaps between the spheres. The gaps between the spheres, is kept at zero value. Replace each sphere by a point atom-pair. Depending on the content of the sphere, the height of the peak at the centre may be equal to sum of products of atomic-numbers. The radius of the spheres is chosen so that all the interatomic pairs are taken into account. Use the iteration. See whether we can obtain more peaks. Then replace these peaks in the lowest Patterson function by point-atom pairs. These peaks may involve light atoms. Repeat the iterations. In the final result, we have only point atom-pairs. The sum of the peak heights represents all the interatomic pairs. This gives the peaks for all interatomic pairs. Hence all the peaks of the Patterson function of a crystal may be resolved.

If result of sharpening the Patterson function of the unit cell (Yuen (2011f) is available, this result can be used to monitor the iterations in this section.

2.4. Removal of the peak at the origin and known Patterson peaks

One variance is to remove the peak at the origin of the Patterson function (Buerger (1959), p. 56-58, Lipson & Cochran (1966), p. 169-170). Use

$$\left|F(\mathbf{h})\right|^2 - \sum_n f_n{}^2(\mathbf{h}) \tag{29}$$

Then apply Sect. 2.1 - 2.2 to this expression. Since the large peak in the origin has been removed, multiply each sharpened Patterson function by a constant so that the height of the highest peaks is the same as those of the Patterson function. Superimpose all these Patterson function and sharpened Patterson functions. As in Sect. 2.1, obtain a lowest Patterson function.

When a pair of Patterson peaks from the n-th and m-th atoms is obtained, one variance is to remove these peaks from the Patterson function (Buerger (1959), p.58-59). Use

$$\left|F(\mathbf{h})\right|^2 - 2f_n(\mathbf{h})f_m(\mathbf{h})\cos[2\pi i\mathbf{h}\cdot(\mathbf{r}_n - \mathbf{r}_m)]. \tag{30}$$

Remove all the known pairs of Patterson peaks.
Then apply Sect. 2.1 - 2.2 to the final expression obtained. A lowest Patterson function is obtained.

One variance is to remove the peak at the origin and the known pair of Patterson peaks by combining expressions (29) and (30). Use

$$\left|F(\mathbf{h})\right|^2 - \sum_n f_n{}^2(\mathbf{h}) - 2f_n(\mathbf{h})f_m(\mathbf{h})\cos[2\pi i\mathbf{h}\cdot(\mathbf{r}_n - \mathbf{r}_m)].$$

Remove all the known pairs of Patterson peaks.
Then apply Sect. 2.1 - 2.2 to the final expression obtained. Use the height of the highest peaks. A lowest Patterson function is obtained.

Add the peaks obtained in these variances to the lowest Patterson function obtained in Sect. 2.2. Then apply Sect. 2.3 to the resultant lowest Patterson function.

2.5. Demonstrations with crystal structures reported in the literature.

Choose a large number of crystal structures reported in the literature, of various sizes, small, mediate, or macromolecular, and with or without heavy atoms. Apply Sect. 2.1, 2.2, and 2.3. Find all non-negative sharpening functions. Obtain the lowest Patterson function. Examine application of iterative algorithm. Use superresolution by extrapolation to deal with the Fourier series truncation problem. Examine the result of resolving all Patterson peaks.

3. Discussions and conclusion

In Sec. 2.1, various sharpening of the Patterson function of a crystal is presented. The atomic scattering factors have approximately same shapes (Eq. (2)). This can be used as an advantage. Starting from the heaviest atoms, we use product of their atomic scattering factors and sharpen the Patterson function. The peaks are sharpened in orders. This helps to locate the peaks and identify their atomic species.

We have assumed that a very large number of reflections are obtained, in fact, an infinite number of reflections. The errors due to the Fourier series truncation of the sharpened Patterson functions are negligible. As the sharpened Patterson functions are non-negative, the value of a sharpened Patterson functions is as valid as the values of the Patterson functions and other sharpened Patterson functions. Hence we can define and obtain the lowest Patterson function of a crystal. The problem of sharpening the Patterson function reduces to the problem of using all the sharpening functions such that the sharpened Patterson functions are non-negative.

Sect 2.2 presents an analytical method of sharpening the Patterson function of a crystal. We have an important conclusion: Investigation of sharpening is for all crystals. The result can then be used for any specific crystal, as needed.

Tchebycheff approximation is used as an illustration. When all the non-negative sharpened Patterson functions are used and the lowest Patterson function obtained, the ripples are wiped out (Sect. 2.1, 2.2).

Sect. 2.3 then deals with the two effects due to Fourier series truncation: large peak half-widths and large negative and positive ripples. These effects are due to truncation of the sharpened peak ${}^{s}G(\mathbf{r})$.

As shown in Sect. 2.1, 2.1, the ripples are wiped out when all the non-negative sharpened Patterson functions are used and the lowest Patterson function obtained.

Hence we need only deal with the peak half-widths. Employing iterative algorithm and superresolution by extrapolation and point atom-pairs, the peaks for all interatomic pairs may be obtained. All the peaks of the Patterson function of a crystal may be resolved. The solution may not be unique. The regions of non-zero values of the sharpened Patterson function come from the light atoms. It may be difficult to make use of these regions, for examples, plateaus. The main purpose is to quantify these regions into point atom-pairs. So long as we can divide these regions into Patterson peaks, the problem may not be severe even though the peaks are not exact.

In applying iterative algorithms in structure determination of nonperiodic or periodic objects, the most important step is to have appropriate initial phases. These may decide whether or not the iterations will converge to the correct solution. A common practice is to employ random initial phases. This is not very satisfactory. Yuen (2011b) presents an attempt of obtaining more realistic initial phases for determination of electron density of crystals. If we start from the lowest Patterson function and apply iterative algorithms, there is no initial phase problem. In applying iterative algorithms to the Patterson function, the known quantities are the observed intensities. The unknown quantities are the unobserved intensities; there are no phases to be determined. We have superresolution by extrapolation of the magnitudes. Hence the result of extrapolation will be more accurate.

Because of (2), the treatment presented is an approximation.

The peak at the origin and all known Patterson peaks may be removed (Sect. 2.4). As the structure factor expressed in (5) is an approximation, the removal of the peaks is not complete.

This article presents a general and effective method of sharpening the Patterson function so that the atomic coordinates of crystal containing large number of atoms can be determined. The effect of Fourier series truncation is also treated. The method can be applied to a large number of crystals of known structures, including macromolecular crystals, to find the effectiveness of the method and to improve the method (Sect. 2.5).

As mentioned in Sect. 1, after all the peaks of the Patterson function are resolved, we can proceed to determine the crystal atomic coordinates from these peaks. Software incorporating the sharpening method in Sect. 2.1 – 2.3 is needed.

To simplify the treatment, thermal vibrations of the atoms have not been mentioned. These could be included in the atomic scattering factors (Ladd & Palmer (1985), pp. 165-166).

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27

Dual-real-spaces iterative algorithms applied to electron density function and Patterson function of a crystal

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Abstract
This article suggests dual-real-spaces iterative algorithms, applying to the point-atom models of electron density function and Patterson function. The atomicity, point-atom and other constraints of the electron density are employed. All non-negative sharpening functions of the Patterson function of the crystal are used. The lowest Patterson function is obtained. The Patterson peaks are replaced by point-atom pairs. We have superresolution by extrapolation of the magnitudes. Crystal atomic coordinates of much larger crystal may be determined, than if only Patterson function is employed.

Keywords: Atomicity; point-atom; point-atom pair; number constraint; atomic radius constraint; interatomic distance constraint; structure constraint; non-negative sharpening functions; superresolution by extrapolation of magnitudes; lowest Patterson function.

1. Introduction

For a unit cell of a crystal containing N atoms, the number of peaks in the Patterson function is equal to $N(N-1)$. For a macromolecular crystal, the number of Patterson peaks will be larger than the number of observed Bragg intensities. Hence it may be difficult to determine these peaks from the Bragg intensities, although these observed diffractions are sufficient for the determination of atomic coordinates. Moreover, the Patterson peaks can overlap; the atomicity is lost. In applying iterative algorithms to Patterson function (Yuen (2011h), Sect. 2.3), the strong and powerful

number constraint (number of atoms in a unit cell N)
atomic radius constraint

interatomic distance constraint

structure constraint

cannot be applied. Hence it may be more effective to apply iterative algorithms to the electron density function of the crystal.

In apply iterative algorithms to the electron density function, the known quantities are the magnitudes of the observed Fourier coefficients. The unknown quantities are the phases of the observed Fourier coefficients, and the magnitudes and phases of the unobserved Fourier coefficients. These have to be determined in the iterative algorithms. In applying iterative algorithms to the Patterson function, the known quantities are the observed intensities. The unknown quantities are the unobserved intensities; there are no phases to be determined. We have superresolution by extrapolation of the magnitudes. Hence the result of extrapolation will be more accurate.

To combine the advantages of applying iterative algorithms to the electron density and to the Patterson function, this article suggest a dual-real-spaces iterative algorithms, applying to the point-atom models of electron density function and Patterson function.

2.1. Dual-real-spaces iterative algorithms

Let $$\hat{f}(\mathbf{h}) = (\sum f_n(\mathbf{h}))/(\sum Z_n) \quad (1)$$

where Z_n and $f_n(\mathbf{h})$ are the atomic number and atomic scattering factor of the n-th atom respectively.

$\hat{f}(\mathbf{h})$ is the atomic scattering factor of a hypothetical atom with $Z = 1$.

$$f_n(\mathbf{h}) \approx Z_n \hat{f}(\mathbf{h}) \quad (2)$$

Divide the structure factors $F(\mathbf{h})$ by $\hat{f}(\mathbf{h})$.

$$F(\mathbf{h}) / \hat{f}(\mathbf{h}) \approx \sum Z_n \delta(\mathbf{r}' - \mathbf{r}_n) \exp[2\pi i(\mathbf{h} \cdot \mathbf{r}_n)] \quad (3)$$

The right-hand side of (3) represents a point-atom crystal (Yuen (2011e), Sect. 2.1).

Then we apply a dual-real-spaces iterative algorithms, applying to the point-atom models of electron density function and Patterson function:

Use sharpening function $\left[\hat{f}(\mathbf{h})\right]^{-1}$.

(i) Use $I_{obs}(\mathbf{h})\left[\hat{f}(\mathbf{h})\right]^{-2}$. $I_{obs}(\mathbf{h})$ is the observed Bragg intensities (Yuen (2011h), Sect. 2.1. Then calculate the Patterson function which contains approximately point-atom pairs. Call this the first sharpened Patterson function.

(ii) Following Yuen (2011h), Sect. 2.1, 2.2. Use all non-negative sharpening functions, and obtain the lowest Patterson function with its peaks. The lowest Patterson function with its peaks will be used as a reference in the following iterations. Replace all peaks by point-atom pairs with height equal to the product of the atomic numbers in the first sharpened Patterson function (Yuen (2011h), Sect. 2.3). If the peak is due to more than one interatomic pairs, use the sum of their products of atomic numbers. Because a nearby Patterson peak may exist, we use a very small radius, for example 0.001Å for a peak. Calculate the Fourier coefficients which are non-negative. If any partial structure of the electron density can be identified, it will be used in (iii).

(iii) For the reciprocal lattice vectors $\mathbf{h}$ for which the Bragg intensities are measured, use $\left[I_{obs}(\mathbf{h})\right]^{1/2}\left[\hat{f}(\mathbf{h})\right]^{-1}$ as the magnitudes. For the other reciprocal lattice vectors $\mathbf{h}$, use the positive square root of the magnitudes of the calculated Fourier coefficients as the magnitudes.

If any partial structure of the electron density is identified in (ii), calculate Fourier coefficients of the electron density of this partial structure. Use the phases of these coefficients as initial phases. If partial structure is not obtained in (ii), use more realistic initial phases obtained in Yuen (2011c) or random initial phases.

Using these magnitudes and phases, **calculate the electron density** from the Fourier series. If any partial structure of the electron density is identified in (ii), it is included in this electron density.

(iv) Then follow Yuen (2011d), Sect. 2.2, Yuen (2011e), Sect. 2.1. Apply

number constraint (number of atoms in a unit cell N)
height constraint
atomic radius constraint and point-atom constraint
interatomic distance constraint
structure constraint,

and the seven constraints in Yuen (2011a), Sect. 2.3, or the six constraints in Yuen (2011b), Sect. 2.2. From this electron density, calculate the Fourier coefficients. For the reciprocal lattice vectors $\mathbf{h}$ for which the Bragg intensities are measured, use $I_{obs}(\mathbf{h})\left[\hat{f}(\mathbf{h})\right]^{-2}$. For the other reciprocal lattice vectors $\mathbf{h}$, use square of the magnitudes of the calculated Fourier coefficients. **Calculate the Patterson function.**

Note that oversampling is not used (Yuen (2011f, 2011g)).

(v) Compare the peaks of this Patterson function with the lowest Patterson function with its peaks in (ii). If they are consistent, add these peaks to the lowest Patterson function. If there is any contradiction, do not use these peaks, or change the corresponding point atoms in the electron density. The lowest Patterson function may be a useful reference in the iterations.

If any partial structure of the electron density is identified, add it to (iii).

Follow (ii) and replace all peaks by point-atom pairs.

(vi) Apply (iii). But use the phases of the Fourier coefficients of the electron density obtained in (iv).

Repeat the processes (iv), (v), (vi) and the iterations until convergence is reached.

If result of dual-real-spaces iterative algorithms with oversampling applied to electron density function and Patterson function of a unit cell (Yuen (2011i) is available, this result can be used to monitor the iterations in this section.

2.2 Other sharpening functions

In Sect. 2.1, in stead of the sharpening function $\left[\hat{f}(\mathbf{h})\right]^{-1}$ other sharpening functions may be used (see (Yuen (2011h), Sect. 2.1):

Use sharpening function $\left[f(\mathbf{h})\right]^{-1}$, where $f(\mathbf{h})$ is the atomic scattering factor of the heaviest atom. In (iii), use $\left[I_{obs}(\mathbf{h})\right]^{1/2}\left[f(\mathbf{h})\right]^{-1}$. In (i), use $I_{obs}(\mathbf{h})\left[f(\mathbf{h})\right]^{-2}$. The purpose of this choice of sharpening function is to locate and identify the Patterson peaks involving two heaviest atoms, or one heaviest atom.

Then use sharpening function $\left[f(\mathbf{h})\right]^{-1}$, where $f(\mathbf{h})$ is the atomic scattering factor of the second heaviest atom. In (iii), use $\left[I_{obs}(\mathbf{h})\right]^{1/2}\left[f(\mathbf{h})\right]^{-1}$. In (i), use $I_{obs}(\mathbf{h})\left[f(\mathbf{h})\right]^{-2}$. The purpose of this choice of sharpening function is to locate and identify the Patterson peaks involving two second heaviest atoms, or one second heaviest atom.

So on.

Using this slow but sure process, we hope to locate and identify all Patterson peaks.

Or use $[\sum f_n^2(\mathbf{h})]^{-1/2}$. In (iii), use $\left[I_{obs}(\mathbf{h})\right]^{1/2}[\sum f_n^2(\mathbf{h})]^{-1/2}$. In (i), use $\left[I_{obs}(\mathbf{h})\right][\sum f_n^2(\mathbf{h})]^{-1}$.

Or use. $[\sum f_n^m(\mathbf{h})]^{-1/m}$ for any positive integer m >2. In (iii), use $\left[I_{obs}(\mathbf{h})\right]^{1/2}[\sum f_n^m(\mathbf{h})]^{-1/m}$. In (i), use $\left[I_{obs}(\mathbf{h})\right][\sum f_n^m(\mathbf{h})]^{-2/m}$.

3. Discussions and conclusion

This article suggests dual-real-spaces iterative algorithms for determination of crystal atomic coordinates. The atomicity, point-atom and other constraints of the electron density are employed. All non-negative sharpening functions of the Patterson function of the crystal are used. The lowest Patterson function with its peaks is obtained. This may be a useful reference in the iterations. The Patterson peaks are replaced by point-atom pairs. We have superresolution by extrapolation of the magnitudes.

A common practice is to resolve the peaks of the Patterson function. The crystal atomic coordinates can then be determined from these peaks (Yuen (2011h), Sect. 1). In the dual-real-spaces iterative algorithms presented in this article, we have simultaneously the resolving of the Patterson peaks and the determination of atomic coordinates.

The number of atoms in a unit cell N is much smaller than the number of Patterson peaks $N(N-1)$. By means of this dual-real-spaces iterative algorithms, applying to the electron density function and Patterson function, we may be able to determine crystal atomic coordinates of much larger crystal, than if only Patterson function is employed.

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28

Sayre's equation applied to the Patterson function of equal-atom crystal. Extension of Bragg intensities

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Abstract

Sayre's equation is applied to the Patterson function of equal-atom crystal, which is used for extension of the Bragg intensities. An exact equation is obtained.

$$I(\mathbf{h}) = \frac{g(\mathbf{h})}{g^{sq}(\mathbf{h})V} \sum_{\mathbf{k}} I(\mathbf{k}) I(\mathbf{h}-\mathbf{k})$$

Keywords: Sayre equation; point-atom.

1. Introduction

Sayre (1952) presents the important Sayre's equation. David (1987) applies this equation to the determination of intensities of completely overlapping reflections in powder diffraction patterns. This article presents extension of Bragg intensity for single crystal.

In Sayre (1980), p. 277 it is mentioned that the tangent formula presents only half of the phase-determining relations in (8.7) to (8.10). The magnitude is not used and information is lost. In this article, Sayre's equation is applied to the Patterson function of equal-atom crystal, which is used for extension of Bragg intensities, or the magnitudes (Sect. 2.1, 2.2). An exact equation is obtained.

For references about Sayre's equation, see Yuen (2011e), Sect. 1.

Determination of the scattering factor of the 'squared atom' is discussed in Sayre

(1952), p. 61, Sayre (1974), p. 181, Debaerdemaeker et al. (1985), p.287, Main (1990), p. 375, Zhang & Main (1990), p. 378, Shiono & Woolfson (1991), p.527, Cowtan & Main (1993), Sect. 4.3, Zhang (1993), p. 215, and Sato (1994), p. 378

In Sect. 2.3, $g(\mathbf{h})/g^{sq}(\mathbf{h})$ is evaluated empirically.

For extension of Bragg intensities by modifying the Patterson function, see Karle & Hauptman (1964), Seeman et al. (1976) and Langs (1998).

2.1. Sayre's equation applied to the Patterson function of equal-point-atom crystal

For a crystal of equal point-atoms with height = 1 (Yuen (2011e), (1)), the square of electron density of the crystal, the squared crystal is identical to the crystal.

$$F'(\mathbf{h}) = \frac{1}{V}\sum_{\mathbf{k}} F'(\mathbf{k})F'(\mathbf{h}-\mathbf{k}) \qquad (1)$$

is the Sayre's equation for this equal-point-atom crystal (Sayre (1952), (1.3), Fan (1998), (5), Yuen (2011e), (6)). $F'(\mathbf{h})$ is the structure factor of the equal-point-atom crystal.

Now consider the Patterson function of this equal-point-atom crystal, with the large peak at the origin removed (Giacovazzo (1998), p. 307, Yuen (2011a), Sect. 2.4). All the Patterson peaks are point atom-pairs of height = 1. The square of the Patterson function is identical to the Patterson function. $\mathbf{h}$ is a reciprocal lattice vector. The Fourier coefficient of the Patterson function is the Bragg intensity $I'(\mathbf{h})$ of the crystal. Hence, from (1),

$$I'(\mathbf{h}) = \frac{1}{V}\sum_{\mathbf{k}} I'(\mathbf{k})I'(\mathbf{h}-\mathbf{k}) \qquad (2)$$

This is an exact equation.

2.2. Sayre's equation applied to the Patterson function of equal-atom crystal

Consider a crystal of real electron density. Let $p(\mathbf{r})$ be the Patterson function of an equal-atom crystal with the large peak at the origin removed.

For a crystal with N equal atoms in a unit cell, the structure factor

$$F(\mathbf{h}) = f(\mathbf{h})\sum_{n=1}^{N}\exp(2\pi i\mathbf{h}\cdot\mathbf{r}_n)\,, \tag{3}$$

$f(\mathbf{h})$ is the atomic scattering factor

There are $N(N-1)$, interatomic vector, or $N(N-1)$ Patterson peaks.

From Yuen (2011d), (10) and (11),

$$p(\mathbf{r}) = \frac{1}{V}\sum_{m=1}^{N(N-1)} G(\mathbf{r}-\mathbf{r}_m) \tag{4}$$

where $G(\mathbf{r}) = \sum_{\mathbf{h}}\left|f(\mathbf{h})\right|^2 \exp[-2\pi i(\mathbf{h}\cdot\mathbf{r})]$ (5)

$\mathbf{r}_m$ is an interatomic vector.

Let $g(\mathbf{h})$ be the scattering factor of one Patterson peak (an atom-pair).

$g(\mathbf{h})$ = the Fourier transform of $G(\mathbf{r})$ in (5). (6)

The integration is over one unit cell of the Patterson function which has the same periods as the electron density.

For a reciprocal lattice vector $\mathbf{h}$,

$$g(\mathbf{h}) = \left|f(\mathbf{h})\right|^2 \tag{7}$$

The Fourier coefficient of the Patterson function

= the Bragg intensity of crystal, $I(\mathbf{h})$

= the structure factor of the Patterson function

$$I(\mathbf{h}) = g(\mathbf{h})\sum_{m=1}^{N(N-1)}\exp(2\pi i\mathbf{h}\cdot\mathbf{r}_m) \tag{8}$$

The squared Patterson function

$$p^2(\mathbf{r}) = p(\mathbf{r})\,p(\mathbf{r}) \tag{9}$$

The structure factor of the squared Patterson function

$$P^{sq}(\mathbf{h}) = g^{sq}(\mathbf{h})\sum_{m=1}^{N(N-1)}\exp(2\pi i\mathbf{h}\cdot\mathbf{r}_m) \tag{10}$$

$g^{sq}(\mathbf{h})$ is the scattering factor of the squared Patterson peak.

From Yuen (2011e), (10),

$$P^{sq}(\mathbf{h}) = \frac{1}{V}\sum_{\mathbf{k}} I(\mathbf{k})I(\mathbf{h}-\mathbf{k}) \qquad (11)$$

From Yuen (2011e), (11),

$$I(\mathbf{h}) = [g(\mathbf{h})/g^{sq}(\mathbf{h})]P^{sq}(\mathbf{h})\,. \qquad (12)$$

Since $I(\mathbf{h})$, $g(\mathbf{h})$ and $g^{sq}(\mathbf{h})$ are non-negative, $P^{sq}(\mathbf{h})$ is non-negative. We may denote $P^{sq}(\mathbf{h})$ by $I^{sq}(\mathbf{h})$. (11) becomes

$$I^{sq}(\mathbf{h}) = \frac{1}{V}\sum_{\mathbf{k}} I(\mathbf{k})I(\mathbf{h}-\mathbf{k}) \qquad (13)$$

From (12) and (13), or Fan (1998), (5), Yuen (2011e), (12),

$$I(\mathbf{h}) = \frac{g(\mathbf{h})}{g^{sq}(\mathbf{h})V}\sum_{\mathbf{k}} I(\mathbf{k})I(\mathbf{h}-\mathbf{k}) \qquad (14)$$

This is an exact equation.

2.3. Evaluation of $g(\mathbf{h})/g^{sq}(\mathbf{h})$

There is much overlapping in the Patterson peaks. The condition of atomicity in Fan (1998), p. 79, (ii) is violated. In order to evaluate $g(\mathbf{h})/g^{sq}(\mathbf{h})$, Cowtan & Main (1993), (18) may be followed. An empirical method employing (14) is used.

$$\frac{g(h)}{g^{sq}(h)} = V\left\langle \frac{I(\mathbf{h})}{\sum_{\mathbf{k}} I(\mathbf{k})I(\mathbf{h}-\mathbf{k})} \right\rangle_h \qquad (15)$$

The average is over spherical shells each covering a narrow ranges of $|\mathbf{h}|$. $I(\mathbf{h})$ and $I(\mathbf{h}-\mathbf{k})$ are experimental values.

To obtain $g(h)/g^{sq}(h)$ for which $I(\mathbf{h})$ is not observed, a Gaussian function is used to fit to the $g(h)/g^{sq}(h)$ obtained in (15) from experimental values of $I(\mathbf{h})$ and $I(\mathbf{h}-\mathbf{k})$ (Main (1990), (15b), Zhang (1993), (6), Cowtan & Main (1993), p.151)

$$\frac{g(h)}{g^{sq}(h)} = C\exp(-Ah^2) \qquad (16)$$

The extrapolated value can then be used.

An alternative method is to choose a similar but known structure (Main (1990), p.375, Zhang & Main (1990), p.378). $g(h)/g^{sq}(h)$ can then be calculated for all values of $|\mathbf{h}|$. Compare with $g(h)/g^{sq}(h)$ obtained in (15) from experimental values of $I(\mathbf{h})$ and $I(\mathbf{h}-\mathbf{k})$, value of $g(h)/g^{sq}(h)$ for unobserved $I(\mathbf{h})$ can be obtained.

2.4. Verification with crystal structures reported in the literature.

Choose a large number of crystal structures with high resolutions, reported in the literature, of various sizes, small, mediate, or macromolecular, and with or without heavy atoms. Apply 2.2 and 2.3. Examine the verification of Eq. (14). In particular, in crystal structures with small number of atoms, the overlapping of the Patterson peaks is small, and the conditions for Sayre's equations (non-negativity, atomicity and equal-atom structure (Fan (1998), p. 79) are satisfied.

3. Discussions and conclusion

Sayre's equation is applied to the Patterson function of equal-point-atom crystal (Sect. 2.1). An exact equation (2) is obtained.

Sayre's equation is applied to the Patterson function of equal-atom crystal (Sect. 2.2). An exact equation (14) is obtained. It is used for extension of the Bragg intensities. The extended Bragg intensities obtained can then be used in various methods for determination of crystal structure. The Fourier series truncation effect is reduced. This is useful in macromolecular crystals which do not diffract to atomic resolution. Moreover, for macromolecular crystals which contain mostly light atoms, the effect of violating equal-atom condition will be small (Fan (1998), p. 79). Unlike the superresolution by extrapolation in iterative algorithms (for example, Yuen (2011b), these extended intensities are fixed during the iterations. The only quantities to be determined are the phases of the observed and unobserved structure factors. This may be an improvement.

$g(\mathbf{h})/g^{sq}(\mathbf{h})$ is evaluated empirically in Sect. 2.3. Other methods of determining the scattering factor of the squared Patterson peak $g^{sq}(\mathbf{h})$ can also be investigated. Determination of the scattering factor of the 'squared atom' in the references in Sect. 1 may be consulted. According to Cowtan & Main (1993), p.152, formulae based on analytical approximations of atomic shape have been used to evaluate $g(\mathbf{h})/g^{sq}(\mathbf{h})$. But the empirical method employed is more reliable over a range of structures. Yuen (2011c), Sect. 2.4 presents a method of evaluating $g(\mathbf{h})/g^{sq}(\mathbf{h})$.

It is useful to investigate the applicability of (14) when the three conditions (non-negativity, atomicity and equal-atom structure (Fan (1998), p. 79)) are not satisfied (Shiono & Woolfson (1991), Fan (1998), p. 81), notably the overlapping of the Patterson peaks. The result of applying (14) to a large number of crystal structures of high resolutions, reported in the literature can be examined (Sect. 2.4).

Although the extended Bragg intensities obtained may not be too accurate because of the overlapping of the Patterson peaks, but it was found in Main (1990), p. 376 that in direct determination of phases, using zeros for the unobserved reflexions, is better than left them out completely. The extended Bragg intensities obtained by applying Sayre's equation to the Patterson function of a crystal should be better than zero values.

The equation obtained, (14) can be combined with the Sayre's equation for crystal (Sayre (1952), (1.3), Fan (1998), (5), Yuen (2011e), (12)) or the tangent formula (Fan (1998), (21), Yuen (1911c), (13)) for crystal atomic-coordinates determination.

We would like to mention once more that the structure factor expression

$$F(\mathbf{h}) = f(\mathbf{h})\sum_n \exp[2\pi i(\mathbf{h}\cdot\mathbf{r}_n)]$$

for an equal-atom crystal is, strictly speaking, an approximation as atoms of same species, but in different environments, have slightly different electron density distributions.

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29

Accelerated convergence of extended Bragg intensities obtained by applying Sayre’s equation to the Patterson function of equal-atom crystal

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Abstract

Modification of the shape of Patterson peak is used to accelerate convergence of extended Bragg intensities obtained by applying Sayre’s equation to the Patterson function of equal-atom crystal. The modified Patterson function has same peak coordinates as the Patterson function. An effective method is obtained: The problem of accelerated convergence of extended Bragg intensities reduces to the problem of choosing $c(\mathbf{h})$ such that $c(\mathbf{h})/[c(\mathbf{h})\otimes c(\mathbf{h})]$ is large for the $I''(\mathbf{h})$ from the experimentally observed $\left|F(\mathbf{h})\right|^2$, and small otherwise. $c(\mathbf{h})/[c(\mathbf{h})\otimes c(\mathbf{h})]$ tends to 0 as $h\to\infty$. Choices of $c(\mathbf{h})$ are suggested. These have to be studied by numerical calculations. Further investigations are needed to have optimum choices of $c(\mathbf{h})$ for three-dimensional crystal, and that the modified Patterson peaks satisfy the conditions of non-negativity and atomicity. It is found that accelerated convergence of the extension of the Bragg intensities by means of the Sayre’s equation, is, in fact, for crystals in general. Moreover, it provides a method of evaluating $g(\mathbf{h})/g^{sq}(\mathbf{h})$. For simple equal-atom crystals, the atomic coordinates of which are determined by symmetry, the Bragg intensities with extension, the *ab initio* electron densities and the $g(\mathbf{h})/g^{sq}(\mathbf{h})$ obtained can be regarded as fundamental data in solid state physics. These will be useful in studies of solid state physics. Extension of Bragg intensities

may be a breakthrough in crystal structure analysis and solid state physics.

Keywords: Point-atom crystal; most general modified Patterson peak; most general modified Patterson function; $g(\mathbf{h})/g^{sq}(\mathbf{h})$; $c(\mathbf{h})/[c(\mathbf{h})\otimes c(\mathbf{h})]$; Gaussian atom; macromolecular crystal; fundamental data; solid state physics.

1. Introduction

In Yuen (2011d), (14), we have presented an exact equation for extension of Bragg intensities by applying Sayre's equation to the Patterson function of equal-atom crystal.

$$I(\mathbf{h}) = \frac{g(\mathbf{h})}{g^{sq}(\mathbf{h})V}\sum_{\mathbf{k}} I(\mathbf{k})I(\mathbf{h}-\mathbf{k}) \tag{1}$$

Starting with experimentally observed $I(\mathbf{h})$ and $I(\mathbf{h}-\mathbf{k})$, $I(\mathbf{h})$ for all $\mathbf{h}$ are built up.

It would be ideal if $g(\mathbf{h})/g^{sq}(\mathbf{h})$ is large for the experimentally observed $I(\mathbf{h})$, and then drops to almost zero for other values of $\mathbf{h}$. Then the calculation of the extension by means of (1) will converge fast, and an accurate collection of $I(\mathbf{h})$ for all $\mathbf{h}$ will be obtained. In general, this is not the case. As mentioned in Giacovazzo (1998), p. 307, Sayre's equation has asymptotical validity; it requires a large number of terms in the summation in (1). In order to achieve this purpose, modification of Patterson function, leading to accelerated convergence of extension of Bragg intensities, is studied in this article.

Because of atomicity, when the Patterson peak is sharpened, the peak coordinates remain unchanged. We can then use sharpening of the Patterson function to resolve the peaks. In this article, the Patterson peak is modified to other shapes, while the peak coordinates remain unchanged. This is used for accelerated convergence of extension of Bragg intensities (Sect. 2.1). Thus, whether the shape of the Patterson peak is sharpened or modified, it can be useful. An effective method for accelerated convergence is presented.

Choices of $c(\mathbf{h})$ are suggested in Sect. 2.2.

The Bragg intensities are extended one by one (Sect. 2.3).

In the accelerated convergence of extension of Bragg intensities, $g(\mathbf{h})/g^{sq}(\mathbf{h})$ is not used. It is found that accelerated convergence of the extension of the Bragg intensities by means of the Sayre's equation, is, in fact, for crystals in general. Moreover, it provides a method of evaluating $g(\mathbf{h})/g^{sq}(\mathbf{h})$ (Sect. 2.4).

For simple equal-atom crystals, the atomic coordinates of which are determined by symmetry, the Bragg intensities with extension, the *ab initio* electron densities and the $g(\mathbf{h})/g^{sq}(\mathbf{h})$ obtained can be regarded as fundamental data in solid state physics (Sect. 2.5). They will be useful in studies of solid state physics.

2.1. Accelerated convergence of extended Bragg intensities

Consider a crystal of equal atoms. The structure factor

$$F(\mathbf{h}) = f(\mathbf{h})\sum_{n}\exp[2\pi i(\mathbf{h}\cdot\mathbf{r}_n)] \tag{2}$$

The Patterson function

$$p(\mathbf{r}) = \frac{1}{V}\sum_{\mathbf{h}}\left|F(\mathbf{h})\right|^2\exp[-2\pi i(\mathbf{h}\cdot\mathbf{r})] \tag{3}$$

The Fourier coefficients in (3)

$$= \frac{I(\mathbf{h})}{V}$$

$$= \frac{\left|F(\mathbf{h})\right|^2}{V} \tag{4}$$

$I(\mathbf{h})$ is the intensity

The Patterson function

$$p(\mathbf{r}) = \frac{1}{V}\sum_{n=1}^{N}\sum_{m=1}^{N}G[\mathbf{r}-(\mathbf{r}_m-\mathbf{r}_n)] \tag{5}$$

where $G(\mathbf{r}) = \sum_{\mathbf{h}}\left|f(\mathbf{h})\right|^2\exp[-2\pi i(\mathbf{h}\cdot\mathbf{r})]$ (6)

(see Yuen (2011c), (8) and (9)). $G(\mathbf{r})$ has the form of a peak at the origin.

By applying Sayre's equation to the Patterson function (3), we obtain

$$I(\mathbf{h}) = \frac{g(\mathbf{h})}{g^{sq}(\mathbf{h})V}\sum_{\mathbf{k}} I(\mathbf{k})I(\mathbf{h}-\mathbf{k}) \qquad (1)$$

where $g(\mathbf{h})$ is the scattering factor of one Patterson peak (an atom-pair).

$g^{sq}(\mathbf{h})$ is the scattering factor of the squared Patterson peak.

(see Yuen (2011d), (14)).

From (2) and Yuen (2011e), (1),

$$F(\mathbf{h})/f(\mathbf{h}) = \sum \delta(\mathbf{r}'-\mathbf{r}_n)\exp[2\pi i(\mathbf{h}\cdot\mathbf{r}_n)] \qquad (7)$$

This represents an equal-point-atom crystal.

From (2011c), (20), the Patterson function consisting of equal point-atom-pairs, is given by

$$^{s}p(\mathbf{r}) = \frac{1}{V}\sum_{\mathbf{h}} \frac{|F(\mathbf{h})|^2}{[f(\mathbf{h})]^2}\exp[-2\pi i(\mathbf{h}\cdot\mathbf{r})] \qquad (8)$$

The Fourier coefficients in (8)

$$= \frac{I'(\mathbf{h})}{V}$$

$$= \frac{|F(\mathbf{h})|^2}{V[f(\mathbf{h})]^2} \qquad (9)$$

Corresponding to (1), we have

$$I'(\mathbf{h}) = \frac{1}{V}\sum_{\mathbf{k}} I'(\mathbf{k})I'(\mathbf{h}-\mathbf{k})\,, \qquad (10)$$

from Yuen (2011d), (2).

The most general modified Patterson peak is given by Yuen (2011c), (22)

$$^{s}G(\mathbf{r}) = \sum_{\mathbf{h}} c(\mathbf{h})\exp[-2\pi i(\mathbf{h}\cdot\mathbf{r})] \qquad (11)$$

$c(\mathbf{h})$ is the scattering factor of the modified Patterson peak $^{s}G(\mathbf{r})$.

$c(\mathbf{h})$ = Fourier transform of $^{s}G(\mathbf{r})$.

$c(\mathbf{h})$ is chosen such that $^{s}G(\mathbf{r})$ is non-negative.

For $\mathbf{h}$ a reciprocal lattice vector, $c(\mathbf{h})$ is chosen positive.

The most general modified Patterson function is given by Yuen (2011c), (23).

$$^{s}p(\mathbf{r}) = \frac{1}{V}\sum_{\mathbf{h}} \frac{|F(\mathbf{h})|^{2}}{[f(\mathbf{h})]^{2}} c(\mathbf{h}) \exp[-2\pi i(\mathbf{h}\cdot\mathbf{r})] \tag{12}$$

The Fourier coefficients in (12)

$$= \frac{I''(\mathbf{h})}{V}$$

$$= \frac{|F(\mathbf{h})|^{2}}{V[f(\mathbf{h})]^{2}} c(\mathbf{h}) \tag{13}$$

The most general modified Patterson function is given by Yuen (2011c), (15)

$$^{s}p(\mathbf{r}) = \frac{1}{V}\sum_{n=1}^{N}\sum_{m=1}^{N} {}^{s}G[\mathbf{r} - (\mathbf{r}_{m} - \mathbf{r}_{n})] \tag{14}$$

with $^{s}G(\mathbf{r})$ given by (11) (see Yuen (2011c), (15), (22)).

The squared modified-Patterson-peak

$$\left[{}^{s}G(\mathbf{r})\right]^{2} = {}^{s}G(\mathbf{r})\,{}^{s}G(\mathbf{r}) \tag{15}$$

The squared modified-Patterson-function

$$\left[{}^{s}p(\mathbf{r})\right]^{2} = {}^{s}p(\mathbf{r})\,{}^{s}p(\mathbf{r})$$

The structure factor of the squared modified-Patterson-function

$$= c^{sq}(\mathbf{h}) \sum_{m=1}^{N(N-1)} \exp(2\pi i\mathbf{h}\cdot\mathbf{r}_{m}) \tag{16}$$

$c^{sq}(\mathbf{h})$ is the scattering factor of the squared modified-Patterson-peak $\left[{}^{s}G(\mathbf{r})\right]^{2}$.

$c^{sq}(\mathbf{h})$ = Fourier transform of $\left[{}^{s}G(\mathbf{r})\right]^{2}$. (17)

Regard $\mathbf{h}$ is any vector in the reciprocal space and use the convolution theorem. From (15), $c^{sq}(\mathbf{h}) = c(\mathbf{h}) \otimes c(\mathbf{h})$ (18)

$\otimes$ denotes convolution.

From Yuen (2011d), (14), we have

$$I''(\mathbf{h}) = \frac{c(\mathbf{h})}{c^{sq}(\mathbf{h})V} \sum_{\mathbf{k}} I''(\mathbf{k}) I''(\mathbf{h}-\mathbf{k}) \tag{19}$$

The problem of accelerated convergence of extended Bragg intensities reduces to the problem of choosing $c(\mathbf{h})$ such that $c(\mathbf{h})/[c(\mathbf{h}) \otimes c(\mathbf{h})]$ is large for the $I''(\mathbf{h})$ from the experimentally observed $|F(\mathbf{h})|^2$ (see (13)), and small otherwise. $c(\mathbf{h})/[c(\mathbf{h}) \otimes c(\mathbf{h})]$ tends to 0 as $h \to \infty$. This is necessary for the convergence of the summation in (19). From (19), $I''(\mathbf{h})$ from the experimentally observed $|F(\mathbf{h})|^2$ is large, and small otherwise. There may be more than one choice of $c(\mathbf{h})$.

After the $c(\mathbf{h})$ is chosen, calculate all $I''(\mathbf{h})$ from the experimentally observed $|F(\mathbf{h})|^2$ (see (13)) and the chosen $c(\mathbf{h})$. Then use (19) to build up $I''(\mathbf{h})$ for all $\mathbf{h}$. For the $I''(\mathbf{h})$ which is not yet obtained by extension, use zero. Because of the choice of $c(\mathbf{h})$, there is accelerated convergence for evaluation of the extended $I''(\mathbf{h})$. Then use $I''(\mathbf{h})$ for all $\mathbf{h}$, and obtain $\dfrac{|F(\mathbf{h})|^2}{V[f(\mathbf{h})]^2}$ from (13). That is, we obtain the Patterson function consisting of equal point-atom-pairs ((8) and (9)). Since $f(\mathbf{h})$ is known, $I(\mathbf{h}) = |F(\mathbf{h})|^2$ for all $\mathbf{h}$ (see (4)), is found. This is the result that we are seeking. We have achieved the aim of accelerated convergence in the evaluation of the extended Bragg intensities.

2.2 Choices of $c(\mathbf{h})$

Consider an one-dimensional crystal of lattice constant a.

According to Sayre (1952), p. 61,

an Gaussian atom = $\exp[-2\pi(ar)^2]$

r is in fractional unit.

The atomic scattering factor = $[1/(\sqrt{2}a)]\exp[-\pi h^2/(2a^2)]$. (20)

The squared atom (or the Patterson peak) = $\exp[-4\pi(ar)^2]$. (21)

The scattering factor of the squared atom = $g(\mathbf{h})$ in (1)

$$= [1/(2a)]\exp[-\pi h^2/(4a^2)] \qquad (22)$$

The squared Patterson peak = $\exp[-8\pi(ar)^2]$. (23)

The scattering factor of the squared Patterson peak

= $g^{sq}(\mathbf{h})$ in (1)

$$= [1/(2\sqrt{2}a)]\exp[-\pi h^2/(8a^2)] \qquad (24)$$

$$\frac{g(\mathbf{h})}{g^{sq}(\mathbf{h})} = \sqrt{2}\exp[-\pi h^2/(8a^2)] \qquad (25)$$

It tends to 0 as $h \to \infty$. This can then be used in (1) for extending the Bragg intensities.

We intend to accelerate the convergence of extending the Bragg intensities by choosing $c(\mathbf{h})$ such that $c(\mathbf{h})/[c(\mathbf{h}) \otimes c(\mathbf{h})]$ is large for the $I''(\mathbf{h})$ from the experimentally observed $|F(\mathbf{h})|^2$, and small otherwise, tending to 0 as $h \to \infty$ (Sect. 2.1). Choices:

(i) Use (22).

$c(\mathbf{h}) = \exp[-\pi h^2/(4a^2)]$ for the $I''(\mathbf{h})$ from the experimentally observed $|F(\mathbf{h})|^2$,

or $= \eta\exp[-\pi h^2/(4a^2)]$, otherwise.

(ii) Use (24) and (22).

$c(\mathbf{h}) = \exp[-\pi h^2/(8a^2)]$ for the $I''(\mathbf{h})$ from the experimentally observed $|F(\mathbf{h})|^2$, or $= \eta \exp[-\pi h^2/(4a^2)]$, otherwise.

(iii) Use

$c(\mathbf{h}) = \exp[-\pi h^2/(na^2)]$ for the $I''(\mathbf{h})$ from the experimentally observed $|F(\mathbf{h})|^2$. n is an integer, $n \geq 9$

or $= \eta \exp[-\pi h^2/(4a^2)]$, otherwise.

η is to be chosen (for examples, 0.01, 0.001, 0.0001).

$c(\mathbf{h})$ is chosen to have sharp discontinuity. $c(\mathbf{h}) \otimes c(\mathbf{h})$ is a smooth function of $\mathbf{h}$ (Hecht (1989), Fig. 11.20, 11.23, 11.24). Hence $c(\mathbf{h})/[c(\mathbf{h}) \otimes c(\mathbf{h})]$ has the sharp discontinuity of $c(\mathbf{h})$, and is large for the $I''(\mathbf{h})$ from the experimentally observed $|F(\mathbf{h})|^2$, and small otherwise.

We expect that choices (i), (ii) or (iii) will have $c(\mathbf{h})/[c(\mathbf{h}) \otimes c(\mathbf{h})]$ tends to 0 as $h \to \infty$. This has to be verified by numerical calculations.

In case (i), (ii) and (iii) do not satisfy $c(\mathbf{h})/[c(\mathbf{h}) \otimes c(\mathbf{h})]$ tends to 0 as $h \to \infty$, we can try the other direction:

(iv) Use (20) and (22).

$c(\mathbf{h}) = \exp[-\pi h^2/(2a^2)]$ for the $I''(\mathbf{h})$ from the experimentally observed $|F(\mathbf{h})|^2$,

or $= \eta \exp[-\pi h^2/(4a^2)]$, otherwise.

(v) Use

$c(\mathbf{h}) = \exp(-\pi h^2/a^2)$ for the $I''(\mathbf{h})$ from the experimentally observed $|F(\mathbf{h})|^2$,

or $= \eta \exp[-\pi h^2/(4a^2)]$, otherwise.

As in Sayres (1952), p. 61, the Patterson peaks (21) are not entirely free of overlap. The modified Patterson peaks in the choices (i) – (v) may overlap, and have negative regions. Hence there is approximation. However, this is less important (Shiono & Woolfson (1991), Woolfson (1991), p. 44, Fan (1998), p. 81). Further investigations are needed to have optimum choices of $c(\mathbf{h})$ for three-dimensional crystal, and that the modified Patterson peaks satisfy the conditions of non-negativity and atomicity (Fan (1998), p. 79)). May use functions other than Gaussian functions.

2.3. Scheme of extension

The Bragg intensities are extended one by one:

Choose a $\mathbf{h}_1$ which is close to some $\mathbf{h}$ with strong $I(\mathbf{h})$. Use Sect. 2.1, 2.2 to extend the Bragg intensities, and obtain $I(\mathbf{h}_1)$. Then add $I(\mathbf{h}_1)$ to the list of $I(\mathbf{h})$ which is determined experimentally or by extension. Using these determined $I(\mathbf{h})$, repeat the extension, and get one more $I(\mathbf{h})$ determined by extension. Repeat the extension until all $I(\mathbf{h})$ for the intended resolution are obtained.

To refine the $I(\mathbf{h})$ determined by extension, arrange these extended $I(\mathbf{h})$ in descending order. Start from the strongest $I(\mathbf{h})$, repeat the extension of the Bragg intensities one by one.

2.4. Evaluation of $g(\mathbf{h})/g^{sq}(\mathbf{h})$

In the evaluation of $I(\mathbf{h})$ for all $\mathbf{h}$ in Sect. 2.1, we have not used (1) and $g(\mathbf{h})/g^{sq}(\mathbf{h})$. This is because we have used $F(\mathbf{h})/f(\mathbf{h})$ in (7) to transform the crystal to an equal-point-atom crystal. Here the property of the original crystal is not used. This means that **accelerated convergence of the extension of the Bragg intensities by means of the Sayre's equation, is, in fact, for crystals in general.** A similar conclusion is found in the sharpening of Patterson function (Yuen (2011c), Sect. 2.2). From the equal-point-atom crystal, we can then treat the most general modified Patterson peak in (11) and the most general modified Patterson function in (12). $c(\mathbf{h})$ chosen in Sect. 2.1 and 2.2 has no physical meaning. It is just a mathematical technique for accelerating the convergence. There can be more than one optimum choices of $c(\mathbf{h})$. Transform the crystal to equal-point-atom crystal avoids the problem of evaluating $g(\mathbf{h})/g^{sq}(\mathbf{h})$. In fact, from $I(\mathbf{h})$ for all $\mathbf{h}$, we can use (1) to obtain $g(\mathbf{h})/g^{sq}(\mathbf{h})$. This is another achievement of acceleration of the convergence of extension of Bragg intensities.

2.5. Application to simple equal-atom crystal. Fundamental data in solid state physics

Sect 2.1 and 2.2 can be applied to simple equal-atom crystals, the atomic coordinates of which are determined by symmetry, for examples, Be, Mg, Zn, C, Si, Ge (Kittel (1986), pp. 19-20). The Bragg intensities are extended. *Ab initio* electron densities can then be determined by refinement with iterative algorithms and oversampling (Yuen (2011a), Sect. 2.3, Yuen (2011b), Sect. 2.2). $g(\mathbf{h})/g^{sq}(\mathbf{h})$ can also be obtained (Sect. 2.4). The Bragg intensities with extension, the *ab initio* electron densities and the $g(\mathbf{h})/g^{sq}(\mathbf{h})$ obtained can be regarded as fundamental data in solid state physics. They will be useful in studies of solid state physics. The $g(\mathbf{h})/g^{sq}(\mathbf{h})$ obtained provides more understanding of atomic properties.

3. Discussions and conclusion

When Sayre's equation is applied to the Patterson function of equal-atom crystal, an exact equation for extension of Bragg intensities has been obtained. By means of modifying the shape of the Patterson peak, we obtain accelerated convergence of extension of Bragg intensities (Sect. 2.1). The modified Patterson function has same peak coordinates as the Patterson function. An effective method is obtained: The problem of accelerated convergence of extended Bragg intensities reduces to the problem of choosing $c(\mathbf{h})$ such that $c(\mathbf{h})/[c(\mathbf{h})\otimes c(\mathbf{h})]$ is large for the $I''(\mathbf{h})$ from the experimentally observed $|F(\mathbf{h})|^2$, and small otherwise. $c(\mathbf{h})/[c(\mathbf{h})\otimes c(\mathbf{h})]$ tends to 0 as $h\to\infty$. This is necessary for the convergence of the summation in (19). The method is applicable to macromolecular crystal.

Choices of $c(\mathbf{h})$ are suggested in Sect. 2.2. These have to be studied by numerical calculations. Further investigations are needed to have optimum choices of $c(\mathbf{h})$ for three-dimensional crystal, and that the modified Patterson peaks satisfy the conditions of non-negativity and atomicity (Fan (1998), p. 79)). Different optimum choices should lead to the same extended Bragg intensities. In fact, different optimum choices leading to the same result indicates that the result is correct.

$g(\mathbf{h})/g^{sq}(\mathbf{h})$ is not used in the accelerated convergence of extension of Bragg intensities. It is found that accelerated convergence of the extension of the Bragg intensities by means of the Sayre's equation, is, in fact, for crystals in general. Moreover, it provides a method of evaluating $g(\mathbf{h})/g^{sq}(\mathbf{h})$ (Sect. 2.4). It can be compared with the results obtained by other method of evaluating $g(\mathbf{h})/g^{sq}(\mathbf{h})$ in Yuen (2011d), Sect. 2.3.

It is useful to investigate the applicability of (19) and the chosen $c(\mathbf{h})$ when the three conditions (non-negativity, atomicity and equal-atom structure (Fan (1998), p. 79) are not satisfied (Shiono & Woolfson (1991), Fan (1998), p. 81), notably if there is overlapping of the peaks in the modified Patterson function (12). We may choose an optimum $c(\mathbf{h})$, while the non-negativity condition of the modified Patterson function is violated. It is mentioned in Woolfson (1991), p. 44 that, in terms of the normalized structure factors, 'Sayre equation holds reasonably well over a very large range of conditions'.

The higher the resolution of the experimental reflections collected, the less will be the overlapping of the peaks of the modified Patterson function. Hence the resolution of the experimental reflections collected should be as high as possible. The result of applying the accelerated convergence of extension of Bragg intensities (Sect. 2.1) to a large number of crystal structures of high resolutions, reported in the literature, can be examined.

Extension of the Bragg intensities is very useful. This is of great help in crystal structure determination. The number of Bragg intensities is increased, while the number of atomic coordinates to be determined remains the same. Extension of Bragg intensities reduces the Fourier series truncation error, producing sharper peaks and reducing the ripples. This helps to identify the atoms in the process of solving the structure, for examples, in sharpened Patterson functions (Yuen (2011c)). The atomic coordinates and electron densities obtained will be more accurate. More accurate knowledge of electron density will be helpful in understanding of chemical and biological properties of molecules.

For crystals containing a very small number of atoms, the atomic coordinates can be determined by symmetry (Sect. 2.5). If the Bragg intensities are extended, the more accurate electron density obtained will be very useful in studies of solid state physics. In this case, the three conditions of the applicability of the Sayre's equation (non-negativity, atomicity and equal-atom structure (Fan (1998), p. 79)) are satisfied. The Bragg intensities with extension, the *ab initio* electron densities and the $g(\mathbf{h})/g^{sq}(\mathbf{h})$ obtained can be regarded as fundamental data in solid state physics. The extension of the Bragg intensities presented in Yuen (2011d) and this article is based upon the physical property of the crystal (non-negativity, atomicity and equal-atom structure), and the extended intensities are obtained by means of the Patterson function and Sayre's equation. They are based on strong physical ground. Extension of Bragg intensities may be a breakthrough in crystal structure analysis and solid state physics.

In Zanotti (2011), p. 733, it is stressed that the extrapolated data obtained in the free-lunch method can be used in solving the phase problem and to obtain a model of the crystal structure. But they are not experimental data, and should be discarded after a structure model is built. But, as just mentioned, the extension of the Bragg intensities presented is based upon the physical property of the crystal, and the extended intensities are obtained by means of the Patterson function and Sayre's equation.

The accelerated convergence of extended Bragg intensities can be applied to the many crystal structures reported in the literature. Iterative algorithms with oversampling may then be applied to refine the electron densities (Yuen (2011a), Sect. 2.3, Yuen (2011b), Sect. 2.2).

Extension of Bragg intensities may be regarded as the first step in crystal structure analysis. The Bragg intensities are first extended, and then various methods can be applied to solve the structure. For articles to be published in the future, the extended Bragg intensities may be included.

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30

Dual-real-spaces iterative algorithms applied to electron density function and Patterson function of a crystal, plus extension of Bragg intensities from application of Sayre's equation to the Patterson function

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By applying Sayre's equation to the Patterson function of a crystal, the Bragg intensities can be extended (Yuen (2011b). The convergence of the extension can be accelerated (Yuen (2011c)), and all extended intensities are obtained. This means that there is no Fourier series truncation error. These extended intensities can then be used in dual-real-spaces iterative algorithms applied to electron density function and Patterson function of a crystal (Yuen (2011a)). Unlike the superresolution by extrapolation employed in Yuen (2011a), these extended intensities are fixed during the iterations. The only quantities to be determined are the phases of the observed and unobserved structure factors. This may be an improvement.

It will be useful to incorporate the dual-real-spaces iterative algorithms plus extension of Bragg intensities with other methods of X-ray crystallography, such as direct methods (Ladd & Palmer (1980), Schenk (1991), Fortier (1998), Giacovazzo (1998)), isomorphous replacement (Rossmann & Arnold (2001)), single-wavelength anomalous diffraction (SAD) and multiwavelength anomalous diffraction (MAD).

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31

Accelerated convergence of extension of structure factors by means of Sayre's equation for equal-atom crystal

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Abstract

Modification of the shape of the atoms is used to accelerate convergence of extended structure factors obtained by applying Sayre's equation to an equal-atom crystal. The modified electron density has same atomic coordinates as the original crystal. An effective method is obtained: The problem of accelerated convergence of extended structure factors reduces to the problem of choosing $c(\mathbf{h})$ such that $c(\mathbf{h})/[c(\mathbf{h})\otimes c(\mathbf{h})]$ is large for the modified structure factors $F''(\mathbf{h})$ from the known $F(\mathbf{h})$, and small otherwise. $c(\mathbf{h})/[c(\mathbf{h})\otimes c(\mathbf{h})]$ tends to 0 as $h\to\infty$. The method is applicable to macromolecular crystal. Choices of $c(\mathbf{h})$ are suggested. These have to be studied by numerical calculations. Further investigations are needed to have optimum choices of $c(\mathbf{h})$ for three-dimensional crystal, and that the modified atoms satisfy the conditions of non-negativity and atomicity. It is found that accelerated convergence of the extension of the structure factors by means of the Sayre's equation, is, in fact, for crystals in general. Moreover, it provides a method of evaluating $f(\mathbf{h})/f^{sq}(\mathbf{h})$.

Keywords: Point-atom crystal; most general modified atom; most general modified electron density; $f(\mathbf{h})/f^{sq}(\mathbf{h})$; $c(\mathbf{h})/[c(\mathbf{h})\otimes c(\mathbf{h})]$; Gaussian atom; macromolecular crystal.

1. Introduction

Sayre (1952) has presented a very useful and important exact equation for extension of structure factors for equal-atom crystal (Fan (1998), (5), Yuen (2011d), (12)).

$$F(\mathbf{h}) = \frac{f(\mathbf{h})}{f^{sq}(\mathbf{h})V} \sum_{\mathbf{k}} F(\mathbf{k})F(\mathbf{h}-\mathbf{k}) \tag{1}$$

Starting with known $F(\mathbf{h})$ and $F(\mathbf{h}-\mathbf{k})$, $F(\mathbf{h})$ for all $\mathbf{h}$ are built up.

Sayre (1952), p. 64 mentions that for very sharp atoms and very short X-ray wavelength, a very large number of structure factors are obtained. Cochran (1952), Sect. 3 deals with partial series. Zachariasen (1952), p. 70 states that by applying a large structure factor, it is possible to use a small number of structure factors in (1). In *SAYTAN i*n Woolfson & Fan (1995), Sect. 3.5.5, there is an overall scaling constant K for the fact that only structure factors with large magnitudes are included in the right-hand side of (1). All these publications concern the number of terms in the right-hand side of (1) and their effects to the determination of phases. Accelerated convergence of extension of structure factors by means of Sayre’s equation will be useful in these problems.

It would be ideal if $f(\mathbf{h})/f^{sq}(\mathbf{h})$ is large for the known $F(\mathbf{h})$, and then drops to almost zero for other values of $\mathbf{h}$. Then the calculation of the extension by means of (1) will converge fast, and an accurate collection of $F(\mathbf{h})$ for all $\mathbf{h}$ will be obtained. In general, this is not the case. In order to achieve this purpose, modification of the electron density, leading to accelerated convergence of extension of structure factors, is studied in this article. The development follows that of Yuen (2011c).

Because of atomicity, when the Patterson peak is sharpened, the peak coordinates remain unchanged. We can then use sharpening of the Patterson function to resolve the peaks. In this article, the atoms are modified to other shape, while the atomic coordinates remain unchanged. This is used for accelerated convergence of extension of structure factors (Sect. 2.1). Thus, whether the shape of the Patterson peak is sharpened or the shape of the atom is modified, it can be useful (Sayre (1952), p. 64, Zachariasen (1952), p. 70). An effective method for accelerated convergence is presented. The method is applicable to macromolecular crystal.

Choices of $c(\mathbf{h})$ are suggested in Sect. 2.2.

The structure factors are extended one by one (Sect. 2.3).

In the accelerated convergence of extension of structure factors, $f(\mathbf{h})/f^{sq}(\mathbf{h})$ is not used. It is found that accelerated convergence of the extension of the structure factors by means of the Sayre's equation, is, in fact, for crystals in general. Moreover, it provides a method of evaluating $f(\mathbf{h})/f^{sq}(\mathbf{h})$ (Sect. 2.4).

Extension of structure factors may be applicable to low-resolution phasing. Low-resolution phasing is presented in Gilmore (2000), Gomis-Rüth & Coll (2001), Lunin et al. (2007) and Liu et al. (2011).

2.1. Accelerated convergence of extended structure factors

Consider a real or complex crystal of equal atoms. The structure factor

$$F(\mathbf{h}) = f(\mathbf{h})\sum_{n}\exp[2\pi i(\mathbf{h}\cdot\mathbf{r}_n)] \tag{2}$$

The electron density

$$\rho(\mathbf{r}) = \frac{1}{V}\sum_{\mathbf{h}} F(\mathbf{h})\exp[-2\pi i(\mathbf{h}\cdot\mathbf{r})] \tag{3}$$

The Fourier coefficients in (3)

$$= \frac{F(\mathbf{h})}{V} \tag{4}$$

Sayre's equation (Fan (1998), (5), Yuen (2011d), (12)) is

$$F(\mathbf{h}) = \frac{f(\mathbf{h})}{f^{sq}(\mathbf{h})V}\sum_{\mathbf{k}} F(\mathbf{k})F(\mathbf{h}-\mathbf{k}) \tag{1}$$

where $f^{sq}(\mathbf{h})$ is the scattering factor of the squared atom.

From (2) and Yuen (2011d), (1),

$$F'(\mathbf{h}) = F(\mathbf{h})/f(\mathbf{h}) = \sum\delta(\mathbf{r}'-\mathbf{r}_n)\exp[2\pi i(\mathbf{h}\cdot\mathbf{r}_n)] \tag{5}$$

This represents an equal-point-atom crystal.

From (3) and (5), the electron density of this equal-point-atom crystal

$$\rho'(\mathbf{r}) = \frac{1}{V}\sum_{\mathbf{h}} F'(\mathbf{h})\exp[-2\pi i(\mathbf{h}\cdot\mathbf{r})]$$

$$= \frac{1}{V}\sum_{\mathbf{h}} \frac{F(\mathbf{h})}{f(\mathbf{h})}\exp[-2\pi i(\mathbf{h}\cdot\mathbf{r})] \tag{6}$$

The Fourier coefficients in (6)

$$= \frac{F'(\mathbf{h})}{V}$$

$$= \frac{F(\mathbf{h})}{Vf(\mathbf{h})} \tag{7}$$

Corresponding to (1), we have

$$F'(\mathbf{h}) = \frac{1}{V}\sum_{\mathbf{k}} F'(\mathbf{k})F'(\mathbf{h}-\mathbf{k}) \tag{8}$$

from Yuen (2011d), (6).

From (5), the most general modified structure factor is given by

$$F''(\mathbf{h}) = \frac{F(\mathbf{h})}{f(\mathbf{h})}c(\mathbf{h}) = c(\mathbf{h})\sum \delta(\mathbf{r}'-\mathbf{r}_n)\exp[2\pi i(\mathbf{h}\cdot\mathbf{r}_n)] \tag{9}$$

Note that the atomic coordinates are the same as those of the crystal (see (2)).

$c(\mathbf{h})$ is the scattering factor (that is, the Fourier transform) of the modified atom $\xi(\mathbf{r})$. $\xi(\mathbf{r})$ is the most general modified atom. $c(\mathbf{h})$ and $\xi(\mathbf{r})$ have moderate shapes.

$c(\mathbf{h})$ is chosen such that the modified atom $\xi(\mathbf{r})$ is non-negative and confined to a radius of about 0.65Å, and, for Gaussian atom, $c(\mathbf{h})$ has a radius of 1.3Å^{-1} (Sayre (1952), p. 61).

For $\mathbf{h}$ a reciprocal lattice vector, $c(\mathbf{h})$ is chosen positive.

The modified electron density of the crystal of modified atoms,

$$\rho''(\mathbf{r}) = \frac{1}{V}\sum_{\mathbf{h}} F''(\mathbf{h})\exp[-2\pi i(\mathbf{h}\cdot\mathbf{r})]$$

$$= \frac{1}{V}\sum_{\mathbf{h}} \frac{F(\mathbf{h})}{f(\mathbf{h})}c(\mathbf{h})\exp[-2\pi i(\mathbf{h}\cdot\mathbf{r})] \tag{10}$$

$\rho''(\mathbf{r})$ is the most general modified electron density.

The Fourier coefficients in (10)

$$= \frac{F''(\mathbf{h})}{V} \tag{11}$$

$$= \frac{F(\mathbf{h})}{Vf(\mathbf{h})} c(\mathbf{h}) \tag{12}$$

The squared modified-atom

$$[\xi(\mathbf{r})]^2 = \xi(\mathbf{r})\xi(\mathbf{r}) \tag{13}$$

The electron density of the squared modified-crystal

$$[\rho''(\mathbf{r})]^2 = \rho''(\mathbf{r})\rho''(\mathbf{r}) \tag{14}$$

The structure factor of the squared modified-crystal

$$= c^{sq}(\mathbf{h}) \sum_n \exp(2\pi i \mathbf{h} \cdot \mathbf{r}_n) \tag{15}$$

$c^{sq}(\mathbf{h})$ is the scattering factor of the squared modified-atom $[\xi(\mathbf{r})]^2$.

$$c^{sq}(\mathbf{h}) = \text{Fourier transform of } [\xi(\mathbf{r})]^2 . \tag{16}$$

Regard $\mathbf{h}$ is any vector in the reciprocal space and use the convolution theorem. From (13), $c^{sq}(\mathbf{h}) = c(\mathbf{h}) \otimes c(\mathbf{h})$ (17)

$\otimes$ denotes convolution.

From Yuen (2011d), (12), we have

$$F''(\mathbf{h}) = \frac{c(\mathbf{h})}{c^{sq}(\mathbf{h})V} \sum_{\mathbf{k}} F''(\mathbf{k})F''(\mathbf{h}-\mathbf{k}) \tag{18}$$

The problem of accelerated convergence of extended structure factors reduces to the problem of choosing $c(\mathbf{h})$ such that $c(\mathbf{h})/[c(\mathbf{h}) \otimes c(\mathbf{h})]$ is large for the modified structure factors $F''(\mathbf{h})$ from the known $F(\mathbf{h})$ (magnitude and phase) (see (12)), and small otherwise. $c(\mathbf{h})/[c(\mathbf{h}) \otimes c(\mathbf{h})]$ tends to 0 as $h \to \infty$. This is necessary for the convergence of the summation in (18). There may be more than one choice.

After the $c(\mathbf{h})$ is chosen, calculate all $F''(\mathbf{h})$ from the known $F(\mathbf{h})$ (see (12)) and the chosen $c(\mathbf{h})$. Then use (18) to build up $F''(\mathbf{h})$ for all $\mathbf{h}$. For the $F''(\mathbf{h})$ which is not yet obtained by extension, use zero. Because of the choice of $c(\mathbf{h})$, there is accelerated convergence for evaluation of the extended $F''(\mathbf{h})$. Then use $F''(\mathbf{h})$ for all $\mathbf{h}$, and obtain $\dfrac{F(\mathbf{h})}{Vf(\mathbf{h})}$ from (12). That is, we obtain the electron density consisting of equal point atoms ((6) and (7)). Since $f(\mathbf{h})$ is known, $F(\mathbf{h})$ for all $\mathbf{h}$, is found. This is the result that we are seeking. We have achieved the aim of accelerated convergence in the evaluation of the extended structure factors. The method is applicable to macromolecular crystal.

2.2 Choices of $c(\mathbf{h})$

See Yuen (2011c) Sect. 2.2. Consider an one-dimensional crystal of lattice constant a. According to Sayre (1952), p. 61,

an Gaussian atom $= \exp[-2\pi(ar)^2]$ (19)

r is in fractional unit.

The atomic scattering factor = $f(\mathbf{h})$ in (1)

$$= [1/(\sqrt{2}a)]\exp[-\pi h^2/(2a^2)] \qquad (20)$$

The squared atom $= \exp[-4\pi(ar)^2]$. (21)

The scattering factor of the squared atom = $f^{sq}(\mathbf{h})$ in (1)

$$= [1/(2a)]\exp[-\pi h^2/(4a^2)] \qquad (22)$$

$$\frac{f(\mathbf{h})}{f^{sq}(\mathbf{h})} = \sqrt{2}\exp[-\pi h^2/(4a^2)] \qquad (23)$$

It tends to 0 as $h \to \infty$. This can then be used in (1) for extending the structure factors.

We intend to accelerate the convergence of extending the structure factors by choosing $c(\mathbf{h})$ such that $c(\mathbf{h})/[c(\mathbf{h}) \otimes c(\mathbf{h})]$ is large for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$, and small otherwise. $c(\mathbf{h})/[c(\mathbf{h}) \otimes c(\mathbf{h})]$ tends to 0 as $h \to \infty$ (Sect. 2.1). Choices:

(i) Use (20).

$c(\mathbf{h}) = \exp[-\pi h^2/(2a^2)]$ for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$,

or $= \eta \exp[-\pi h^2/(2a^2)]$, otherwise.

(ii) Use (22) and (20).

$c(\mathbf{h}) = \exp[-\pi h^2/(4a^2)]$ for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$,

or $= \eta \exp[-\pi h^2/(2a^2)]$, otherwise.

(iii) Use

$c(\mathbf{h}) = \exp[-\pi h^2/(na^2)]$ for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$,

n is an integer, $n \geq 5$

or $= \eta \exp[-\pi h^2/(2a^2)]$, otherwise.

η is to be chosen (for examples, 0.01, 0.001, 0.0001).

$c(\mathbf{h})$ is chosen to have sharp discontinuity. $c(\mathbf{h}) \otimes c(\mathbf{h})$ is a smooth function of $\mathbf{h}$ (Hecht (1989), Fig. 11.20, 11.23, 11.24). Hence $c(\mathbf{h})/[c(\mathbf{h}) \otimes c(\mathbf{h})]$ has the sharp discontinuity of $c(\mathbf{h})$, and is large for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$, and small otherwise.

We expect that choices (i), (ii) or (iii) will have $c(\mathbf{h})/[c(\mathbf{h}) \otimes c(\mathbf{h})]$ tends to 0 as $h \to \infty$. This has to be verified by numerical calculations.

In case (i), (ii) and (iii) do not satisfy $c(\mathbf{h})/[c(\mathbf{h}) \otimes c(\mathbf{h})]$ tends to 0 as $h \to \infty$, we can try the other direction:

(iv) Use

$c(\mathbf{h}) = \exp(-\pi h^2/a^2)$ for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$,

or $= \eta \exp[-\pi h^2/(2a^2)]$, otherwise.

As in Sayres (1952), p. 61, the atoms (19) are not entirely free of overlap. The modified atoms in the choices (i) – (iv) may overlap, and have negative regions. Hence there is approximation. However, this is less important (Shiono & Woolfson (1991), Woolfson (1991), p. 44, Fan (1998), p. 81). Further investigations are needed to have optimum choices of $c(\mathbf{h})$ for three-dimensional crystal, and that the modified atoms satisfy the conditions of non-negativity and atomicity (Fan (1998), p. 79)). May use functions other than Gaussian functions.

2.3. Scheme of extension

The scheme of extension in Yuen (2011c), Sect. 2.3 is used; the structure factors are extended one by one until all structure factors for the intended resolution are obtained.

To refine the structure factors determined by extension, arrange the extended structure factors in descending order according to their magnitudes. Start from the largest magnitude, repeat the extension of the structure factors one by one.

2.4. Evaluation of $f(\mathbf{h})/f^{sq}(\mathbf{h})$

In the evaluation of $F(\mathbf{h})$ for all $\mathbf{h}$ in Sect. 2.1, we have not used (1) and $f(\mathbf{h})/f^{sq}(\mathbf{h})$. This is because we have used $F(\mathbf{h})/f(\mathbf{h})$ in (5) to transform the crystal to an equal-point-atom crystal. Here the property of the original crystal is not used, for example, the original crystal may be real or complex. This means that **accelerated convergence of the extension of the structure factors by means of the Sayre's equation is, in fact, for crystals in general.** A similar conclusion is found in the sharpening of Patterson function (Yuen (2011a), Sect. 2.2) and extension of Bragg intensities (Yuen (2011b), Sect. 2.4). From the equal-point-atom crystal, we can then treat the most general modified atom $\xi(\mathbf{r})$ (from (9)) and the most general modified electron density in (10). $c(\mathbf{h})$ chosen in Sect. 2.1 and 2.2 has no physical meaning. It is just a mathematical technique for accelerating the convergence. There can be more than one optimum choices of $c(\mathbf{h})$. Transform the crystal to equal-point-atom crystal avoids the problem of evaluating $f(\mathbf{h})/f^{sq}(\mathbf{h})$. In fact, from $F(\mathbf{h})$ for all $\mathbf{h}$, we can use (1) to obtain $f(\mathbf{h})/f^{sq}(\mathbf{h})$. This is another achievement of acceleration of the convergence of extension of structure factors.

3. Discussions and conclusion

By means of modifying the shape of the atoms, we obtain accelerated convergence of extension of structure factors (Sect. 2.1). The modified electron density has same atomic coordinates as the original crystal. An effective method is obtained: The problem of accelerated convergence of extended structure factors reduces to the problem of choosing $c(\mathbf{h})$ such that $c(\mathbf{h})/[c(\mathbf{h})\otimes c(\mathbf{h})]$ is large for the modified structure factors $F''(\mathbf{h})$ from the known $F(\mathbf{h})$, and small otherwise. $c(\mathbf{h})/[c(\mathbf{h})\otimes c(\mathbf{h})]$ tends to 0 as $h\rightarrow\infty$. This is necessary for the convergence of the summation in (18). The method is applicable to macromolecular crystal. Extension of structure factors may be applicable to low-resolution phasing.

Choices of $c(\mathbf{h})$ are suggested in Sect. 2.2. These have to be studied by numerical calculations. Further investigations are needed to have optimum choices of $c(\mathbf{h})$ for three-dimensional crystal, and that the modified atoms satisfy the conditions of non-negativity and atomicity (Fan (1998), p. 79)). Different optimum choices should lead to the same extended structure factors. In fact, different optimum choices leading to the same result indicates that the result is correct.

$f(\mathbf{h})/f^{sq}(\mathbf{h})$ is not used in the accelerated convergence of extension of structure factors. It is found that accelerated convergence of the extension of the structure factors by means of the Sayre's equation, is, in fact, for crystals in general. Moreover, it provides a method of evaluating $f(\mathbf{h})/f^{sq}(\mathbf{h})$ (Sect. 2.4). It can be compared with the results obtained by other method of determining the scattering factor of the 'squared atom' in the references in Yuen (2011b), Sect. 1.

As accelerated convergence of the extension of the structure factors by means of the Sayre's equation, is, in fact, for crystals in general, the modified structure factors may be regarded as more 'fundamental'. Direct methods may be applied to the structure factors or modified structure factors.

It is useful to investigate the applicability of (18) and the chosen $c(\mathbf{h})$ when the three conditions (non-negativity, atomicity and equal-atom structure) are not satisfied by the modified electron density (Shiono & Woolfson (1991), Fan (1998), p. 79, 81). It is mentioned in Woolfson (1991), p. 44 that, in terms of the normalized structure factors, 'Sayre equation holds reasonably well over a very large range of conditions'.

If the overlapping of the Patterson peaks is not a problem, the magnitudes of the structure factors are first extended with acceleration by applying Sayre's equation to the Patterson function of the crystal (Yuen (2011b, 2011c). Then we need only obtain the phases of the extended structure factors.

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32

A solution of the phase problem of one-dimensional centrosymmetric equal-atom crystal by means of accelerated convergence of Sayre’s equation

August 25, 2001

Abstract

A deterministic and routine method is provided for the solution of the phase problem of a one-dimensional centrosymmetric equal-atom crystal. Accelerated convergence of Sayre’s equation is employed.

Keywords: Deterministic method; centrosymmetric; macromolecular crystal.

1. Introduction

Sayre (1952) presents the very useful and important Sayre’s equation. As Sayre’s equation has asymptotical validity, it requires a large number of terms in the summation (Giacovazzo (1998), p. 307). This has experimental limitation. To overcome this difficultly and to reduce the amount of computation, accelerated convergence is introduced (Yuen (2011).

In this article, a deterministic and routine method is provided for the solution of the phase problem of a one-dimensional centrosymmetric equal-atom crystal. The experimental $\left|F(\mathbf{h})\right|$ are divided into two groups. Group A for calculating the structure, Group B for discriminating the signs of the structure factors in Group A. The method can be applied to the phase problem for three-dimensional crystal.

2. Method

From Yuen (2011), (18),

$$F''(\mathbf{h}) = \frac{c(\mathbf{h})}{c^{sq}(\mathbf{h})V} \sum_{\mathbf{k}} F''(\mathbf{k}) F''(\mathbf{h}-\mathbf{k}) \qquad (1)$$

$$c^{sq}(\mathbf{h}) = c(\mathbf{h}) \otimes c(\mathbf{h})$$

$\mathbf{h}$ are all the reciprocal lattice vectors, whether the structure factors are related by symmetry or not. For centrosymmtric crystal, the structure factors of a centrosymmetric pair may be used.

$c(\mathbf{h})$ is chosen such that the modified atom $\xi(\mathbf{r})$ (Yuen (2011), Sect. 2.1) is non-negative and confined to a radius of about 0.65Å, and, for Gaussian atom, $c(\mathbf{h})$ has a radius of 1.3Å^{-1} (Sayre (1952), p. 61).

For $\mathbf{h}$ a reciprocal lattice vector, $c(\mathbf{h})$ is chosen positive.

From Yuen (2011), (9), the modified structure factor

$$F''(\mathbf{h}) = \frac{F(\mathbf{h})}{f(\mathbf{h})} c(\mathbf{h}) \qquad (2)$$

$c(\mathbf{h})$ is chosen such that $c(\mathbf{h})/[c(\mathbf{h}) \otimes c(\mathbf{h})]$ is large for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$ (magnitude and phase) (see (2)), and small otherwise. $c(\mathbf{h})/[c(\mathbf{h}) \otimes c(\mathbf{h})]$ tends to 0 as $h \to \infty$. From (1), $F''(\mathbf{h})$ from the known $F(\mathbf{h})$ are large, and small otherwise.

Use the one-dimensional example of Sayre (1952), Sect. 2. Calculate the structure factors from $h = 1$ to 40. Regard their magnitudes as experimental data. Use (2) to convert $F(\mathbf{h})$ to the modified structure factors $F''(\mathbf{h})$. Divide the structure factors (and modified structure factors) into two groups. Group A is for $h = 1$ to 26. Group A is for calculating the structure. Group B is for $h = 27$ to 40. Group B is for discrimination.

For each modified structure factor in Group A, there are two choices of sign. For the 26 modified structure factors in Group A, there are $2^{26} = 6.71 \times 10^7$ combinations of signs. If for each combination, we calculate the crystal structure, much computation is needed. Instead, accelerated convergence of extension of structure factors will be used. Choose the strongest reflection, say $|F''(\mathbf{h}_1)|$, in Group B. For each combination of the signs in Group A, calculate $F''(\mathbf{h}_1)$ from (1). The summation in the right-hand side is limited to modified structure factors in Group A. If the calculated $\left|F''(\mathbf{h}_1)\right|$ agrees with the experimental $\left|F''(\mathbf{h}_1)\right|$, this combination of signs in Group A will be regarded as acceptable. Find all the acceptable combinations for $\left|F''(\mathbf{h}_1)\right|$. Choose the next strongest reflection $\left|F''(\mathbf{h}_2)\right|$ in Group B. Out of all the acceptable combinations for $\left|F''(\mathbf{h}_1)\right|$, find the combinations such that the calculated $\left|F''(\mathbf{h}_2)\right|$ agrees with the experimental $\left|F''(\mathbf{h}_2)\right|$. Repeat the process until a unique combination is obtained. Calculate the crystal structure. See whether it is the correct crystal structure. If the structure is correct, use all the structure factors in Group A and Group B to obtain more accurate value.

3. Discussions and conclusion

A deterministic and routine method is provided for the solution of the phase problem of a one-dimensional centrosymmetric equal-atom crystal. Accelerated convergence of extension of structure factors by means of Sayre's equation is employed.

Investigation of applying the method to three-dimensional crystal is needed, in particular, macromolecular crystal. It is useful to incorporate with other methods in crystallography. Direct methods may be employed to obtain the signs of some reflections, so that amount of computation can be reduced.

It is useful to investigate the applicability of (1) when the non-negativity and atomicity conditions are not satisfied by the modified electron density (Fan (1998), p. 79, 81). It is mentioned in Woolfson (1991), p. 44 that, in terms of the normalized structure factors, 'Sayre equation holds reasonably well over a very large range of conditions'. Simplifications such as reducing the radius of the sphere of reflections, or, only strongest 25% and weakest 25% reflections are included in the summation in (1), can be explored. By intuition, we may think that reflections in the middle range come from random distributions of the atoms. The strongest and weakest reflections come from very specific distributions. They are sensitive to variations of atomic coordinates, and are effective in determination of atomic coordinate. The weakest reflections are as important as the strongest reflections (see also Fan (1998), p. 85, Tate (1991), p. 426).

'Sharpening' of $c(\mathbf{h})$ in the reciprocal space may be applied. The radius of the sphere of reflections is reduced. Hence smaller number of reflections in Group A is used. $c(\mathbf{h})$ is the scattering factor (that is, the Fourier transform) of the modified atom $\xi(\mathbf{r})$ ((Yuen (2011), Sect. 2.1). $\xi(\mathbf{r})$ is broadened. There is overlapping of the modified atoms. We may use functions other than the Gaussian functions, so that $\xi(\mathbf{r})$ has the size of atomic radius while $c(\mathbf{h})$ is very sharp. For example, the Fourier transform of the rectangular function is $\sin c(s) = \sin(\pi s)/(\pi s)$. For an atom of the form of a rectangular function with radius 0.65Å, the first zero of $\sin c(s)$ occurs at 0.769 Å^{-1}. Hence the number of reflections in Group A will be much reduced (Compare with the 1.3Å^{-1} for Gaussian atom (Yuen (2011), Sect. 2.1). Investigation is needed to have optimum choice.

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33

Accelerated convergence of extension of structure factors by means of modifications of Sayre's equation for crystal of unequal atoms

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Abstract

Modification of the shape of the atoms is used to accelerate convergence of extended structure factors obtained by applying modifications of Sayre's equation to crystal containing two, three, or more than three types of atoms. An effective method is obtained: For crystal containing three types of atoms, the problem of accelerated convergence of extended structure factors reduces to the problem of choosing $c(\mathbf{h})$ such that $A''_{PQR}(\mathbf{h})$, $B''_{PQR}(\mathbf{h})$ and $C''_{PQR}(\mathbf{h})$ are large for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$, and small otherwise. $A''_{PQR}(\mathbf{h})$, $B''_{PQR}(\mathbf{h})$ and $C''_{PQR}(\mathbf{h})$ tend to 0 as $h \to \infty$. The method is applicable to macromolecular crystal. Choices of $c(\mathbf{h})$ are suggested. These have to be studied by numerical calculations. Further investigations are needed to have optimum choices of $c(\mathbf{h})$ for three-dimensional crystal, and that the modified atoms satisfy the conditions of non-negativity and atomicity. It is found that accelerated convergence of the extension of the structure factors by means of modifications of the Sayre's equation, is, in fact, for crystals in general. Moreover, it provides relations between $A_{PQ}(\mathbf{h})$ and $B_{PQ}(\mathbf{h})$, and between $A_{PQR}(\mathbf{h})$, $B_{PQR}(\mathbf{h})$ and $C_{PQR}(\mathbf{h})$. These are useful in evaluation of

$A_{PQ}(\mathbf{h})$ and $B_{PQ}(\mathbf{h})$, $A_{PQR}(\mathbf{h})$, $B_{PQR}(\mathbf{h})$ and $C_{PQR}(\mathbf{h})$.

Keywords: Point-atom crystal; most general modified atom; most general modified electron density; Gaussian atom; macromolecular crystal.

1. Introduction

This is a sequel article of Yuen (2011c) which deals with equal-atom crystal. Woolfson (1958a) has presented two very useful and important exact equations for extension of structure factors for crystal containing two or more than two types of atoms, respectively:

$$F(\mathbf{h}) = A(\mathbf{h})V^{-1}\sum_{\mathbf{k}} F(\mathbf{k})F(\mathbf{h}-\mathbf{k}) + B(\mathbf{h})V^{-2}\sum_{\mathbf{k}}\sum_{\mathbf{l}} F(\mathbf{k})F(\mathbf{l})F(\mathbf{h}-\mathbf{k}-\mathbf{l})$$

$$F(\mathbf{h}) = A(\mathbf{h})V^{-1}\sum_{\mathbf{k}} F(\mathbf{k})F(\mathbf{h}-\mathbf{k}) + B(\mathbf{h})V^{-2}\sum_{\mathbf{k}}\sum_{\mathbf{l}} F(\mathbf{k})F(\mathbf{l})F(\mathbf{h}-\mathbf{k}-\mathbf{l})$$

$$+ C(\mathbf{h})V^{-3}\sum_{\mathbf{k}}\sum_{\mathbf{l}}\sum_{\mathbf{m}} F(\mathbf{k})F(\mathbf{l})F(\mathbf{m})F(\mathbf{h}-\mathbf{k}-\mathbf{l}-\mathbf{m}) + \ldots$$

These are modifications of Sayre's equation (Sayre (1952)). Starting with known structure factors, $F(\mathbf{h})$ for all $\mathbf{h}$ are built up.

This article deals with accelerated convergence of extension of structure factors by means of modifications of Sayre's equation for crystal of unequal atoms. Accelerated convergence is achieved by modifying the electron density. The development follows those of Yuen (2011b) and (2011c).

Accelerated convergence of extension of structure factors for crystal of two types of atoms is presented in Sect. 2.1.

Choices of $c(\mathbf{h})$ are suggested in Sect. 2.2.

The structure factors are extended one by one (Sect. 2.3).

In the accelerated convergence of extension of structure factors, $A_{PQ}(\mathbf{h})$ and $B_{PQ}(\mathbf{h})$ are not used. It is found that accelerated convergence of the extension of the structure factors by means of the modified Sayre's equation, is, in fact, for crystals in general. Moreover, it provides a relation between $A_{PQ}(\mathbf{h})$ and $B_{PQ}(\mathbf{h})$, which can be used in the evaluation of $A_{PQ}(\mathbf{h})$ and $B_{PQ}(\mathbf{h})$ (Sect. 2.4).

Treatments of crystal containing three types of atoms are presented in Sect. 2.5 to 2.8.

For crystal containing more than three types of atoms, see Sect. 2.9.

For studies of a crystal containing more than one species of atoms, see Woolfson (1958b), Rothbauer (1976, 1977, 1978, 1980), Roach et al. (2001) and Roach & Carter (2002, 2003).

Extension of structure factors may be applicable to low-resolution phasing. Low-resolution phasing is presented in Guo et al. (2000), Lunina et al. (2003), Müller et al. (2006) and Abrescia et al. (2011).

2.1. Accelerated convergence of extended structure factors of crystal containing two types of atoms

Consider a real or complex, centrosymmetric or non-centrosymmetric crystal of two types of atoms, P and Q. The structure factor

$$F(\mathbf{h}) = \sum_{n} f_n(\mathbf{h}) \exp[2\pi i(\mathbf{h} \cdot \mathbf{r}_n)] \tag{1}$$

The electron density

$$\rho(\mathbf{r}) = \frac{1}{V} \sum_{\mathbf{h}} F(\mathbf{h}) \exp[-2\pi i(\mathbf{h} \cdot \mathbf{r})] \tag{2}$$

The Fourier coefficients in (2)

$$= \frac{F(\mathbf{h})}{V} \tag{3}$$

Use the modified Sayre equation (Woolfson (1958a), (10), Shiono & Woolfson (1991), (12)),

$F(\mathbf{h})$

$$= A_{PQ}(\mathbf{h})V^{-1}\sum_{\mathbf{k}} F(\mathbf{k})F(\mathbf{h}-\mathbf{k}) + B_{PQ}(\mathbf{h})V^{-2}\sum_{\mathbf{k}}\sum_{\mathbf{l}} F(\mathbf{k})F(\mathbf{l})F(\mathbf{h}-\mathbf{k}-\mathbf{l}) \tag{4}$$

$A_{PQ}(\mathbf{h})$ and $B_{PQ}(\mathbf{h})$ can be determined from the scattering factors of the electron densities and their squares and cubes, of the two types of atoms (Woolfson (1958a), Sect. 2, 3).

Let Z_n and $f_n(\mathbf{h})$ be the atomic number and atomic scattering factor of the n-th atom respectively. The atomic scattering factors have approximately same shapes. Let

$$\hat{f}(\mathbf{h}) = (\sum f_n(\mathbf{h}))/(\sum Z_n) \tag{5}$$

$\hat{f}(\mathbf{h})$ is the atomic scattering factor of a hypothetical atom with $Z = 1$.

$$f_n(\mathbf{h}) \approx Z_n \hat{f}(\mathbf{h}) \tag{6}$$

$$F'(\mathbf{h}) = \frac{F(\mathbf{h})}{\hat{f}(\mathbf{h})} = \sum_n Z_n \exp[2\pi i(\mathbf{h}\cdot\mathbf{r}_n)] \tag{7}$$

This represents a point-atom crystal.

From (2) and (7), the electron density of this point-atom crystal

$$\begin{aligned}\rho'(\mathbf{r}) &= \frac{1}{V}\sum_{\mathbf{h}} F'(\mathbf{h})\exp[-2\pi i(\mathbf{h}\cdot\mathbf{r})] \\ &= \frac{1}{V}\sum_{\mathbf{h}} \frac{F(\mathbf{h})}{\hat{f}(\mathbf{h})}\exp[-2\pi i(\mathbf{h}\cdot\mathbf{r})] \end{aligned} \tag{8}$$

The Fourier coefficients in (8)

$$= \frac{F'(\mathbf{h})}{V} \tag{9}$$

$$= \frac{F(\mathbf{h})}{V\hat{f}(\mathbf{h})} \tag{10}$$

Corresponding to (4), we have

$$F'(\mathbf{h})$$

$$= A'_{PQ}(\mathbf{h})V^{-1}\sum_{\mathbf{k}} F'(\mathbf{k})F'(\mathbf{h}-\mathbf{k}) + B'_{PQ}(\mathbf{h})V^{-2}\sum_{\mathbf{k}}\sum_{\mathbf{l}} F'(\mathbf{k})F'(\mathbf{l})F'(\mathbf{h}-\mathbf{k}-\mathbf{l}) \tag{11}$$

Let Z_P and Z_Q be the atomic numbers of atoms of types P and Q respectively.

From (6), Woolfson (1958a), (5) and (6),

$$A'_{PQ}(\mathbf{h})Z_P{}^2 + B'_{PQ}(\mathbf{h})Z_P{}^3 = Z_P \tag{12}$$

$$A'_{PQ}(\mathbf{h})Z_Q{}^2 + B'_{PQ}(\mathbf{h})Z_Q{}^3 = Z_Q \tag{13}$$

From (12) and (13), $A'_{PQ}(\mathbf{h})$ and $B'_{PQ}(\mathbf{h})$ are determined. For point-atom crystal, $A'_{PQ}(\mathbf{h})$ and $B'_{PQ}(\mathbf{h})$ are constants, independent of $\mathbf{h}$.

From (7), the most general modified structure factor is given by

$$F''(\mathbf{h}) = \frac{F(\mathbf{h})}{\hat{f}(\mathbf{h})}c(\mathbf{h}) = c(\mathbf{h})\sum_n Z_n \exp[2\pi i(\mathbf{h}\cdot\mathbf{r}_n)] \tag{14}$$

Note that the atomic coordinates are the same as those of the crystal (see (1)).
$c(\mathbf{h})$ is the scattering factor (that is, the Fourier transform) of the modified atom $\xi(\mathbf{r})$. $\xi(\mathbf{r})$ is the most general modified atom. $c(\mathbf{h})$ and $\xi(\mathbf{r})$ have moderate shapes.
$c(\mathbf{h})$ is chosen such that the modified atom $\xi(\mathbf{r})$ is non-negative and confined to a radius of about 0.65Å, and, for Gaussian atom, $c(\mathbf{h})$ has a radius of 1.3Å^{-1} (Sayre (1952), p. 61).
For $\mathbf{h}$ a reciprocal lattice vector, $c(\mathbf{h})$ is chosen positive.
The modified electron density of the crystal of modified atoms,

$$\rho''(\mathbf{r}) = \frac{1}{V}\sum_{\mathbf{h}} F''(\mathbf{h})\exp[-2\pi i(\mathbf{h}\cdot\mathbf{r})]$$

(In-between Fourier Coefficients Not Employed)

$$= \frac{1}{V}\sum_{\mathbf{h}} \frac{F(\mathbf{h})}{\hat{f}(\mathbf{h})} c(\mathbf{h}) \exp[-2\pi i(\mathbf{h}\cdot\mathbf{r})] \tag{15}$$

$\rho''(\mathbf{r})$ is the most general modified electron density.

The Fourier coefficients in (15)

$$= \frac{F''(\mathbf{h})}{V} \tag{16}$$

$$= \frac{F(\mathbf{h})}{V\hat{f}(\mathbf{h})} c(\mathbf{h}) \tag{17}$$

Corresponding to (4), we have

$$F''(\mathbf{h}) = A''_{PQ}(\mathbf{h})V^{-1}\sum_{\mathbf{k}} F''(\mathbf{k})F''(\mathbf{h}-\mathbf{k})$$

$$+ B''_{PQ}(\mathbf{h})V^{-2}\sum_{\mathbf{k}}\sum_{\mathbf{l}} F''(\mathbf{k})F''(\mathbf{l})F''(\mathbf{h}-\mathbf{k}-\mathbf{l}) \tag{18}$$

	P	Q	
The modified atom	$\xi(\mathbf{r})Z_P$	$\xi(\mathbf{r})Z_Q$	(19)
Square of the modified atom	$[\xi(\mathbf{r})]^2 Z_P{}^2$	$[\xi(\mathbf{r})]^2 Z_Q{}^2$	
Cube of the modified atom	$[\xi(\mathbf{r})]^3 Z_P{}^3$	$[\xi(\mathbf{r})]^3 Z_Q{}^3$	

The scattering factor of the modified atoms and their squares and cubes

P	Q	
$c(\mathbf{h})Z_P$	$c(\mathbf{h})Z_Q$	(20)
$c(\mathbf{h})\otimes c(\mathbf{h})Z_P{}^2$.	$c(\mathbf{h})\otimes c(\mathbf{h})Z_Q{}^2$	(21)
$c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})Z_P{}^3$	$c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})Z_Q{}^3$	(22)

$\mathbf{h}$ is any vector in the reciprocal space.

From (20), (21), (22), Woolfson (1958a), (5) and (6),

$$A''_{PQ}(\mathbf{h})c(\mathbf{h})\otimes c(\mathbf{h})Z_P{}^2 + B''_{PQ}(\mathbf{h})c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})Z_P{}^3 = c(\mathbf{h})Z_P \tag{23}$$

$$A''_{PQ}(\mathbf{h})c(\mathbf{h})\otimes c(\mathbf{h})Z_Q{}^2 + B''_{PQ}(\mathbf{h})c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})Z_Q{}^3 = c(\mathbf{h})Z_Q \tag{24}$$

From (23) and (24), $A''_{PQ}(\mathbf{h})$ and $B''_{PQ}(\mathbf{h})$ are determined.

Starting with known structure factors, $F''(\mathbf{h})$ for all $\mathbf{h}$ are built up by means of (18). **The problem of accelerated convergence of extended structure factors reduces to the problem of choosing $c(\mathbf{h})$ such that $A''_{PQ}(\mathbf{h})$ and $B''_{PQ}(\mathbf{h})$ are large for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$ (magnitude and phase) (see (17)), and small otherwise. $A''_{PQ}(\mathbf{h})$ and $B''_{PQ}(\mathbf{h})$ tend to 0 as $h \to \infty$.** This is necessary for the convergence of the summation in (18). There may be more than one choice.

After the $c(\mathbf{h})$ is chosen, with $A''_{PQ}(\mathbf{h})$ and $B''_{PQ}(\mathbf{h})$ determined, calculate all $F''(\mathbf{h})$ from the known $F(\mathbf{h})$ (see (17)) and the chosen $c(\mathbf{h})$. Then use (18) to build up $F''(\mathbf{h})$ for all $\mathbf{h}$. For the $F''(\mathbf{h})$ which is not yet obtained by extension, use zero. Because of the choice of $c(\mathbf{h})$, there is accelerated convergence for evaluation of the extended $F''(\mathbf{h})$. Then use $F''(\mathbf{h})$ for all $\mathbf{h}$, and obtain $\dfrac{F(\mathbf{h})}{V\hat{f}(\mathbf{h})}$ from (17). That is, we obtain the electron density consisting of point atoms ((8) and (10)). Since $\hat{f}(\mathbf{h})$ is known, $F(\mathbf{h})$ for all $\mathbf{h}$, is found. This is the result that we are seeking. We have achieved the aim of accelerated convergence in the evaluation of the extended structure factors. The method is applicable to macromolecular crystal.

2.2. Choices of $c(\mathbf{h})$

See Yuen (2011c), Sect. 2.2. According to Woolfson (1958a), Sect. 3, consider a one-dimensional crystal of Gaussian atoms of types P and Q.

The electron density distributions:

	P	Q	
ρ	$\exp[-2\pi(ar)^2]$	$2\exp[-2\pi(ar)^2]$	(25)
ρ^2	$\exp[-4\pi(ar)^2]$	$4\exp[-4\pi(ar)^2]$	
ρ^3	$\exp[-6\pi(ar)^2]$	$8\exp[-6\pi(ar)^2]$	

r is in fractional unit.

The scattering factor of the atoms ρ, ρ^2 and ρ^3:

P	Q	
$\frac{0.7071}{a}\exp[-\pi h^2/(2a^2)]$	$\frac{1.414}{a}\exp[-\pi h^2/(2a^2)]$	(26)
$\frac{0.5}{a}\exp[-\pi h^2/(4a^2)]$	$\frac{2}{a}\exp[-\pi h^2/(4a^2)]$	(27)
$\frac{0.4082}{a}\exp[-\pi h^2/(6a^2)]$	$\frac{3.266}{a}\exp[-\pi h^2/(6a^2)]$	(28)

From (26), (27), (28), Woolfson (1958a), (5) and (6),

$$\frac{0.5}{a}\exp\left[-\frac{\pi h^2}{4a^2}\right]A_{PQ}(\mathbf{h})+\frac{0.4082}{a}\exp\left[-\frac{\pi h^2}{6a^2}\right]B_{PQ}(\mathbf{h})$$

$$=\frac{0.7071}{a}\exp\left[-\frac{\pi h^2}{2a^2}\right]$$

$$\frac{2}{a}\exp\left[-\frac{\pi h^2}{4a^2}\right]A_{PQ}(\mathbf{h})+\frac{3.266}{a}\exp\left[-\frac{\pi h^2}{6a^2}\right]B_{PQ}(\mathbf{h})=\frac{1.414}{a}\exp\left[-\frac{\pi h^2}{2a^2}\right]$$

From these two equations, $A_{PQ}(\mathbf{h})$ and $B_{PQ}(\mathbf{h})$ are found to be

$$A_{PQ}(\mathbf{h}) = 2.121\exp[-0.25\pi h^2 / a^2]$$

$$B_{PQ}(\mathbf{h}) = -0.8659\exp[-0.3333\pi h^2 / a^2]$$

$A_{PQ}(\mathbf{h})$ and $B_{PQ}(\mathbf{h})$ tend to 0 as $h \to \infty$. This can then be used in (4) for extending the structure factors.

We intend to accelerate the convergence of extending the structure factors by choosing $c(\mathbf{h})$ such that $A''_{PQ}(\mathbf{h})$ and $B''_{PQ}(\mathbf{h})$ are large for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$, and small otherwise. $A''_{PQ}(\mathbf{h})$ and $B''_{PQ}(\mathbf{h})$ tend to 0 as $h \to \infty$ (Sect. 2.1). Choices:

(i) Use (26).

$c(\mathbf{h}) = \exp[-\pi h^2 /(2a^2)]$ for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$,
or $= \eta\exp[-\pi h^2 /(2a^2)]$, otherwise.

(ii) Use (27) and (26).

$c(\mathbf{h}) = \exp[-\pi h^2 /(4a^2)]$ for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$,
or $= \eta\exp[-\pi h^2 /(2a^2)]$, otherwise.

(iii) Use

$c(\mathbf{h}) = \exp[-\pi h^2 /(na^2)]$ for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$,
n is an integer, $n \ge 5$. For example, (28).
or $= \eta\exp[-\pi h^2 /(2a^2)]$, otherwise.

η is to be chosen (for examples, 0.01, 0.001, 0.0001).

$c(\mathbf{h})$ is chosen to have sharp discontinuity. $c(\mathbf{h}) \otimes c(\mathbf{h})$ and $c(\mathbf{h}) \otimes c(\mathbf{h}) \otimes c(\mathbf{h})$ are smooth functions of $\mathbf{h}$ (Hecht (1989), Fig. 11.20, 11.23, 11.24). Hence $A''_{PQ}(\mathbf{h})$ and $B''_{PQ}(\mathbf{h})$ determined in (23) and (24) have the sharp discontinuity of $c(\mathbf{h})$, and are large for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$, and small otherwise.

It needs to verify by numerical calculations that choices (i), (ii) or (iii) will have $A''_{PQ}(\mathbf{h})$ and $B''_{PQ}(\mathbf{h})$ tend to 0 as $h \to \infty$. In case that this is not true, we can try the other direction:

(iv) Use

$c(\mathbf{h}) = \exp(-\pi h^2 / a^2)$ for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$,

or $= \eta \exp[-\pi h^2 /(2a^2)]$, otherwise.

As in Sayres (1952), p. 61, Woolfson (1958a), p. 279, the atoms (25) are not entirely free of overlap. The modified atoms in the choices (i) – (iv) may overlap, and have negative regions. Hence there is approximation. However, this is less important (Shiono & Woolfson (1991), Woolfson (1991), p. 44, Fan (1998), p. 81). Further investigations are needed to have optimum choices of $c(\mathbf{h})$ for three-dimensional crystal, and that the modified atoms satisfy the conditions of non-negativity and atomicity (Fan (1998), p. 79)). May use functions other than Gaussian functions.

2.3. Scheme of extension

The scheme of extension in Yuen (2011b), Sect. 2.3 is used; the structure factors are extended one by one until all structure factors for the intended resolution are obtained.

To refine the structure factors determined by extension, arrange these extended structure factors in descending order according to their magnitudes. Start from the largest magnitude, repeat the extension of the structure factors one by one.

2.4. Evaluation of $A_{PQ}(\mathbf{h})$ and $B_{PQ}(\mathbf{h})$

In the evaluation of $F(\mathbf{h})$ for all $\mathbf{h}$ in Sect. 2.1, we have not used (4) and $A_{PQ}(\mathbf{h})$ and $B_{PQ}(\mathbf{h})$. This is because we have used $F(\mathbf{h})/\hat{f}(\mathbf{h})$ in (7) to transform the crystal to a point-atom crystal. Here the property of the original crystal is not used, for example, the original crystal may be real or complex. This means that **accelerated convergence of the extension of the structure factors by means of the modified Sayre equation is, in fact, for crystals in general.** A similar conclusion is found in the sharpening of Patterson function (Yuen (2011a), Sect. 2.2) and extension of Bragg intensities (Yuen (2011b), Sect. 2.4). From the point-atom crystal, we can then treat the most general modified atom $\xi(\mathbf{r})$ (from (14)) and the most general modified electron density in (15). $c(\mathbf{h})$ chosen in Sect. 2.1 and 2.2 has no physical meaning. It is just a mathematical technique for accelerating the convergence. There can be more than one optimum choices of $c(\mathbf{h})$. Transform the crystal to point-atom crystal avoids the problem of evaluating $A_{PQ}(\mathbf{h})$ and $B_{PQ}(\mathbf{h})$. In fact, from $F(\mathbf{h})$ for all $\mathbf{h}$, we can use (4) to obtain a relation between $A_{PQ}(\mathbf{h})$ and $B_{PQ}(\mathbf{h})$, which can be used in the evaluation of $A_{PQ}(\mathbf{h})$ and $B_{PQ}(\mathbf{h})$. This is another achievement of acceleration of the convergence of extension of structure factors.

2.5. Accelerated convergence of extended structure factors of crystal containing three types of atoms

Follow Sect. 2.1. Consider a real or complex, centrosymmetric or non-centrosymmetric crystal of three types of atoms P, Q and R. The structure factor

$$F(\mathbf{h}) = \sum_n f_n(\mathbf{h}) \exp[2\pi i(\mathbf{h}\cdot\mathbf{r}_n)] \tag{29}$$

The electron density

$$\rho(\mathbf{r}) = \frac{1}{V}\sum_{\mathbf{h}} F(\mathbf{h}) \exp[-2\pi i(\mathbf{h}\cdot\mathbf{r})]$$

The Fourier coefficients in $\rho(\mathbf{r})$

$$= \frac{F(\mathbf{h})}{V}$$

Use Woolfson (1958a), (23) for extension of the structure factors,

$$F(\mathbf{h}) = A_{PQR}(\mathbf{h})V^{-1}\sum_{\mathbf{k}} F(\mathbf{k})F(\mathbf{h}-\mathbf{k})$$

$$+ B_{PQR}(\mathbf{h})V^{-2}\sum_{\mathbf{k}}\sum_{\mathbf{l}} F(\mathbf{k})F(\mathbf{l})F(\mathbf{h}-\mathbf{k}-\mathbf{l})$$

$$+ C_{PQR}(\mathbf{h})V^{-3}\sum_{\mathbf{k}}\sum_{\mathbf{l}}\sum_{\mathbf{m}} F(\mathbf{k})F(\mathbf{l})F(\mathbf{m})F(\mathbf{h}-\mathbf{k}-\mathbf{l}-\mathbf{m}) \tag{30}$$

$A_{PQR}(\mathbf{h})$, $B_{PQR}(\mathbf{h})$ and $C_{PQR}(\mathbf{h})$ can be determined from the scattering factors of the electron densities and their squares, cubes and quadruples, of the three types of atoms (Woolfson (1958a), Sect. 2, 3).

Note that $A_{PQR}(\mathbf{h})$ and $B_{PQR}(\mathbf{h})$ are different from $A_{PQ}(\mathbf{h})$ and $B_{PQ}(\mathbf{h})$ for two types of atoms (Sect. 2.1).

$$F'(\mathbf{h}) = \frac{F(\mathbf{h})}{\hat{f}(\mathbf{h})} = \sum_{n} Z_n \exp[2\pi i(\mathbf{h}\cdot\mathbf{r}_n)] \tag{31}$$

represents a point-atom crystal.

The electron density of this point-atom crystal

$$\rho'(\mathbf{r}) = \frac{1}{V}\sum_{\mathbf{h}} F'(\mathbf{h})\exp[-2\pi i(\mathbf{h}\cdot\mathbf{r})]$$

$$= \frac{1}{V}\sum_{\mathbf{h}} \frac{F(\mathbf{h})}{\hat{f}(\mathbf{h})}\exp[-2\pi i(\mathbf{h}\cdot\mathbf{r})] \tag{32}$$

The Fourier coefficients in (32)

$$= \frac{F'(\mathbf{h})}{V} \tag{33}$$

$$= \frac{F(\mathbf{h})}{V\hat{f}(\mathbf{h})} \tag{34}$$

Corresponding to (30), we have

$$F'(\mathbf{h}) = A'_{PQR}(\mathbf{h})V^{-1}\sum_{\mathbf{k}} F'(\mathbf{k})F'(\mathbf{h}-\mathbf{k})$$

$$+B'_{PQR}(\mathbf{h})V^{-2}\sum_{\mathbf{k}}\sum_{\mathbf{l}}F'(\mathbf{k})F'(\mathbf{l})F'(\mathbf{h}-\mathbf{k}-\mathbf{l})$$

$$+C'_{PQ!R}(\mathbf{h})V^{-3}\sum_{\mathbf{k}}\sum_{\mathbf{l}}\sum_{\mathbf{m}}F'(\mathbf{k})F'(\mathbf{l})F'(\mathbf{m})F'(\mathbf{h}-\mathbf{k}-\mathbf{l}-\mathbf{m})$$

(35)

Let Z_R the atomic numbers of atoms of type R. From Woolfson (1958a), (5) and (6),

$$A'_{PQR}(\mathbf{h})Z_P{}^2+B'_{PQR}(\mathbf{h})Z_P{}^3+C'_{PQR}(\mathbf{h})Z_P{}^4=Z_P \tag{36}$$

$$A'_{PQR}(\mathbf{h})Z_Q{}^2+B'_{PQR}(\mathbf{h})Z_Q{}^3+C'_{PQR}(\mathbf{h})Z_Q{}^4=Z_Q \tag{37}$$

$$A'_{PQR}(\mathbf{h})Z_R{}^2+B'_{PQR}(\mathbf{h})Z_R{}^3+C'_{PQR}(\mathbf{h})Z_R{}^4=Z_R \tag{38}$$

From (36), (37) and (38), $A'_{PQR}(\mathbf{h})$, $B'_{PQR}(\mathbf{h})$ and $C'_{PQR}(\mathbf{h})$ are determined. For point-atom crystal, $A'_{PQR}(\mathbf{h})$, $B'_{PQR}(\mathbf{h})$ and $C'_{PQR}(\mathbf{h})$ are constants, independent of $\mathbf{h}$.

From (31), the most general modified structure factor is given by

$$F''(\mathbf{h})=\frac{F(\mathbf{h})}{\hat{f}(\mathbf{h})}c(\mathbf{h})=c(\mathbf{h})\sum_n Z_n\exp[2\pi i(\mathbf{h}\cdot\mathbf{r}_n)] \tag{39}$$

Note that the atomic coordinates are the same as those of the crystal (see (29)).

$c(\mathbf{h})$ is the scattering factor (that is, the Fourier transform) of the modified atom $\xi(\mathbf{r})$. $\xi(\mathbf{r})$ is the most general modified atom. $c(\mathbf{h})$ and $\xi(\mathbf{r})$ have moderate shapes.

$c(\mathbf{h})$ is chosen such that the modified atom $\xi(\mathbf{r})$ is non-negative and confined to a radius of about 0.65Å, and, for Gaussian atom, $c(\mathbf{h})$ has a radius of 1.3Å^{-1} (Sayre (1952), p. 61).

For $\mathbf{h}$ a reciprocal lattice vector, $c(\mathbf{h})$ is chosen positive.

The modified electron density of the crystal of modified atoms,

$$\rho''(\mathbf{r}) = \frac{1}{V}\sum_{\mathbf{h}} F''(\mathbf{h}) \exp[-2\pi i(\mathbf{h}\cdot\mathbf{r})]$$

$$= \frac{1}{V}\sum_{\mathbf{h}} \frac{F(\mathbf{h})}{\hat{f}(\mathbf{h})} c(\mathbf{h}) \exp[-2\pi i(\mathbf{h}\cdot\mathbf{r})] \tag{40}$$

$\rho''(\mathbf{r})$ is the most general modified electron density.

The Fourier coefficients in (40)

$$= \frac{F''(\mathbf{h})}{V} \tag{41}$$

$$= \frac{F(\mathbf{h})}{V\hat{f}(\mathbf{h})} c(\mathbf{h}) \tag{42}$$

Corresponding to (30), we have

$$\begin{aligned} F''(\mathbf{h}) = {} & A''_{PQR}(\mathbf{h}) V^{-1} \sum_{\mathbf{k}} F''(\mathbf{k}) F''(\mathbf{h}-\mathbf{k}) \\ & + B''_{PQR}(\mathbf{h}) V^{-2} \sum_{\mathbf{k}}\sum_{\mathbf{l}} F''(\mathbf{k}) F''(\mathbf{l}) F''(\mathbf{h}-\mathbf{k}-\mathbf{l}) \\ & + C''_{PQR}(\mathbf{h}) V^{-3} \sum_{\mathbf{k}}\sum_{\mathbf{l}}\sum_{\mathbf{m}} F''(\mathbf{k}) F''(\mathbf{l}) F''(\mathbf{m}) F''(\mathbf{h}-\mathbf{k}-\mathbf{l}-\mathbf{m}) \end{aligned} \tag{43}$$

	P	Q	R
The modified atom	$\xi(\mathbf{r})Z_P$	$\xi(\mathbf{r})Z_Q$	$\xi(\mathbf{r})Z_R$
Square of the modified atom	$[\xi(\mathbf{r})]^2 Z_P{}^2$	$[\xi(\mathbf{r})]^2 Z_Q{}^2$	$[\xi(\mathbf{r})]^2 Z_R{}^2$
Cube of the modified atom	$[\xi(\mathbf{r})]^3 Z_P{}^3$	$[\xi(\mathbf{r})]^3 Z_Q{}^3$	$[\xi(\mathbf{r})]^3 Z_R{}^3$
Quadruple of the modified atom	$[\xi(\mathbf{r})]^4 Z_P{}^4$	$[\xi(\mathbf{r})]^4 Z_Q{}^4$	$[\xi(\mathbf{r})]^4 Z_R{}^4$

(44)

The scattering factors of the modified atoms and their squares and cubes

$$\begin{array}{ccc}
P & Q & R \\
c(\mathbf{h})Z_P & c(\mathbf{h})Z_Q & c(\mathbf{h})Z_R \\
c(\mathbf{h})\otimes c(\mathbf{h})Z_P{}^2\,. & c(\mathbf{h})\otimes c(\mathbf{h})Z_Q{}^2 & c(\mathbf{h})\otimes c(\mathbf{h})Z_R{}^2 \\
c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})Z_P{}^3 & c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})Z_Q{}^3 & \\
 & & c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})Z_R{}^3
\end{array}$$

$$c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})Z_P{}^4$$

$$c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})Z_Q{}^4$$

$$c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})Z_R{}^4$$

$\mathbf{h}$ is any vector in the reciprocal space.

(45)

From these scattering factors, and Woolfson (1958a), (5) and (6),

$$A''_{PQR}(\mathbf{h})c(\mathbf{h})\otimes c(\mathbf{h})Z_P{}^2 + B''_{PQR}(\mathbf{h})c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})Z_P{}^3$$

$$+ C''_{PQR}(\mathbf{h})c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})Z_P{}^4 = c(\mathbf{h})Z_P$$

(46)

$$A''_{PQR}(\mathbf{h})c(\mathbf{h})\otimes c(\mathbf{h})Z_Q{}^2 + B''_{PQR}(\mathbf{h})c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})Z_Q{}^3$$

$$+C''_{PQR}(\mathbf{h})c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})Z_Q{}^4 = c(\mathbf{h})Z_Q \tag{47}$$

$$A''_{PQR}(\mathbf{h})c(\mathbf{h})\otimes c(\mathbf{h})Z_R{}^2 + B''_{PQR}(\mathbf{h})c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})Z_R{}^3$$

$$+C''_{PQR}(\mathbf{h})c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})Z_R{}^4 = c(\mathbf{h})Z_R \tag{48}$$

From (46), (47) and (48), $A''_{PQR}(\mathbf{h})$, $B''_{PQR}(\mathbf{h})$ and $C''_{PQR}(\mathbf{h})$ are determined.

Starting with known structure factors, $F''(\mathbf{h})$ for all $\mathbf{h}$ are built up by means of (43). **The problem of accelerated convergence of extended structure factors reduces to the problem of choosing $c(\mathbf{h})$ such that $A''_{PQR}(\mathbf{h})$, $B''_{PQR}(\mathbf{h})$ and $C''_{PQR}(\mathbf{h})$ are large for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$ (magnitude and phase) (see (42)), and small otherwise. $A''_{PQR}(\mathbf{h})$, $B''_{PQR}(\mathbf{h})$ and $C''_{PQR}(\mathbf{h})$ tend to 0 as $h \to \infty$.** This is necessary for the convergence of the summation in (43). There may be more than one choice.

After the $c(\mathbf{h})$ is chosen, with $A''_{PQR}(\mathbf{h})$, $B''_{PQR}(\mathbf{h})$ and $C''_{PQR}(\mathbf{h})$ determined, calculate all $F''(\mathbf{h})$ from the known $F(\mathbf{h})$ (see (42)) and the chosen $c(\mathbf{h})$. Then use (43) to build up $F''(\mathbf{h})$ for all $\mathbf{h}$. For the $F''(\mathbf{h})$ which is not yet obtained by extension, use zero. Because of the choice of $c(\mathbf{h})$, there is accelerated convergence for evaluation of the extended $F''(\mathbf{h})$. Then use $F''(\mathbf{h})$ for all $\mathbf{h}$, and obtain $\dfrac{F(\mathbf{h})}{V\hat{f}(\mathbf{h})}$ from (42). That is, we obtain the electron density consisting of point atoms ((32) and (34)). Since $\hat{f}(\mathbf{h})$ is known, $F(\mathbf{h})$ for all $\mathbf{h}$, is found. This is the result that we are seeking. We have achieved the aim of accelerated convergence in the evaluation of the extended structure factors. The method is applicable to macromolecular crystal.

2.6. Choices of $c(\mathbf{h})$

See Yuen (2011c), Sect. 2.2. According to Woolfson (1958a), Sect. 3, consider a one-dimensional crystal of Gaussian atoms of types P, Q and R.

The electron density distributions:

$$\begin{array}{llll} & P & Q & R \\ \rho & \exp[-2\pi(ar)^2] & 2\exp[-2\pi(ar)^2] & 4\exp[-2\pi(ar)^2] \\ \rho^2 & \exp[-4\pi(ar)^2] & 4\exp[-4\pi(ar)^2] & 16\exp[-4\pi(ar)^2] \\ \rho^3 & \exp[-6\pi(ar)^2] & 8\exp[-6\pi(ar)^2] & 64\exp[-6\pi(ar)^2] \\ \rho^4 & \exp[-8\pi(ar)^2] & 16\exp[-8\pi(ar)^2] & 256\exp[-8\pi(ar)^2] \end{array} \tag{49}$$

r is in fractional unit.

The scattering factor of the atoms ρ, ρ^2, ρ^3 and ρ^4:

$$\begin{array}{lll} P & Q & R \\ \frac{0.7071}{a}\exp[-\pi h^2/(2a^2)] & \frac{1.414}{a}\exp[-\pi h^2/(2a^2)] & \\ & & \frac{2.828}{a}\exp[-\pi h^2/(2a^2)] \end{array} \tag{50}$$

$$\begin{array}{lll} \frac{0.5}{a}\exp[-\pi h^2/(4a^2)] & \frac{2}{a}\exp[-\pi h^2/(4a^2)] & \\ & & \frac{8}{a}\exp[-\pi h^2/(4a^2)] \end{array} \tag{51}$$

$$\frac{0.4082}{a}\exp[-\pi h^2/(6a^2)] \qquad \frac{3.266}{a}\exp[-\pi h^2/(6a^2)]$$

$$\frac{26.12}{a}\exp[-\pi h^2/(6a^2)]$$

(52)

$$\frac{0.3536}{a}\exp[-\pi h^2/(8a^2)] \qquad \frac{5.657}{a}\exp[-\pi h^2/(8a^2)]$$

$$\frac{90.51}{a}\exp[-\pi h^2/(8a^2)]$$

(53)

From (50) – (53), Woolfson (1958a), (5) and (6),

$$\frac{0.5}{a}\exp\left[-\frac{\pi h^2}{4a^2}\right]A_{PQR}(\mathbf{h})+\frac{0.4082}{a}\exp\left[-\frac{\pi h^2}{6a^2}\right]B_{PQR}(\mathbf{h})$$

$$+\frac{0.3536}{a}\exp\left[-\frac{\pi h^2}{8a^2}\right]C_{PQR}(\mathbf{h})=\frac{0.7071}{a}\exp\left[-\frac{\pi h^2}{2a^2}\right]$$

$$\frac{2}{a}\exp\left[-\frac{\pi h^2}{4a^2}\right]A_{PQR}(\mathbf{h})+\frac{3.266}{a}\exp\left[-\frac{\pi h^2}{6a^2}\right]B_{PQR}(\mathbf{h})$$

$$+\frac{5.657}{a}\exp\left[-\frac{\pi h^2}{8a^2}\right]C_{PQR}(\mathbf{h})=\frac{1.414}{a}\exp\left[-\frac{\pi h^2}{2a^2}\right]$$

$$\frac{8}{a}\exp\left[-\frac{\pi h^2}{4a^2}\right]A_{PQR}(\mathbf{h})+\frac{26.12}{a}\exp\left[-\frac{\pi h^2}{6a^2}\right]B_{PQR}(\mathbf{h})$$

$$+\frac{90.51}{a}\exp\left[-\frac{\pi h^2}{8a^2}\right]C_{PQR}(\mathbf{h})=\frac{2.828}{a}\exp\left[-\frac{\pi h^2}{2a^2}\right]$$

From these three equations, $A_{PQR}(\mathbf{h})$, $B_{PQR}(\mathbf{h})$ and $C_{PQR}(\mathbf{h})$ are found to be

$$A_{PQR}(\mathbf{h})=2.476\exp[-0.25\pi h^2/a^2]$$

$$B_{PQR}(\mathbf{h}) = 1.517 \exp[-0.3333\pi h^2 / a^2]$$

$$C_{PQR}(\mathbf{h}) = 0.2499 \exp[-0.375\pi h^2 / a^2]$$

$A_{PQR}(\mathbf{h})$, $B_{PQR}(\mathbf{h})$ and $C_{PQR}(\mathbf{h})$ tend to 0 as $h \to \infty$. This can then be used in (30) for extending the structure factors.

We intend to accelerate the convergence of extending the structure factors by choosing $c(\mathbf{h})$ such that $A''_{PQR}(\mathbf{h})$, $B''_{PQR}(\mathbf{h})$ and $C''_{PQR}(\mathbf{h})$ are large for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$, and small otherwise. $A''_{PQR}(\mathbf{h})$, $B''_{PQR}(\mathbf{h})$ and $C''_{PQR}(\mathbf{h})$ tend to 0 as $h \to \infty$ (Sect. 2.5). Choices:

(i) Use (50).

$c(\mathbf{h}) = \exp[-\pi h^2 /(2a^2)]$ for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$,

or $= \eta \exp[-\pi h^2 /(2a^2)]$, otherwise.

(ii) Use (51) and (50).

$c(\mathbf{h}) = \exp[-\pi h^2 /(4a^2)]$ for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$,

or $= \eta \exp[-\pi h^2 /(2a^2)]$, otherwise.

(iii) Use

$c(\mathbf{h}) = \exp[-\pi h^2 /(na^2)]$ for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$,

n is an integer, $n \geq 5$. For example, (52), (53).

or $= \eta \exp[-\pi h^2 /(2a^2)]$, otherwise.

η is to be chosen (for examples, 0.01, 0.001, 0.0001).

$c(\mathbf{h})$ is chosen to have sharp discontinuity. $c(\mathbf{h}) \otimes c(\mathbf{h})$, $c(\mathbf{h}) \otimes c(\mathbf{h}) \otimes c(\mathbf{h})$ and $c(\mathbf{h}) \otimes c(\mathbf{h}) \otimes c(\mathbf{h}) \otimes c(\mathbf{h})$ are smooth functions of $\mathbf{h}$. Hence $A''_{PQR}(\mathbf{h})$, $B''_{PQR}(\mathbf{h})$ and $C''_{PQR}(\mathbf{h})$ determined in (46), (47) and (48) have the sharp discontinuity of $c(\mathbf{h})$, and are large for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$, and small otherwise.

It needs to verify by numerical calculations that choices (i), (ii) or (iii) will have $A''_{PQR}(\mathbf{h})$, $B''_{PQR}(\mathbf{h})$ and $C''_{PQR}(\mathbf{h})$ tend to 0 as $h \to \infty$. In case that this is not true, we can try the other direction:

(iv) Use

$c(\mathbf{h}) = \exp(-\pi h^2 / a^2)$ for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$,

or $= \eta \exp[-\pi h^2 /(2a^2)]$, otherwise.

As in Sayres (1952), p. 61, Woolfson (1958a), p. 279, the atoms (49) are not entirely free of overlap. The modified atoms in the choices (i) – (iv) may overlap, and have negative regions. Hence there is approximation. However, this is less important (Shiono & Woolfson (1991), Woolfson (1991), p. 44, Fan (1998), p. 81). Further investigations are needed to have optimum choices of $c(\mathbf{h})$ for three-dimensional crystal, and that the modified atoms satisfy the conditions of non-negativity and atomicity (Fan (1998), p. 79)). May use functions other than Gaussian functions.

2.7. Scheme of extension

Same as Sect. 2.3.

2.8. Evaluation of $A_{PQR}(\mathbf{h})$, $B_{PQR}(\mathbf{h})$ and $C_{PQR}(\mathbf{h})$

In the evaluation of $F(\mathbf{h})$ for all $\mathbf{h}$ in Sect. 2.5, we have not used (30) and $A_{PQR}(\mathbf{h})$, $B_{PQR}(\mathbf{h})$ and $C_{PQR}(\mathbf{h})$. This is because we have used $F(\mathbf{h})/\hat{f}(\mathbf{h})$ in (31) to transform the crystal to a point-atom crystal. Here the property of the original crystal is not used, for example, the original crystal may be real or complex. This means that **accelerated convergence of the extension of the structure factors by means of the modification of Sayre equation is, in fact, for crystals in general.** A similar conclusion is found in the sharpening of Patterson function, and extension of

Bragg intensities. From the point-atom crystal, we can then treat the most general modified atom $\xi(\mathbf{r})$ (from (39)) and the most general modified electron density in (40). $c(\mathbf{h})$ chosen in Sect. 2.5 and 2.6 has no physical meaning. It is just a mathematical technique for accelerating the convergence. There can be more than one optimum choices of $c(\mathbf{h})$. Transform the crystal to point-atom crystal avoids the problem of evaluating $A_{PQR}(\mathbf{h})$, $B_{PQR}(\mathbf{h})$ and $C_{PQR}(\mathbf{h})$. In fact, from $F(\mathbf{h})$ for all $\mathbf{h}$, we can use (30) to obtain we a relation between $A_{PQR}(\mathbf{h})$, $B_{PQR}(\mathbf{h})$ and $C_{PQR}(\mathbf{h})$, which can be used in the evaluation of $A_{PQR}(\mathbf{h})$, $B_{PQR}(\mathbf{h})$ and $C_{PQR}(\mathbf{h})$. This is another achievement of acceleration of the convergence of extension of structure factors.

2.9. Accelerated convergence of extended structure factors of crystal containing more than three types of atoms

Similar treatment can be used, employing Woolfson (1958), (23),

$$F(\mathbf{h}) = A(\mathbf{h})V^{-1}\sum_{\mathbf{k}} F(\mathbf{k})F(\mathbf{h}-\mathbf{k}) + B(\mathbf{h})V^{-2}\sum_{\mathbf{k}}\sum_{\mathbf{l}} F(\mathbf{k})F(\mathbf{l})F(\mathbf{h}-\mathbf{k}-\mathbf{l})$$
$$+C(\mathbf{h})V^{-3}\sum_{\mathbf{k}}\sum_{\mathbf{l}}\sum_{\mathbf{m}} F(\mathbf{k})F(\mathbf{l})F(\mathbf{m})F(\mathbf{h}-\mathbf{k}-\mathbf{l}-\mathbf{m}) + \ldots \tag{54}$$

3. Discussions and conclusion

By means of modifying the shape of the atoms, we obtain accelerated convergence of extension of structure factors for crystals containing two, three or more than three types of atoms (Sect. 2.1, 2.5, 2.9). The modified electron density has same atomic coordinates as the original crystal. An effective method is obtained: For crystal containing three types of atoms, the problem of accelerated convergence of extended structure factors reduces to the problem of choosing $c(\mathbf{h})$ such that $A''_{PQR}(\mathbf{h})$, $B''_{PQR}(\mathbf{h})$ and $C''_{PQR}(\mathbf{h})$ are large for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$ (magnitude and phase) (see (42)), and small otherwise. $A''_{PQR}(\mathbf{h})$, $B''_{PQR}(\mathbf{h})$ and $C''_{PQR}(\mathbf{h})$ tend to 0 as $h \to \infty$. This is necessary for the convergence of the summation in (43). Results for crystal containing two types of atoms are similar. The method is applicable to macromolecular crystal. Extension of structure factors may be applicable to low-resolution phasing.

Choices of $c(\mathbf{h})$ are suggested in Sect. 2.2, 2.6. These have to be studied by numerical calculations. Further investigations are needed to have optimum choices of $c(\mathbf{h})$ for three-dimensional crystal, and that the modified atoms satisfy the conditions of non-negativity and atomicity (Fan (1998), p. 79)). Different optimum choices should lead to the same extended structure factors. In fact, different optimum choices leading to the same result indicates that the result is correct.

$A_{PQ}(\mathbf{h})$ and $B_{PQ}(\mathbf{h})$, $A_{PQR}(\mathbf{h})$, $B_{PQR}(\mathbf{h})$ and $C_{PQR}(\mathbf{h})$ are not used in the accelerated convergence of extension of structure factors. It is found that accelerated convergence of the extension of the structure factors by means of the modifications of Sayre's equation, is, in fact, for crystals in general. Moreover, it provides relations between $A_{PQ}(\mathbf{h})$ and $B_{PQ}(\mathbf{h})$, and between $A_{PQR}(\mathbf{h})$, $B_{PQR}(\mathbf{h})$ and $C_{PQR}(\mathbf{h})$. These are useful in evaluation of $A_{PQ}(\mathbf{h})$ and $B_{PQ}(\mathbf{h})$, $A_{PQR}(\mathbf{h})$, $B_{PQR}(\mathbf{h})$ and $C_{PQR}(\mathbf{h})$ (Sect. 2.4, 2.8). It can be compared with the results obtained from the scattering factors of the electron densities and their squares, cubes and quadruples, of the two or three types of atoms.

It is useful to investigate the applicability of (18), (43), and the chosen $c(\mathbf{h})$ when the conditions of non-negativity and atomicity are not satisfied by the modified electron density (Shiono & Woolfson (1991), Fan (1998), p. 79, 81). It is mentioned in Woolfson (1991), p. 44 that, in terms of the normalized structure factors, 'Sayre equation holds reasonably well over a very large range of conditions'.

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34

A solution of the phase problem of one-dimensional centrosymmetric unequal-atom crystal by means of accelerated convergence of modifications of Sayre's equation

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Abstract

A deterministic and routine method is provided for the solution of the phase problem of a one-dimensional centrosymmetric unequal-atom crystal. Accelerated convergence of modifications of Sayre's equation is employed.

Keywords: Deterministic method; centrosymmetric; macromolecular crystal.

1. Introduction

Sayre (1952) presents the very useful and important Sayre's equation for equal-atom crystal. Woolfson (1958) presents very useful and important modifications for unequal-atom crystal. As Sayre's equation and its modifications have asymptotical validity, they require a large number of terms in the summation (Giacovazzo (1998), p. 307). This has experimental limitation. To overcome this difficultly and to reduce the amount of computation, accelerated convergence is introduced (Yuen (2011a, 2011c).

A solution of the phase problem of one-dimensional centrosymmetric equal-atom crystal by means of accelerated convergence of Sayre's equation is presented in Yuen (2011b). Using the same approach, and employing accelerated convergence of modifications of Sayre's equation, a deterministic and routine method is provided, in this article, for the solution of the phase problem of a one-dimensional centrosymmetric crystal of two types of atoms (Sect. 2.1) and four types of atoms (Sect. 2.2). The method can be applied to the phase problem for three-dimensional crystal.

2.1. Solution of the phase problem of one-dimensional centrosymmetric crystal containing two types of atoms, P and Q

From Yuen (2011c), (18),

$$F''(\mathbf{h}) = A''_{PQ}(\mathbf{h})V^{-1}\sum_{\mathbf{k}} F''(\mathbf{k})F''(\mathbf{h}-\mathbf{k})$$

$$+ B''_{PQ}(\mathbf{h})V^{-2}\sum_{\mathbf{k}}\sum_{\mathbf{l}} F''(\mathbf{k})F''(\mathbf{l})F''(\mathbf{h}-\mathbf{k}-\mathbf{l}) \qquad (1)$$

From Yuen (2011c), (14), the modified structure factor

$$F''(\mathbf{h}) = \frac{F(\mathbf{h})}{f(\mathbf{h})}c(\mathbf{h}) \qquad (2)$$

$c(\mathbf{h})$ is chosen such that the modified atom $\xi(\mathbf{r})$ (Yuen (2011c), Sect. 2.1) is non-negative and confined to a radius of about 0.65Å, and, for Gaussian atom, $c(\mathbf{h})$ has a radius of 1.3Å^{-1} (Sayre (1952), p. 61).

For $\mathbf{h}$ a reciprocal lattice vector, $c(\mathbf{h})$ is chosen positive.

$c(\mathbf{h})$ is chosen such that $A''_{PQ}(\mathbf{h})$ and $B''_{PQ}(\mathbf{h})$ are large for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$ (magnitude and phase) (see (2)), and small otherwise. $A''_{PQ}(\mathbf{h})$ and $B''_{PQ}(\mathbf{h})$ tend to 0 as $h \to \infty$. From (1), $F''(\mathbf{h})$ from the known $F(\mathbf{h})$ are large, and small otherwise.

Consider the one-dimensional example of Woolfson (1958), Sect. 3. Use (1) for acceleration of extension of modified structure factors, and follow Yuen (2011b), Sect. 2. The crystal structure is determined.

2.2. Solution of the phase problem of one-dimensional centrosymmetric crystal containing four types of atoms, P, Q, R and S

From Woolfson (1958), (23), and Yuen (2011c), (54),

$$F''(\mathbf{h}) =$$

$$
\begin{aligned}
& A''_{PQRS}(\mathbf{h}) V^{-1} \sum_{\mathbf{k}} F''(\mathbf{k}) F''(\mathbf{h}-\mathbf{k}) \\
& + B''_{PQRS}(\mathbf{h}) V^{-2} \sum_{\mathbf{k}} \sum_{\mathbf{l}} F''(\mathbf{k}) F''(\mathbf{l}) F''(\mathbf{h}-\mathbf{k}-\mathbf{l}) \\
& + C''_{PQRS}(\mathbf{h}) V^{-3} \sum_{\mathbf{k}} \sum_{\mathbf{l}} \sum_{\mathbf{m}} F''(\mathbf{k}) F''(\mathbf{l}) F''(\mathbf{m}) F''(\mathbf{h}-\mathbf{k}-\mathbf{l}-\mathbf{m}) \\
& + D''_{PQRS}(\mathbf{h}) V^{-4} \sum_{\mathbf{k}} \sum_{\mathbf{l}} \sum_{\mathbf{m}} \sum_{\mathbf{n}} F''(\mathbf{k}) F''(\mathbf{l}) F''(\mathbf{m}) F''(\mathbf{n}) F''(\mathbf{h}-\mathbf{k}-\mathbf{l}-\mathbf{m}-\mathbf{n})
\end{aligned}
\tag{3}
$$

For $\mathbf{h}$ a reciprocal lattice vector, $c(\mathbf{h})$ is chosen positive.

$c(\mathbf{h})$ is chosen such that $A''_{PQRS}(\mathbf{h})$, $B''_{PQRS}(\mathbf{h})$, $C''_{PQRS}(\mathbf{h})$ and $D''_{PQRS}(\mathbf{h})$ are large for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$ (magnitude and phase) (see (2)), and small otherwise. $A''_{PQRS}(\mathbf{h})$, $B''_{PQRS}(\mathbf{h})$, $C''_{PQRS}(\mathbf{h})$ and $D''_{PQRS}(\mathbf{h})$ tend to 0 as $h \to \infty$. From (3), $F''(\mathbf{h})$ from the known $F(\mathbf{h})$ are large, and small otherwise.

Consider the one-dimensional example of Woolfson (1958), Sect. 7. Use (3) for acceleration of extension of modified structure factors, and follow Yuen (2011b), Sect. 2. The crystal structure is determined.

3. Discussions and conclusion

A deterministic and routine method is provided for the solution of the phase problem of a one-dimensional centrosymmetric crystal of two and four types of atoms. Accelerated convergence of extension of structure factors by means of modifications of Sayre's equation is employed.

Investigation of applying the method to three-dimensional crystal is needed, in particular, macromolecular crystal. It is useful to incorporate with other methods in crystallography. Direct methods may be employed to obtain the signs of some reflections, so that amount of computation can be reduced.

It is useful to investigate the applicability of (1) and (3) when the non-negativity and atomicity conditions are not satisfied by the modified electron density (Fan (1998), p. 79, 81).

Simplifications such as reducing the radius of the sphere of reflections, or, only strongest 25% and weakest 25% reflections are included in the summation in (1) and (3), can be explored.

'Sharpening' of $c(\mathbf{h})$ in the reciprocal space may be applied. The radius of the sphere of reflections is reduced. Hence smaller number of reflections in Group A (Yuen (2011b), Sect. 2) is used. $c(\mathbf{h})$ is the scattering factor (that is, the Fourier transform) of the modified atom $\xi(\mathbf{r})$ ((Yuen (2011c), Sect. 2.1, 2.5). $\xi(\mathbf{r})$ is broadened. There is overlapping of the modified atoms. We may use functions other than the Gaussian functions, so that $\xi(\mathbf{r})$ has the size of atomic radius while $c(\mathbf{h})$ is very sharp. For example, the Fourier transform of the rectangular function is $\sin c(s) = \sin(\pi s)/(\pi s)$. For an atom of the form of a rectangular function with radius 0.65Å, the first zero of $\sin c(s)$ occurs at 0.769 Å^{-1}. Hence the number of reflections in Group A will be much reduced (Compare with the 1.3Å^{-1} for Gaussian atom (Yuen (2011c), Sect. 2.1). Investigation is needed to have optimum choice.

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35

Sharpening in reciprocal space. A set of minimum number of structure factors for generating all the structure factors for crystals of equal and unequal atoms

August 30, 2011

Abstract
By means of sharpening in reciprocal space, a set of minimum number of structure factors for generating all the structure factors for a crystal of equal or unequal atoms can be obtained empirically. $c(\mathbf{h})$ is sharp. The modified atom $\xi(\mathbf{r})$ has broad shape.

Keywords: Empirical; non-negativity; atomicity.

1. Introduction

Employing accelerated convergence of extension of structure factor by means of Sayre's equation (Yuen (2011a, 2011c)), all structure factors can be generated by a set of structure factors. If a set of minimum number of structure factors for generating all the structure factors can be determined, the other structure factors can be obtained by extension with acceleration. This is important and useful in X-ray crystallography, in particular, protein crystals which do not diffract to atomic resolution, or sensitive to radiations (Woolfson & Fan (1995), p. 172).

Methods in crystallography, for example, Yuen (2011b, 2011d), may be applied to the set of minimum number of structure factors. The amount of computation is much reduced. This is means that applicability of these methods is extended to larger crystals. In Yuen (2011b, 2011d), Sect. 3, it is mentioned that 'sharpening' of $c(\mathbf{h})$ in the reciprocal space may be applied. The radius of the sphere of reflections is reduced. Hence smaller number of reflections is used. $c(\mathbf{h})$ is the scattering factor of the modified atom $\xi(\mathbf{r})$. $c(\mathbf{h})$ is sharp. $\xi(\mathbf{r})$ has broad shape. There are large regions of overlapping of the modified atoms. $\xi(\mathbf{r})$ may have negative ripples. The three conditions (non-negativity, atomicity and equal-atom structure) for the application of Sayre's equation (Fan (1998), p. 79) may not be satisfied. This article suggests that we determine empirically how sharp $c(\mathbf{h})$ can be, or how severe such violations can be tolerated for a crystal containing two types of atoms (Sect. 2.1) or three types of atoms (Sect. 2.2).

2.1. A set of minimum number of structure factors for generating all the structure factors for a crystal of equal atoms

Use Yuen (2011a), Sect 2.1. $c(\mathbf{h})$ is chosen such that $c(\mathbf{h})/[c(\mathbf{h}) \otimes c(\mathbf{h})]$ is large for the modified structure factors $F''(\mathbf{h})$ from the known $F(\mathbf{h})$, and small otherwise. $c(\mathbf{h})/[c(\mathbf{h}) \otimes c(\mathbf{h})]$ tends to 0 as $h \to \infty$.

For example, in Sayre (1952), p. 61, or Yuen (2011a), (19), we have the Gaussian atom $\exp[-2\pi(ar)^2]$ (1)

It drops to about 0.07 at $ar = 0.65$Å,

The atomic scattering factor = $[1/(\sqrt{2}a)]\exp[-\pi h^2/(2a^2)]$ (2)

It drops to about 0.07 of its central value at $h/a = 1.3$Å^{-1}. (3)

If we use a modified atom $\xi(\mathbf{r}) = \exp[-\pi(ar)^2]$ (4)

It drops to 0.07 at $ar = 0.920$ Å. (5)

This is larger than that in (3), and there is overlapping of the modified atoms $\xi(\mathbf{r})$.

Its scattering factor $c(\mathbf{h}) = (1/a)\exp(-\pi h^2/a^2)$ (6)

It drops to 0.07 at $h/a = 0.920$ Å^{-1}. (7)

Comparing with (5), we have sharpening in the reciprocal space.

$$c(\mathbf{h}) \otimes c(\mathbf{h}) = [1/(\sqrt{2}a)]\exp[-\pi h^2/(2a^2)] \qquad (8)$$

$$c(\mathbf{h})/[c(\mathbf{h}) \otimes c(\mathbf{h})] = \sqrt{2}\exp[-\pi h^2/(2a^2)] \qquad (9)$$

$c(\mathbf{h})/[c(\mathbf{h}) \otimes c(\mathbf{h})]$ tends to 0 as $h \to \infty$.

As in Yuen (2011a), Sect. 2.2, for accelerated convergence of extension of structure factor, we may choose

$c(\mathbf{h}) = \exp(-\pi h^2 / a^2)$ for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$,

or $= \eta \exp[-\pi h^2 / a^2]$, otherwise.

η is to be chosen (for examples, 0.01, 0.001, 0.0001).

For this choice of $c(\mathbf{h})$, we have to verify numerically that $c(\mathbf{h})/[c(\mathbf{h}) \otimes c(\mathbf{h})]$ tends to 0 as $h \to \infty$. Investigations are needed to have optimum choices of $c(\mathbf{h})$ for three-dimensional crystal. May use functions other than Gaussian functions.

Now we intend to investigate how sharp $c(\mathbf{h})$ can be, i.e. what is the minimum number of reflections needed for generating all the structure factors? If the number of reflections is equal to the number of atomic coordinates to be determined, a set of atomic coordinates may be obtained. The solution may not be unique as each atom can be in any position of the crystal. If the structure obtained must satisfy the environment of the atoms, including distances between nearest-neighbors, next nearest-neighbors, bond lengths and bond angles, etc., a unique set of atomic coordinates may be determined. Moreover, if the structure obtained must satisfy such atomic environments, the minimum number of reflections needed may be less than the number of atomic coordinates to be determined. Hence, for a number of crystals of known structures, we can use $c(\mathbf{h})$ as sharp as possible, accelerated convergence of extension of structure factors is then applied. $\xi(\mathbf{r})$ has broad shape. There will be large regions of overlapping of the modified atoms $\xi(\mathbf{r})$. $\xi(\mathbf{r})$ may have negative regions. See how severe violations of the conditions of atomicity and non-negativity can be tolerated. See whether we can obtain all the structure factors. The admissible sharpness of $c(\mathbf{h})$ can be established empirically. For centrosymmetric crystal, the structure factor can only be positive or negative. This is a very strong condition. $c(\mathbf{h})$ can be very sharp, and the minimum number of structure factors may be very small.

2.2. A set of minimum number of structure factors for generating all the structure factors for a crystal of unequal atoms

Follow Sect. 2.1.

For crystal containing two types of atoms, use Yuen (2011c), Sect. 2.1. $c(\mathbf{h})$ is chosen such that $A''_{PQ}(\mathbf{h})$ and $B''_{PQ}(\mathbf{h})$ are large for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$, and small otherwise. $A''_{PQ}(\mathbf{h})$ and $B''_{PQ}(\mathbf{h})$ tend to 0 as $h \to \infty$.

For crystal containing three types of atoms, use Yuen (2011c), Sect. 2.5. $c(\mathbf{h})$ is chosen such that $A''_{PQR}(\mathbf{h})$, $B''_{PQR}(\mathbf{h})$ and $C''_{PQR}(\mathbf{h})$ are large for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$, and small otherwise. $A''_{PQR}(\mathbf{h})$, $B''_{PQR}(\mathbf{h})$ and $C''_{PQR}(\mathbf{h})$ tend to 0 as $h \to \infty$.

Choice of $c(\mathbf{h})$ in Sect. 2.1 may be used.

For a number of crystals containing two, three or more than three types of atoms (Yuen (2011c), Sect. 2.9) and of known structures, we can use $c(\mathbf{h})$ as sharp as possible. Accelerated convergence of extension of structure factors is then applied. See how severe violations of the conditions of atomicity, non-negativity, and equal-atom structure can be tolerated. The admissible sharpness of $c(\mathbf{h})$ is established empirically.

3. Discussions and conclusion

For crystals containing equal or unequal atoms, with optimum choice of $c(\mathbf{h})$, we can obtain empirically a set of minimum number of structure factors or modified structures factors which generate all the structure factors. As this set is obtained empirically from a number of crystals of known structures, the environment of atoms, such as distances between nearest-neighbors, next nearest-neighbors, bond lengths and bond angles, etc, are included implicitly (see McCusker & Baerlocher (2006)). Experimentally we need only measure these reflections. This is very useful, in particular, for macromolecular crystals. ‘We need only measure these reflections’ is ideal; experimentally, it is better to measure the reflections to as high resolution as possible.

In powder diffraction pattern, there is much overlapping (David (1987)). We may be able to resolve the low-order reflections. The higher-order reflections may then be obtained by extension with acceleration. These higher-order reflections can be used for resolving the overlapping higher-order reflections in powder diffraction pattern. It can be combined with the other methods in David & Sivia (2006), Peschar et al. (2006), Sect. 10.4, 10. 5, Giacovazzo et al. (2006), Sect. 11.3, 11.5.

Methods in crystallography, for example, Yuen (2011b, 2011d), may be applied to the set of minimum number of structure factors or modified structure factors. For non-centrosymmetric crystals, single isomorphous replacement or one-wavelength anomalous scattering (Stout & Jensen (1989), Sect. 13.2, 13.3, Woolfson & Fan (1995), Ch. 4, 5, 6) can be used. There is ambiguity in the phases of the two $F(\mathbf{h})$. This is similar to the centrosymmetric case (Yuen (2011b)). As we need only deal with the set of minimum number of structure factors or modified structure factors, this means that applicability of methods in crystallography is extended to larger crystals.

The structure factors can be extended to atomic resolution. Direct methods can then be applied.

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36

Combination of methods in X-ray crystallography and iterative algorithms

November 29, 2011

Abstract
Sayre's equation and its modifications are incorporated with point-atom model, sharpening of Patterson function. Iterative algorithms are employed to alternate between point atoms, sharp atoms, moderate shape and broad shape of $\xi(\mathbf{r})$. The atomic coordinates are fixed in the Sayre's equation and its modifications, and sharpening of Patterson function. The main objective is to determine the atomic coordinates.

Keywords: Point atom; sharpening; moderate shape; broad shape.

1. Introduction

In the above, we have presented articles about iterative algorithms, point-atom model, sharpening of Patterson function, and Sayre's equation and its modifications. Now we try to combine together these approaches.

2.1. Iterative algorithms applied to the most general modified electron density $\rho''(\mathbf{r})$ and the most general modified structure factor $F''(\mathbf{h})$

In Yuen (2011g, 2011h), we have accelerated convergence of extension of structure factors by means of Sayre's equation and its modifications for crystals of equal atoms and unequal atoms. In Yuen (2011g), Sect. 2.1, and Yuen (2011h), Sect. 2.1, 2.5, there are the most general modified structure factor $F''(\mathbf{h})$, the most general modified atom $\xi(\mathbf{r})$, and the most general modified electron density $\rho''(\mathbf{r})$. $c(\mathbf{h})$ and $\xi(\mathbf{r})$ have moderate shapes. $F''(\mathbf{h})$ from the known $F(\mathbf{h})$ is large, and small otherwise. Hence $F''(\mathbf{h})$ for all $\mathbf{h}$ can be used. This means that there is no Fourier series truncation error. Iterative algorithms can be applied to the most general modified electron density $\rho''(\mathbf{r})$ and the most general modified structure factor $F''(\mathbf{h})$ for all $\mathbf{h}$ (Yuen (2011a, 2011b)). No oversampling is used. Initial phases in Yuen (2011c) may be employed.

2.2. Iterative algorithms applied to the minimum number of structure factors

In Yuen (2011i), we have sharpening of $c(\mathbf{h})$ in reciprocal space. The modified atom $\xi(\mathbf{r})$ has broad shape. We obtain a set of minimum number of structure factors for generating all the structure factors for crystal of equal or unequal atoms. Iterative algorithms can be applied to the modified electron density, and the minimum number of structure factors. The modified atoms may have large regions of overlap, and there are negative regions. No oversampling is used. Initial phases in Yuen (2011c) may be employed.

As the number of structure factors used is small, the amount of computation is reduced, at the expense that the modified atoms may have large regions of overlap, and there are negative regions. As mentioned in Yuen (2011g), Sect. 2.1, and Yuen (2011h), Sect. 2.1, 2.5, the atomic coordinates of the modified atoms $\xi(\mathbf{r})$ are the same as those of the crystal. The gross shapes of the molecules and the atomic coordinates can be determined. The shapes of the modified atoms $\xi(\mathbf{r})$ may be difficult to identify because of the large regions of overlapping. Moreover $\xi(\mathbf{r})$ has broad shape which may be very different from the atoms of the crystal.

2.3. Combination of Sect. 2.1 and 2.2.

In Sect. 2.1, $\xi(\mathbf{r})$ and $c(\mathbf{h})$ have moderate shapes. A large number of modified structure factors $F''(\mathbf{h})$ is used.

In Sect. 2.2, $\xi(\mathbf{r})$ has broad shape, and $c(\mathbf{h})$ is sharpened. A minimum number of structure factors is used.

In both Sect. 2.1 and 2.2, the atomic coordinates are the same as those of the crystal.

Hence we can use iterative algorithms which alternate between the processes in Sect. 2.1 and 2.2, i.e. alternate between moderate shape and broad shape of $\xi(\mathbf{r})$. The main objective is to determine the atomic coordinates, which is, in fact, the main purpose of X-ray crystallography. The shape of $\xi(\mathbf{r})$ is immaterial.

2.4. Combination of Sect. 2.3, point-atom model, and sharpening of Patterson function

In dual-real-spaces iterative algorithms applied to electron density function and Patterson function of a crystal, plus acceleration of extension of Bragg intensities (Yuen (2011e, 2011f), point-atom model is used in the electron density function, and sharpened Patterson peaks (hence sharp atoms) are used for the Patterson function (Yuen (2011d).

A large number of structure factors are used.

In Sect. 2.1, 2.2, and sharpening of Patterson function, the atomic coordinates are the same as those of the crystal.

Hence we can use iterative algorithms which alternate between the processes in Yuen (2011f), and Sect. 2.1, 2.2, i.e. alternate between point atoms, sharp atoms, moderate shape and broad shape of $\xi(\mathbf{r})$. The main objective is to determine the atomic coordinates. The shapes of the atoms are immaterial.

3. Discussions and conclusion

We have incorporate Sayre’s equation and its modifications with point-atom model and sharpening of Patterson function. Iterative algorithms are employed to alternate between point atoms, sharp atoms, moderate shape and broad shape of $\xi(\mathbf{r})$. The main objective is to determine the atomic coordinates. The basic idea is to use different shapes of the atoms, with the atomic coordinates fixed in the Sayre’s equation and its modifications, and sharpening of Patterson function, and try to determine the atomic coordinates.

After the atomic coordinates are determined, refinement in X-ray crystallography can be applied to obtain shapes of the atoms.

The next step will be to combine these iterative algorithms with other methods in X-ray crystallography, for examples, isomorphous replacement and direct methods.

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X-ray crystallography

(in-between Fourier coefficients included)

37

Extension of the Patterson function to include the in-between｜Fourier coefficients｜.

Patterson functions of crystal and a unit cell

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Abstract

This article presents the extension of the Patterson function to include the in-between ｜Fourier coefficients｜. The in-between｜Fourier coefficients｜ are included in the Patterson function of a unit cell. The Patterson function of a crystal and the Patterson function of a unit cell are complementary to each other. Together, they can be used for crystal atomic-coordinate determination.

Keywords: Atomicity; periodic convolution; circular convolution; autocorrelation; deconvolution; mid-Bragg intensities; in-between structure factors; Shannon sampling; cardinal series.

1. Introduction

Extension of the conventional methods in X-ray crystallography to include the in-between Fourier coefficients opens a new phase in X-ray crystallography. Many researches can be done. The results obtained from this inclusion and the results obtained by conventional method are complementary to each other. These results, together, can be used for crystal atomic-coordinate determination.

The total number of｜Fourier coefficients｜ and in-between｜Fourier coefficients｜ is eight times that of the｜Fourier coefficients｜ (observed structure factors). We have the same set of atomic coordinates to be determined. This total number is much larger than the number of atomic coordinates. The system is excessively over-determined. The extension of the conventional methods in X-ray crystallography to include the in-between | Fourier coefficients | (in-between structure factors) will be very useful in crystal atomic-coordinate determination, in particular, for macromolecular crystals. From the | Fourier coefficients | , the in-between | Fourier coefficients | can be obtained by interpolation in multidimensions of | Fourier coefficients | or | Fourier coefficients $|^2$ (Bates (1984), p. 216, Kincaid & Cheney (1996), Sect. 6.10, Cheney & Light (2000), Anita (2002), Sect. 4.7, 4.8, Hau-Riege et al. (2004)). From the two interpolated values, a more reliable result can be obtained. The interpolated values are more accurate because the reciprocal lattice points are closely spaced (Yuen (2011a), Appendix). Hence for the same set of observed | structure factors | , we have increased the data eight times by means of interpolation. Extrapolation of observed | Fourier coefficients | has been used and called *free-lunch method* (Milanesio & Viterbo (2011), Sect. 6.3.2.1 and Zanotti (2011), p. 733). Now with interpolation, there are eight times the number of observed | Fourier coefficients | . This is a *free-dinner method*. Moreover, extrapolation is improved by employing these | Fourier coefficients | and in-between | Fourier coefficients | .

The first topic to be investigated is, naturally, the extension of the Patterson function to include the in-between | Fourier coefficients | , which is presented in this article. The in-between | Fourier coefficients | are included in the Patterson function of a unit cell.

Weirich (2006) presents a survey of conventional methods for solving the phase problem.

Sayre (1952) uses Patterson function of a unit cell. Mishnev ((1996), p. 629, (1998), p. 478) shows that the Patterson function of a unit cell 'is less subjected to the peak superposition inherent to the standard Patterson, has more pronounced maxima'. Confined structure is presented in De Caro et al. (2002). Hughes (1953) presents a generalization of the Patterson function.

Giacovazzo et al. (1998, 1999a, 1999b, 1999c) presents the first step for development of direct methods involving rational index structure factors.

2.1. Patterson function of crystal

Consider a crystal of real or complex electron density $\rho(\mathbf{r})$. Form the autocorrelation (Papoulis (1977), p. 64),

$$p(\mathbf{r}) = \rho(\mathbf{r}) \,\overline{\otimes}\, \rho^*(-\mathbf{r}) \tag{1}$$

$$= \int \rho(\mathbf{s})\rho^*(-(\mathbf{r}-\mathbf{s}))d\mathbf{s} = \int \rho(\mathbf{r}+\mathbf{s})\rho^*(\mathbf{s})d\mathbf{s} \tag{2}$$

$\overline{\otimes}$ denotes periodic or circular convolution (Oppenheim et al (1983), pp. 133, 217, 263).

The integration is over one unit cell.

$p(\mathbf{r})$ is also a periodic function with same unit cell constants.

Let $U = abc$, where a,b,c are the unit cell edges, c_{phkl} and $c_{\rho hkl}$, $h,k,l = 0,\pm1,\pm2,\dots$, be the Fourier coefficients of $p(\mathbf{r})$ and $\rho(\mathbf{r})$ respectively.

$$c_{phkl} = Uc_{\rho hkl}c_{\rho hkl}^* = U\left|c_{\rho hkl}\right|^2 \tag{3}$$

(Oppenheim et al (1983), (4.133)).

$p(\mathbf{r})$ is the Patterson function of the crystal (Yuen (2008), (B)).

Because of atomicity, we have the structure factor

$$F(\mathbf{h}) = \sum_{n=1}^{N} f_n(\mathbf{h})\exp(2\pi i\mathbf{h}\cdot\mathbf{r}_n), \tag{4}$$

In terms of the structure factor, the Patterson function of a crystal

$$p(\mathbf{r}) = \frac{1}{V}\sum_{\mathbf{h}}\left|F(\mathbf{h})\right|^2 \exp[-2\pi i(\mathbf{h}\cdot\mathbf{r})] \tag{5}$$

N is the number of atoms in a unit cell, V the volume of one unit cell. The factor V^{-1} is used as in Ladd & Palmer (1985), (6.54), (6.55).

From (4), $\left|F(\mathbf{h})\right|^2 = \sum_{n=1}^{N}\sum_{m=1}^{N} f_n(\mathbf{h})[f_m(\mathbf{h})]^*\exp[2\pi i\mathbf{h}\cdot(\mathbf{r}_n-\mathbf{r}_m)]$,

If $\rho(\mathbf{r})$ is real, the Patterson function is real. (2) becomes

$$p(\mathbf{r}) = \int \rho(\mathbf{s})\rho(-(\mathbf{r}-\mathbf{s}))d\mathbf{s} = \int \rho(\mathbf{s})\rho(-\mathbf{r}+\mathbf{s})d\mathbf{s} = \int \rho(\mathbf{r}+\mathbf{s})\rho(\mathbf{s})d\mathbf{s}$$

(6)

In (6), $p(\mathbf{r}) = p(-\mathbf{r})$. (7)

Hence, for real electron density, the Patterson function is centrosymmetric.

If $\rho(\mathbf{r})$ is non-negative, the Patterson function is non-negative.

Patterson function of complex $\rho(\mathbf{r})$ is discussed in Buerger (1959), pp.77-79, Ramachandran & Srinivasan (1970), pp. 206-210.

Consider real atoms. Let Z_n and $f_n(\mathbf{h})$ be the atomic number and atomic scattering factor of the n-th atom respectively. The atomic scattering factors have approximately same shapes. Let

$$\hat{f}(\mathbf{h}) = (\sum f_n(\mathbf{h}))/(\sum Z_n) \tag{8}$$

$\hat{f}(\mathbf{h})$ is the atomic scattering factor of a hypothetical atom with $Z = 1$ (Harker & Kasper (1948), (11)).

$$f_n(\mathbf{h}) \approx Z_n \hat{f}(\mathbf{h}) \tag{9}$$

Substitute (4) and (9) into (5),

$$p(\mathbf{r}) \approx \frac{1}{V} \sum_{n=1}^{N} \sum_{m=1}^{N} Z_n Z_m G[\mathbf{r} - (\mathbf{r}_m - \mathbf{r}_n)] \tag{10}$$

where $G(\mathbf{r}) = \sum_{\mathbf{h}} \left| \hat{f}(\mathbf{h}) \right|^2 \exp[-2\pi i(\mathbf{h} \cdot \mathbf{r})]$ (11)

$G(\mathbf{r})$ has the form of a peak at the origin. From (10), this peak is shifted to each point given by interatomic vectors $\mathbf{r}_m - \mathbf{r}_n$. The magnitude of the peak is modified by $Z_n Z_m$ (Ladd & Palmer (1985), p. 233, Stout & Jensen (1989), p. 283, Woolfson & Fan (1995), p. 35). Thus, this displacement gives rise to the Patterson peaks which are very useful in crystal atomic-coordinate determination. If $\hat{f}(\mathbf{h})$ =1, $G(\mathbf{r})$ is a point atom.

2.2. Extension of the Patterson function to include the in-between | Fourier

coefficients| . Patterson function of a unit cell

Consider a unit cell of the crystal as an object. $\rho(\mathbf{r})$ is real or complex.

Similar to (1) and (2),

$$p_c(\mathbf{r}) = \rho(\mathbf{r}) \otimes \rho^*(-\mathbf{r}) \tag{12}$$

$$= \int \rho(\mathbf{s})\rho^*(-(\mathbf{r}-\mathbf{s}))d\mathbf{s} = \int \rho(\mathbf{r}+\mathbf{s})\rho^*(\mathbf{s})d\mathbf{s} \tag{13}$$

$\otimes$ denotes convolution.

$p_c(\mathbf{r})$ is the autocorrelation of $\rho(\mathbf{r})$. $p_c(\mathbf{r})$ is the Patterson function of the unit cell. Note that this is not a unit cell of the Patterson function.

$p_c(\mathbf{r})$ has size (support) in $[-a,a]$, $[-b,b]$, $[-c,c]$. Convolution is a smoothing operation; $p_c(\mathbf{r})$ tapers to zero at the boundary.

If $\rho(\mathbf{r})$ is real, from (13), $p_c(\mathbf{r})$ is real and centrosymmetric.

If $\rho(\mathbf{r})$ is non-negative, $p_c(\mathbf{r})$ is non-negative.

Let $P(\mathbf{h})$ and $F(\mathbf{h})$ be the Fourier transforms of $p_c(\mathbf{r})$ and $\rho(\mathbf{r})$ respectively. $\mathbf{h}$ is any vector in the reciprocal space. $F(\mathbf{h})$ is the Fourier transform of a unit cell.

$$P(\mathbf{h}) = F(\mathbf{h})F^*(\mathbf{h}) = |F(\mathbf{h})|^2 \tag{14}$$

This is Yuen (2008), (A).

The Patterson function of a unit cell

$$p_c(\mathbf{r}) = \frac{1}{V}\int |F(\mathbf{h})|^2 \exp(-2\pi i\mathbf{h}\cdot\mathbf{r})d\mathbf{h}$$

V the volume of one unit cell. The factor V^{-1} is used as in Ladd & Palmer (1985), (6.54), (6.55).

We can apply Shannon sampling to $P(\mathbf{h}) = |F(\mathbf{h})|^2$ in (14) and obtain the Patterson function of the unit cell. To simplify the treatment, we treat the one-dimensional case.

$$P(h) = \left|F(h)\right|^2 \tag{15}$$

Consider the object in the interval [0, 1]. The electron density $\rho(x) = 0$ for $x \notin [0,1]$. x is in fractional unit.

Use the Shannon's sampling theory (Marks (1991), p. 1),

$$P(h) = \sum_{m=-\infty}^{\infty} \left|F(m/2)\right|^2 \sin c(2h - m) \tag{16}$$

The $\sin c$ function is defined as $\sin c(x) = \dfrac{\sin(\pi x)}{\pi x}$.

For even m, $F(m/2)$ are the Fourier coefficients, say $F(n)$.

For odd m, $F(m/2)$ are the in-between Fourier coefficients. $\left|F(m/2)\right|^2$ can be obtained by interpolation of | Fourier coefficients | , $\left|F(n)\right|$. The interpolated $\left|F(m/2)\right|$ is then squared. We can also obtain $\left|F(m/2)\right|^2$ by interpolation of | Fourier coefficients | ^{2}, $\left|F(n)\right|^2$. From these two results, a more reliable value is obtained.

The Patterson function of a unit cell $p_c(x)$ is given by the inverse Fourier transform of $P(h)$.

$$p_c(x) = \frac{1}{a}\int P(h)\exp(-2\pi ihx)dh \tag{17}$$

a is the unit cell length.

Substitute (16) into (17). We treat the cardinal series (16) as uniformly convergent (Marks (1991), Sect. 3.3.1, Higgins (1985), p. 70, Th. 1). The summation and the integration in (16) and (17) can be interchanged (Titchmarsh (1968), Sect. 1.71).

$$\int \sin c(h)\exp(-2\pi ihx)dh = \Pi(x)$$

The rectangular function $\Pi(x) = 1$, $|x| < 1/2$, or $=0,\ |x|>1/2,$

or $=1/2,\ |x|=1/2.$

$$\int \sin c(2h-m)\exp(-2\pi ihx)dh = \frac{1}{2}[\exp(-\pi imx)]\Pi(x/2) \tag{18}$$

Hence $$p_c(x) = \frac{1}{2a}\sum_{m=-\infty}^{\infty}|F(m/2)|^2 \cos(\pi mx)\Pi(x/2) \tag{19}$$

Return to the three-dimensional case (14). The object is given by the unit cell with $x \in [0,1]$, $y \in [0,1]$, $z \in [0,1]$. The electron density is zero outside this unit cell. Use the three-dimensional Shannon's sampling theory (Goodman (2005), (2-57)).

$$P(\mathbf{h}) =$$

$$\sum_{m_1,m_2,m_3}|F(m_1/2,m_2/2,m_3/2)|^2 \sin c[2h_1-m_1]\sin c[2h_2-m_2]\sin c[2h_3-m_3]$$

The summations of m_1, m_2, m_3 are from $-\infty$ to ∞.

When one or more of m_1, m_2, and m_3 is odd, the mid-Bragg intensity $|F(m_1/2,m_2/2,m_3/2)|^2$ can be obtained by interpolation of | Fourier coefficients | in multidimensions. The interpolated $|F(m_1/2,m_2/2,m_3/2)|$ is then squared. We can also obtain $|F(m_1/2,m_2/2,m_3/2)|^2$ by interpolation of the Bragg intensity, | Fourier coefficients | ^{2} in multidimensions. From these two results, a more reliable value is obtained.

Similar to (17) - (19), Patterson function of the unit cell

$$p_c(\mathbf{r}) = \frac{1}{8V}\sum_{m_1,m_2,m_3}|F(m_1/2,m_2/2,m_3/2)|^2 \cos[\pi(m_1x+m_2y+m_3z)]$$

$$\Pi(x/2)\Pi(y/2)\Pi(z/2) \tag{20}$$

$p_c(\mathbf{r})$ has size (support) in $[-1,1]$, $[-1,1]$, $[-1,1]$ (in fractional units).

As a unit cell is used as the object, an atom near a boundary surface of the unit cell will be divided into two parts, attaching to the two opposite boundary surfaces. An atom near an edge of the unit cell will be divided into four parts, attaching to four edges of the unit cell. An atom near a corner of the unit cell will be divided into eight parts, attaching to the eight corners of the unit cell. Each part is an atom which is not spherically symmetric. Hence the number of atoms in the unit cell inside the unit cell is increased. The Fourier transforms of $p_c(\mathbf{r})$ becomes

$$F(\mathbf{h}) = \sum_{n=1}^{N'} f_n'(\mathbf{h}) \exp(2\pi i \mathbf{h} \cdot \mathbf{r}_n)\,, \tag{21}$$

where $N' \geq N$ in (4).

$f_n'(\mathbf{h})$ is the atomic scattering factor of an atom if the atom is divided.

$\mathbf{r}_n$ is in fractional units.

When the number of atoms in a unit cell is large, for example, macromolecular crystal,

$$N' \approx N\,. \tag{22}$$

Consider real atoms. Instead of (9), we use an approximation

$$f_n(\mathbf{h}) \approx Z_n' \hat{f}(\mathbf{h}) \tag{23}$$

where Z_n' is for a part of the atom if the atom is divided. Z_n' may or may not be an integer. But for all the parts of a divided atom, the sum of the Z_n' is the atomic number of the atom.

Substitute (21) and (23) into (14).

$$P(\mathbf{h}) \approx \sum_{n=1}^{N'} \sum_{m=1}^{N'} Z_n' Z_m' \left| \hat{f}(\mathbf{h}) \right|^2 \exp[2\pi i \mathbf{h} \cdot (\mathbf{r}_n - \mathbf{r}_m)] \tag{24}$$

The Patterson function of the unit cell,

$$\begin{aligned} p_c(\mathbf{r}) &\approx \frac{1}{V} \sum_{n=1}^{N'} \sum_{m=1}^{N'} Z_n' Z_m' \int \left| \hat{f}(\mathbf{h})^2 \right| \exp[-2\pi i \mathbf{h} \cdot (\mathbf{r} - (\mathbf{r}_n - \mathbf{r}_m))] d\mathbf{h} \\ &\approx \frac{1}{V} \sum_{n=1}^{N'} \sum_{m=1}^{N'} Z_n' Z_m' G_c[\mathbf{r} - (\mathbf{r}_n - \mathbf{r}_m)] \end{aligned} \tag{25}$$

where $G_c(\mathbf{r}) = \int \left| \hat{f}(\mathbf{h}) \right|^2 \exp[-2\pi i \mathbf{h} \cdot \mathbf{r}] d\mathbf{h}$ (26)

$G_c(\mathbf{r})$ has the form of a peak at the origin. From (25), this peak is shifted to each point given by interatomic vectors $\mathbf{r}_n - \mathbf{r}_m$. The magnitude of the peak is modified by $Z_n' Z_m'$. Thus, this displacement gives rise to the Patterson peaks. If $\hat{f}(\mathbf{h})$ =1, $G_c(\mathbf{r})$ is a point atom.

The right-hand side of (25) is an approximation. For an atom-pair near the boundary surfaces, or edges, or corners of the support of the Patterson function of the unit cell, $[-1,1]$, $[-1,1]$, $[-1,1]$, $G_c(\mathbf{r})$ will extend beyond this support. For macromolecular crystal, the effect is reduced.

2.3. Demonstrations with crystal structures reported in the literature. The calculated in-between Fourier coefficients are good for determination of atomic coordinates

Choose a large number of crystal structures reported in the literature, of various sizes, small, mediate, or macromolecular, and with or without heavy atoms. As stated in Yuen (2011b), Sect. 2.2, the calculated in-between Fourier coefficients are reasonably good for determination of electron density. The use of the Patterson function is for determination of atomic coordinates. It is not as complicate as the electron density. Hence the calculated in-between Fourier coefficients are good for determination of atomic coordinates. Large uncertainties can be accepted. In fact, accurate atomic coordinates are determined in conventional method in X-ray crystallography. Hence the calculated in-between Fourier coefficients should be good for determination of atomic coordinates, including the Patterson peaks. The Patterson function of a unit cell of these crystal structures can be calculated. The Patterson peaks can be examined, and compared with the peaks of the Patterson functions of the crystals. The result of division of atoms near a boundary surface, an edge or a corner, can be examined.

3. Discussions and conclusion

The in-between | Fourier coefficients | are included in the Patterson function of a unit cell. An atom near a boundary surface, an edge or a corner of the unit cell are divided into parts. Each part is an atom which is not spherically symmetric. The number of atoms in the unit cell inside the unit cell is increased. The number of interatomic vectors, hence the number of peaks of the Patterson function, is increased. However the peak heights involving these splitted atoms are lower. If the atoms near the boundary surfaces, or edges, or corners of the unit cell are light atoms, these may help in resolving the peaks of the Patterson function (Sect. 2.2).

The method can be applied to a large number of crystals of known structures, including macromolecular crystals, to find the effectiveness of the inclusion of the in-between | Fourier coefficients | (Sect. 2.3).

The Patterson function of a crystal $p(\mathbf{r})$, (Eq. (5)), and the Patterson function of a unit cell $p_c(\mathbf{r})$, (Eq. (20)), are complementary to each other. $p(\mathbf{r})$ and $p_c(\mathbf{r})$, together, can be used for crystal atomic-coordinate determination.

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38

Sharpening of Patterson function of a unit cell.

Lowest Patterson function. Iterative algorithms with oversampling, and superresolution by extrapolation

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Abstract

Various sharpening of the Patterson function of a unit cell is used. The lowest Patterson function is obtained. A general method of sharpening is presented. We found that: The investigation of sharpening is for all crystals. The result can then be used for any specific crystal, as needed. We show an illustration employing Tchebycheff approximation. Shannon’s sampling and cardinal series are used. The effects of Fourier series truncation are due to truncation of the sharpened peak ${}^{s}G_{c}(\mathbf{r})$. The ripples are wiped out when all non-negative sharpening Patterson functions are used. To reduce the half-widths of the peaks, iterative algorithms with oversampling, and superresolution by extrapolation are employed. Using point atom-pairs, all the peaks of the Patterson function of a crystal may be resolved.

Keywords: Fourier transform; atomicity; autocorrelation; ripples; Shannon sampling; cardinal series; Tchebycheff approximation; Fourier series truncation; series termination; half-widths; superresolution by extrapolation; point atom-pair; oversampling.

1. Introduction

This is a sequel article of Yuen (2011d). It deals with the sharpening of the Patterson function of a unit cell.

In Sect. 2.1, various sharpening of the Patterson function of a unit cell is discussed, and the lowest Patterson function introduced. The problem of sharpening the Patterson function reduces to the problem of using all the sharpening functions such that the sharpened Patterson functions are non-negative.

In Sect. 2.2, a general method of sharpening is presented. We found that: The investigation of sharpening is for all crystals. The result can then be used for any specific crystal, as needed. We show an illustration employing Tchebycheff approximation. Shannon's sampling and cardinal series are used. The effects of Fourier series truncation are due to truncation of the sharpened peak ${}^{s}G_c(\mathbf{r})$. When all sharpened Patterson functions of the unit cell are used, and the lowest Patterson function obtained, the ripples are wiped out.

To reduce the half-widths of the peaks, iterative algorithms with oversampling, and superresolution by extrapolation are employed in Sect. 2.3. Using point atom-pairs, all the peaks of the Patterson function of a crystal may be resolved.

The peak at the origin and all known Patterson peaks may be removed (Sect. 2.4).

Waser & Schomaker (1953) presents an extensive treatment of modification functions.

2.1. Various sharpening of Patterson function of a unit cell. Lowest Patterson function

Use a unit cell as the object. The Patterson function of unit cell is given by the inverse Fourier transform of the diffraction intensity curve (Yuen (2011e), (14)).

Follow Yuen (2011d), Sect. 2.1. Assume that a very large number of reflections are obtained; very high resolution is obtained and the errors due to the Fourier series truncation upon the sharpened Patterson functions of a unit cell are negligible. Consider real atoms. Because of the atomicity, the Patterson function of a unit cell shows peaks at positions of the interatomic vectors with height approximately proportional to the product of the atomic numbers of the two atoms (or part of atom for a divided atom). This is the reason why the Patterson function of a unit cell is useful. The maximum width of a peak in the Patterson function is equal to the sum of the maximum widths of the two atoms involved. There are large number of Patterson peaks, and much overlapping. To sharpen the peaks, multiply the diffraction intensity curve by a sharpening function. As Yuen (2011d), Sect. 2.1, various sharpening functions can be used. In particular, use (atomic scattering factor of the heaviest atom)$^{-2}$, (atomic scattering factor of the heaviest atom × atomic factor of the second heaviest atom)$^{-1}$ (atomic scattering factor of the second heaviest atom)$^{-2}$, so on. This will sharpen the peaks in order, so that they can be recognized, and the atomic species identified. Use all sharpening functions such that the sharpened Patterson functions of a unit cell are non-negative. Superimpose all these Patterson function and sharpened Patterson functions of a unit cell. Insert all the peaks that we have recorded. At the other points of the unit cell, use the lowest value of these functions and obtain the lowest Patterson function of a unit cell. This is a sharpened Patterson function. See if there are any peaks not shown in the Patterson function or sharpened Patterson functions.

The problem of sharpening the Patterson function of a unit cell reduces to the problem of using all the sharpening functions such that the sharpened Patterson functions are non-negative.

2.2. General method of sharpening

To find all the sharpening functions of a unit cell, an analytical method is used. Consider a real or complex electron density $\rho(\mathbf{r})$. Form the autocorrelation

$$p_c(\mathbf{r}) = \rho(\mathbf{r}) \otimes \rho^*(-\mathbf{r}) \tag{1}$$

$$= \int \rho(\mathbf{s})\rho^*(-(\mathbf{r}-\mathbf{s}))d\mathbf{s} = \int \rho(\mathbf{r}+\mathbf{s})\rho^*(\mathbf{s})d\mathbf{s} \tag{2}$$

$\otimes$ denotes convolution. The integration is over one unit cell.

$p_c(\mathbf{r})$ is the autocorrelation of $\rho(\mathbf{r})$. $p_c(\mathbf{r})$ is the Patterson function of the unit cell.

$p_c(\mathbf{r})$ has size (support) in $[-1,1]$, $[-1,1]$, $[-1,1]$ (in fractional units).

Convolution is a smoothing operation; $p_c(\mathbf{r})$ tapers to zero at the boundary.

If $\rho(\mathbf{r})$ is real, from (2), $p_c(\mathbf{r})$ is real and centrosymmetric.

If $\rho(\mathbf{r})$ is non-negative, $p_c(\mathbf{r})$ is non-negative.

Let $P(\mathbf{h})$ and $F(\mathbf{h})$ be the Fourier transforms of $p_c(\mathbf{r})$ and $\rho(\mathbf{r})$ respectively.

$F(\mathbf{h})$ is the Fourier transform of a unit cell.

$$P(\mathbf{h}) = F(\mathbf{h})F^*(\mathbf{h}) = |F(\mathbf{h})|^2 \tag{3}$$

The Patterson function of a unit cell

$$p_c(\mathbf{r}) = \frac{1}{V}\int |F(\mathbf{h})|^2 \exp(-2\pi i\mathbf{h}\cdot\mathbf{r})d\mathbf{h} \tag{4}$$

V the volume of one unit cell. The factor V^{-1} is used as in Ladd & Palmer (1985), (6.54), (6.55).

Consider real atoms. Let Z_n and $f_n(\mathbf{h})$ be the atomic number and atomic scattering factor of the n-th atom respectively. The atomic scattering factors have approximately same shapes. Let

$$\hat{f}(\mathbf{h}) = (\sum f_n(\mathbf{h}))/(\sum Z_n) \tag{5}$$

$\hat{f}(\mathbf{h})$ is the atomic scattering factor of a hypothetical atom with $Z = 1$.

Let N be the number of atoms per unit cell.

An atom near a boundary surface, an edge or a corner of the unit cell is divided into parts. Each part is an atom which is not spherically symmetric. The Fourier transforms of $p_c(\mathbf{r})$ becomes

$$F(\mathbf{h}) = \sum_{n=1}^{N'} f_n'(\mathbf{h})\exp(2\pi i\mathbf{h}\cdot\mathbf{r}_n), \tag{6}$$

where $N' \geq N$,

$f_n'(\mathbf{h})$ is the atomic scattering factor of part of an atom if the atom is divided.

When the number of atoms in a unit cell is large, for example, macromolecular crystal, $N' \approx N$.

We use an approximation

$$f_n'(\mathbf{h}) \approx Z_n' \hat{f}(\mathbf{h}) \tag{7}$$

where Z_n' is for a part of the atom if the atom is divided. Z_n' may or may not be an integer. But for all the parts of a divided atom, the sum of the Z_n' is the atomic number of the atom.

Substitute (6) and (7) into (3).

$$P(\mathbf{h}) \approx \sum_{n=1}^{N'}\sum_{m=1}^{N'} Z_n' Z_m' \left|\hat{f}(\mathbf{h})\right|^2 \exp[2\pi i\mathbf{h}\cdot(\mathbf{r}_n - \mathbf{r}_m)] \tag{8}$$

Substitute (8) into (4). The Patterson function of the unit cell,

$$\begin{aligned} p_c(\mathbf{r}) &\approx \frac{1}{V}\sum_{n=1}^{N'}\sum_{m=1}^{N'} Z_n' Z_m' \int \left|\hat{f}(\mathbf{h})^2\right| \exp[-2\pi i\mathbf{h}\cdot(\mathbf{r}-(\mathbf{r}_n - \mathbf{r}_m))]d\mathbf{h} \\ &\approx \frac{1}{V}\sum_{n=1}^{N'}\sum_{m=1}^{N'} Z_n' Z_m' G_c[\mathbf{r}-(\mathbf{r}_n - \mathbf{r}_m)] \end{aligned} \tag{9}$$

where $G_c(\mathbf{r}) = \int \left|\hat{f}(\mathbf{h})\right|^2 \exp[-2\pi i\mathbf{h}\cdot\mathbf{r}]d\mathbf{h}$ (10)

$G_c(\mathbf{r})$ has the form of a peak at the origin. From (9), this peak is shifted to each point given by interatomic vectors $\mathbf{r}_n - \mathbf{r}_m$. The magnitude of the peak is modified by $Z_n' Z_m'$. This displacement gives rise to the Patterson peaks.

The right-hand side of (9) is an approximation. For an atom-pair near the boundary surfaces, or edges, or corners of the support of the Patterson function of the unit cell, $[-1,1]$, $[-1,1]$, $[-1,1]$, $G_c(\mathbf{r})$ will extend beyond this support. For macromolecular crystal, the effect is reduced.

If, in (6), the electrons not only form a cluster around the nuclei but also concentrate in an infinitesimal volume around each nucleus, this gives rise to a system of point atoms. Using (7), (6) becomes

$${}^{s}F(\mathbf{h}) = \frac{F(\mathbf{h})}{\hat{f}(\mathbf{h})} = \sum_n Z_n' \exp[2\pi i(\mathbf{h}\cdot\mathbf{r}_n)] \tag{11}$$

s stands for sharpened.

Replacing $|F(\mathbf{h})|^2$ in (4) by $|{}^sF(\mathbf{h})|^2$, we obtain

$$ {}^s p_c(\mathbf{r}) = \frac{1}{V}\sum_{n=1}^{N'}\sum_{m=1}^{N'} Z_n' Z_m' G_c[\mathbf{r} - (\mathbf{r}_n - \mathbf{r}_m)] \tag{12}$$

with $G_c(\mathbf{r}) = \int \exp[-2\pi i\mathbf{h}\cdot\mathbf{r}]d\mathbf{h}$ (13)

which represents a point atom-pair.

For a point-atom unit cell, there is no division of atoms near a boundary surface, an edge or a corner. The atom on a boundary surface, an edge or a corner, either belongs to one unit cell or another.

(12) becomes

$$ {}^s p_c(\mathbf{r}) = \frac{1}{V}\sum_{n=1}^{N}\sum_{m=1}^{N} Z_n Z_m G_c[\mathbf{r} - (\mathbf{r}_n - \mathbf{r}_m)] \tag{14}$$

with $G_c(\mathbf{r})$ given by (13).

We intend to transform (9) so that the shapes of the peaks are changed while the displacement property is maintained. To obtain the sharpening functions, we use a function $K(\mathbf{r})$ which has same support as $p_c(\mathbf{r})$ (i.e. $[-1,1]$, $[-1,1]$, $[-1,1]$).

Consider a transform of $p_c(\mathbf{r})$:

Let ${}^s p_c(\mathbf{r}) = p_c(\mathbf{r}) \otimes K(\mathbf{r})$ (15)

$$= \int p_c(\mathbf{s})K(\mathbf{r}-\mathbf{s})d\mathbf{s} \tag{16}$$

${}^s p_c(\mathbf{r})$ has size (support) $[-2,2]$, $[-2,2]$, $[-2,2]$ (in fractional units).

Substitute (9) into (16),

$$ {}^s p_c(\mathbf{r}) \approx \frac{1}{V}\sum_{n=1}^{N'}\sum_{m=1}^{N'} Z_n' Z_m' {}^s G_c[\mathbf{r} - (\mathbf{r}_m - \mathbf{r}_n)] \tag{17}$$

where ${}^s G_c(\mathbf{r}) = G_c(\mathbf{r}) \otimes K(\mathbf{r})$

$$= \int G_c(\mathbf{s}) K(\mathbf{r} - \mathbf{s}) d\mathbf{s} \tag{18}$$

The integration is over the region $[-2,2]$, $[-2,2]$, $[-2,2]$.

(17) has exactly the displacement property exhibited in (9). It has sharpened peak ${}^sG_c(\mathbf{r})$ (see (18)) at interatomic vectors $\mathbf{r}_m - \mathbf{r}_n$. ${}^s p_c(\mathbf{r})$ is a sharpened Patterson function of the unit cell.

The right-hand side of (17) is an approximation. For an atom-pair near the boundary surfaces, or edges, or corners of the support of the sharpened Patterson function of the unit cell, $[-2,2]$, $[-2,2]$, $[-2,2]$, ${}^sG_c(\mathbf{r})$ will extend beyond this support. For macromolecular crystal, the effect is reduced.

Express $K(\mathbf{r})$ as the inverse Fourier transform of a function $q(\mathbf{h})$,

$$K(\mathbf{r}) = \frac{1}{4^3} \int q(\mathbf{h}) \exp(-2\pi i \mathbf{h} \cdot \mathbf{r}) d\mathbf{h} \tag{19}$$

$q(\mathbf{h})$ is band-limited.

Substitute (10) and (19) into (18),

$${}^sG_c(\mathbf{r}) = \int \left| \hat{f}(\mathbf{h}) \right|^2 q(\mathbf{h}) \exp(-2\pi i \mathbf{h} \cdot \mathbf{r}) d\mathbf{h} \tag{20}$$

$\left| \hat{f}(\mathbf{h}) \right|^2 q(\mathbf{h})$ is band-limited. ${}^sG_c(\mathbf{r})$ has size (support) $[-2,2]$, $[-2,2]$, $[-2,2]$ (in fractional units).

Compare (20) with (10), we see that the net result of the transformation (16) is the introduction of the coefficient $q(\mathbf{h})$ into (20). This coefficient can be adjusted so that ${}^sG_c(\mathbf{r})$ is a peak of satisfactory shape, while the displacement property is maintained (see (17)).

Conversely, if, from the beginning, we modify the Patterson function of a unit cell (4) by defining

$$^{s}p_c(\mathbf{r}) = \frac{1}{64V}\int |F(\mathbf{h})|^2 q(\mathbf{h})\exp(-2\pi i\mathbf{h}\cdot\mathbf{r})d\mathbf{h} \qquad (21)$$

From (6) and (7),

$$F(\mathbf{h}) \approx \sum_{n=1}^{N'} Z_n{}'\hat{f}(\mathbf{h})\exp(2\pi i\mathbf{h}\cdot\mathbf{r}_n) \qquad (22)$$

Substitute (22) into (21), we obtain (17) with $^{s}G_c(\mathbf{r})$ given by (20). Hence, the displacement property is maintained when coefficient $q(\mathbf{h})$ are introduced into the Patterson function (see (21)). Moreover, (16) is obtained (see Waser & Schomaker (1953), (3-8)).

For the sharpened peak $^{s}G_c(\mathbf{r})$ in (20) and the sharpened Patterson function $^{s}p_c(\mathbf{r})$ in (17) to be real, the coefficient $q(\mathbf{h}) = [q(-\mathbf{h})]^*$. (23)

From (21), the Fourier transform of the sharpened Patterson function of a unit cell

$$^{s}p_c(\mathbf{r}) = \frac{1}{64V}|f(\mathbf{h})|^2 q(\mathbf{h})$$

In general, $q(\mathbf{h})$ is complex. Hence the Fourier transform of the sharpened Patterson function of a unit cell is complex. This is different from the Fourier transform of the Patterson function of a unit cell, which is non-negative (Eq. (3)). Hence we have the conclusion: For a sharpened Patterson function of a unit cell to be the Patterson function of an electron density distribution of the unit cell, $q(\mathbf{h})$ must be non-negative.

If $q(\mathbf{h}) = 1/[\hat{f}(\mathbf{h})]^2$, (21) becomes

$$^{s}p_c(\mathbf{r}) = \frac{1}{V}\int \frac{|F(\mathbf{h})|^2}{[\hat{f}(\mathbf{h})]^2}\exp(-2\pi i\mathbf{h}\cdot\mathbf{r})d\mathbf{h} \qquad (24)$$

This is the sharpened Patterson function of point-atom pairs obtained by substituting (11) into (4). The factor $1/64$ is ignored as it comes from the support $[-2,2]$,

$[-2,2]$, $[-2,2]$ of ${}^{s}p_c(\mathbf{r})$ in (16). For point-atom pairs, the support of ${}^{s}p_c(\mathbf{r})$ reduces to $[-1,1]$, $[-1,1]$, $[-1,1]$.

Choose $q(\mathbf{h}) = c(\mathbf{h})/[\hat{f}(\mathbf{h})]^2$. (25)

(20) becomes

$$ {}^{s}G_c(\mathbf{r}) = \int c(\mathbf{h}) \exp(-2\pi i \mathbf{h}\cdot\mathbf{r}) d\mathbf{h} \tag{26}$$

$c(\mathbf{h})$ is band-limited.

This inverse Fourier transform gives the most general sharpened peak ${}^{s}G_c(\mathbf{r})$, and hence the most general sharpening Patterson function ${}^{s}p_c(\mathbf{r})$ in (17). ${}^{s}G_c(\mathbf{r})$ is for all crystals. Here we have an important conclusion of this article: **Investigation of sharpening is for all crystals. The result can then be used for any specific crystal, as needed.**

Substitute (25) into (21).

$$ {}^{s}p_c(\mathbf{r}) = \frac{1}{64V} \int \frac{|F(\mathbf{h})|^2}{[\hat{f}(\mathbf{h})]^2} c(\mathbf{h}) \exp(-2\pi i \mathbf{h}\cdot\mathbf{r}) d\mathbf{h} \tag{27}$$

$\dfrac{|f(\mathbf{h})|^2}{[f(\mathbf{h})]^2} c(\mathbf{h})$ is band-limited.

The coefficient $c(\mathbf{h})$ in (26) are chosen to give the desired sharpened peak ${}^{s}G_c(\mathbf{r})$. With this coefficient $c(\mathbf{h})$ and the experimentally determined $|F(\mathbf{h})|^2$, Eq. (27) is used to obtain the sharpened Patterson function of a unit cell ${}^{s}p_c(\mathbf{r})$. In (27), instead of the sharpening function $[\hat{f}(\mathbf{h})]^{-2}$, other sharpening functions in Yuen (2011d), Sect. 2.1 can also be used. In particular, starting from the heaviest atoms, we use product of their atomic scattering factors as sharpening functions.

Now consider a non-negative electron density $\rho(\mathbf{r})$. From (2), the Patterson function of the unit cell is non-negative. Consider the case of finite number of diffraction intensities, limited by experimental measurement. There is Fourier series truncation error. There will be large negative and positive ripples in the Patterson function.

We will discuss the method of obtaining the coefficients $c(\mathbf{h})$ in (26) in one-dimension. In (26), $c(h)$ is band-limited. The inverse Fourier transform of $c(h)$ has support $[-2,2]$. Use the Shannon's sampling theory (cardinal series, Marks (1991), p. 1),

$$c(h) = \sum_{m=-\infty}^{\infty} c(m/4) \sin c(4h - m) \tag{28}$$

The $\sin c$ function is defined as $\sin c(x) = \frac{\sin(\pi x)}{\pi x}$.

Substitute (28) into (26). Similar to Yuen (2011e), (19),

$${}^{s}G_{c}(x) = \frac{1}{4} \sum_{m=-\infty}^{\infty} c(m/4) \exp(-\pi i m x/2) \Pi(x/4) \tag{29}$$

Let $Q(x)$ be a real sharpening function in $[-2,2]$ (in fractional unit). $Q(x)=0$ for $x \notin [-2,2]$. We would like to choose the coefficient $c(m/4)$ such that ${}^{s}G_{c}(x)$ in (26) is a good approximation to $Q(x)$. Yuen (2011d), Sect. 2.2 will be followed. The degree of approximation is defined, according to the L_p -norm. As an example, we will use the Tchebycheff approximation.

We will use a finite sum as an approximation of the integral in (26). From (29),

$${}^{s}G_{c}(x) = \frac{1}{4} \sum_{m=-H}^{H} c(m/4) \exp(-\pi i m x/2) \Pi(x/4) \tag{30}$$

$Q(x)$ is real. Hence $c(m/4) = [c(-m/4)]^{*}$.

For centrosymmetric real $Q(x)$, $c(m/4)$ is real and $c(m/4) = c(-m/4)$.

(31)

$$ {}^{s}G_c(x) = \frac{1}{4}c(0)\Pi(x/4) + \frac{1}{2}\sum_{m=1}^{H} c(m/4)\cos(\pi m x/2)\Pi(x/4) \qquad (32) $$

For the Tchebycheff approximation of a given real function over a discrete range by a linear combination of M linearly independent continuous real functions, IBM has a subroutine for calculating the M coefficients.

We show three examples of $Q(x)$:

Consider the interval $[-2,2]$ of x.

(A) $Q(x)$ =100 at $x = 0$.

$Q(x) = 0$ at $x \varepsilon [-\sigma, \sigma]$, but $x \neq 0$. σ is a positive number < 1.

$Q(x) = 0$ at $x = -2$ or 2

$Q(x)$ =0 for $x \notin [-2,2]$.

$Q(x)$ is undefined at other points of x. This is to give flexibility to the approximating function ${}^{s}G_c(x)$.

The purpose of this choice of $Q(x)$ is to have sharp peak at the point $x = 0$.

To approximate $Q(x)$ by ${}^{s}G_c(x)$ in (32), apply the subroutine of IBM. The coefficients $c(0)$ and $c(m/4)$ are obtained. Hence ${}^{s}G_c(x)$ is obtained. Supremum of $\left|{}^{s}G_c(x) - Q(x)\right|$ for $x \in [-2,2]$ is obtained.

In Sect. 2.1, the sharpened Patterson function used is non-negative. Hence we set all the negative value of ${}^{s}G_c(x)$ to zero, and obtain a non-negative sharpened peak ${}^{s}\overline{G}_c(x)$. ${}^{s}\overline{G}_c(x)$ has intervals of zeros.

For a specific crystal, the non-negative sharpened function ${}^{s}\overline{p}_c(x)$ is obtained from

(27).

$\frac{|F(h)|^2}{[\hat{f}(h)]^2}c(h)$ is band-limited.

As (28), Shannon sampling is used. From (30),

$$ {}^{s}\overline{p}_c(x) = \frac{1}{4V}\sum_{m=-H}^{H}\frac{|F(m/4)|^2}{[\hat{f}(m/4)]^2}c(m/4)\exp(-\pi imx/2)\Pi(x/4) \qquad (33)$$

For m = 4 × an integer, $F(m/4)$ are the Fourier coefficients, say $F(n)$.

For $m \neq$ 4 × an integer, $|F(m/4)|^2$ can be obtained by interpolation of | Fourier coefficients | , $|F(n)|$. The interpolated $|F(m/4)|$ is then squared. We can also obtain $|F(m/4)|^2$ by interpolation of | Fourier coefficients | ^{2}, $|F(n)|^2$. From these two results, a more reliable value is obtained.

For different values of σ, the ripple heights and positions change. For all the non-negative sharpened Patterson functions of the unit cell ${}^{s}\overline{p}_c(x)$, obtain the lowest Patterson function as in Sect. 2.1. **The shifted zeros of all** ${}^{s}\overline{G}_c(x)$ **wipe out all the ripples.**

(B) $Q(x)$ is a triangle at the origin, i.e., for $x\varepsilon[-\sigma,\sigma]$.
$Q(x)$ =0 for $x \notin [-2,2]$.
$Q(x)$ is undefined at other points of x.

(C) Unsymmetrical $Q(x)$.
$Q(x) = 0$ at $x\varepsilon[-0.2,0)$.
$Q(x) = 100$ at $x = 0$

$Q(x)=0$ for $x \notin [-2,2]$.

$Q(x)$ is undefined at other points of x.

The purpose of this choice of $Q(x)$ is to force ${}^{s}G_c(x)$ to have a peak very sharp at one side, at the points $x=0$. This may help in resolving the peaks.

Use all reasonable shapes of $Q(x)$, and obtain the best L_p-approximation to $Q(x)$ for all $p \geq 1$ (see Yuen (2011d), (25)).

For three-dimensional crystals, approximation in three dimensions, or product of three one-dimensional approximating functions for the three axes may be used.

Corresponding to (28),

$$c(\mathbf{h}) = \sum_{m_1=-\infty}^{\infty} \sum_{m_2=-\infty}^{\infty} \sum_{m_3=\infty}^{\infty} c(m_1/4, m_2/4, m_3/4) \sin c(4h_1 - m_1) \sin c(4h_2 - m_2) \cdot \sin c(4h_3 - m_3) \tag{34}$$

Corresponding to (29) and similar to Yuen (2011e), (20),

$${}^{s}G_c(\mathbf{r}) = \frac{1}{64} \sum_{m_1,m_2,m_3} \{c(m_1/4, m_2/4, m_3/4) \exp[-\pi i(m_1 x + m_2 y + m_3 z)/2] \cdot \Pi(x/4)\Pi(y/4)\Pi(z/4)\} \tag{35}$$

The three-dimensional equation corresponding to (30),

$${}^{s}G_c(\mathbf{r}) = \frac{1}{64} \sum_{m_1=-H_1}^{H_1} \sum_{m_2=-H_2}^{H_2} \sum_{m_3=-H_3}^{H_3} \{c(m_1/4, m_2/4, m_3/4) \cdot \exp[-\pi i(m_1 x + m_2 y + m_3 z)/2]\Pi(x/4)\Pi(y/4)\Pi(z/4)\} \tag{36}$$

For all $Q(\mathbf{r})$, and apply the subroutine of IBM. Determine the coefficients $c(m_1/4, m_2/4, m_3/4)$ in (36). ${}^{s}G_c(\mathbf{r})$ is obtained. Obtain supremum of

$\left|{}^{s}G_c(\mathbf{r}) - Q(\mathbf{r})\right|$, $\mathbf{r} \in ([-2,2],\ [-2,2],\ [-2,2])$. In Sect. 2.1, the sharpened Patterson function used is non-negative. Hence we set all the negative value of ${}^{s}G_c(\mathbf{r})$ to zero, and obtain a non-negative sharpened peak ${}^{s}\overline{G_c}(\mathbf{r})$. ${}^{s}\overline{G_c}(\mathbf{r})$ has regions of zeros. As these are for all crystals (see (26)), the results may be tabulated.

For a specific crystal, the non-negative sharpened Patterson function ${}^{s}\overline{p}_c(\mathbf{r})$ is obtained in (27), employing the experimentally determined $\left|F(\mathbf{h})\right|^2$. Corresponding to (33),

$$
\begin{aligned}
{}^{s}\overline{p}_c(\mathbf{r}) = \frac{1}{64V} \sum_{m_1=-H_1}^{H_1} \sum_{m_2=H_2}^{H_2} \sum_{m_3=H_3}^{H_3} \Bigg\{ & \frac{\left|F(m_1/4, m_2/4, m_3/4)\right|^2}{[\hat{f}(m_1/4, m_2/4, m_3/4)]^2} \cdot \\
& c(m_1/4, m_2/4, m_3/4) \cdot \\
& \exp[-\pi i(m_1 x + m_2 y + m_3 z)/2] \Pi(x/4) \Pi(y/4) \Pi(z/4) \Bigg\}
\end{aligned}
\tag{37}
$$

$\left|F(m_1/4, m_2/4, m_3/4)\right|^2$ can be obtained by interpolation of | Fourier coefficients | in multidimensions. The interpolated $\left|F(m_1/4, m_2/4, m_3/4)\right|$ is then squared. We can also obtain $\left|F(m_1/4, m_2/4, m_3/4)\right|^2$ by interpolation of | Fourier coefficients | ^{2} in multidimensions. From these two results, a more reliable value is obtained.

For all the non-negative sharpened Patterson functions ${}^{s}\overline{p}_c(\mathbf{r})$, as in Sect. 2.1, multiply each sharpened Patterson function by a constant so that the height of the peak at the origin is the same as that of the Patterson function. Superimpose all these Patterson function and sharpened Patterson functions. Insert all the peaks in the

Patterson function and each sharpened Patterson function. At the other points of the unit cell, use the lowest value of these functions. From this resultant function, see if there are any more peaks not shown in the Patterson function or sharpened Patterson functions. We obtain a lowest Patterson function of the unit cell. The ripples are wiped out by the zeros of all the non-negative sharpened Patterson functions.

The lowest Patterson function of a unit cell can be compared with the lowest Patterson function from the Patterson function of a unit cell (Yuen (2011d), Sect. 2.1, 2.2), to see if any more peaks are revealed.

2.3. Fourier series truncation. Iterative algorithm with oversampling and superresolution by extrapolation

Only a finite number of terms of the diffraction intensity of a crystal are measured. The effects due to Fourier series truncation (termination) are large peak half-widths and large negative and positive ripples. The ripples are due to the ripples of the sharpened peak ${}^{s}G_c(\mathbf{r})$. The effects of series truncation are due to truncation of the sharpened peak ${}^{s}G_c(\mathbf{r})$. Eq. (30) is an example. When all the non-negative sharpened Patterson functions are used and the lowest Patterson function obtained, the ripples are wiped out (Sect. 2.1, 2.2). Hence the remaining problem is the large half-widths of the non-negative sharpened peak ${}^{s}\overline{G_c}(\mathbf{r})$. To amend for this Fourier series truncation error, follow Yuen (2011d), Sect. 2.3. Start from the lowest Patterson function of the unit cell and apply iterative algorithms with oversampling. Superresolution by extrapolation is used. There is no initial phase problem.

In the reciprocal space, the $|F(m_1/4, m_2/4, m_3/4)|^2$, observed or obtained by interpolation in multidimensions, multiplied by $[\,\hat{f}(\mathbf{h})\,]^{-2}$ will be used as the constraint. The oversampling ratio = 4^3 = 64. (38)

Use point atom-pairs. Neglect the effect of atoms near the boundary surface, an edge or a corner of the unit cell, as in macromolecular crystal.

Apply the iterative algorithms with oversampling. All the peaks of the Patterson function of the unit cell may be resolved.

Result of sharpening the Patterson function of the crystal (Yuen (2011d) can be used to monitor the iterations in this section.

2.4. Removal of the peak at the origin and known Patterson peaks

One variance is to remove the peak at the origin of the Patterson function (similar to Buerger (1959), p. 56-58, Lipson & Cochran (1966), p. 169-170),

$$p_c(\mathbf{r}) = \frac{1}{V}\int \left\{ \left|F(\mathbf{h})\right|^2 - \sum_{n=1}^{N'} [f_n'(\mathbf{h})]^2 \right\} \exp(-2\pi i\mathbf{h}\cdot\mathbf{r})d\mathbf{h} \tag{39}$$

$p_c(\mathbf{r})$ has size (support) in $[-1,1]$, $[-1,1]$, $[-1,1]$. Use the Shannon's sampling theory (similar to Yuen (2011e), (20)).

$$\begin{aligned} p_c(\mathbf{r}) = \frac{1}{8V} \sum_{m_1,m_2,m_3} \Big\{ \Big[& \left|F(m_1/2, m_2/2, m_3/2)\right|^2 \\ & - \sum_{n=1}^{N'} [f_n'(m_1/2, m_2/2, m_3/2)]^2 \Big] \cdot \\ & \cos[\pi(m_1x + m_2y + m_3z)] \Pi(x/2)\Pi(y/2)\Pi(z/2) \Big\} \end{aligned} \tag{40}$$

Then apply Sect. 2.1 - 2.2 to this expression. Since the large peak in the origin has been removed, multiply each sharpened Patterson function of the unit cell by a constant so that the height of the highest peaks is the same as those of the Patterson function. Superimpose all these Patterson function and sharpened Patterson functions. As in Sect. 2.1, obtain a lowest Patterson function of the unit cell.

When a pair of Patterson peaks from the j-th and n-th atoms is obtained, one variance is to remove these peaks from the Patterson function (similar to Buerger (1959), p.58-59).

$$p_c(\mathbf{r}) = \frac{1}{V}\int \left\{ |F(\mathbf{h})|^2 - 2 f_j'(\mathbf{h}) f_n'(\mathbf{h}) \cos[2\pi\mathbf{h}\cdot(\mathbf{r}_j - \mathbf{r}_n)] \right\} \exp(-2\pi i\mathbf{h}\cdot\mathbf{r}) d\mathbf{h} \tag{41}$$

Use the Shannon's sampling theory

$$\begin{aligned} p_c(\mathbf{r}) = \frac{1}{8V} \sum_{m_1,m_2,m_3} \Big\{ \Big[& |F(m_1/2, m_2/2, m_3/2)|^2 \\ & - 2 f_j'(m_1/2, m_2/2, m_3/2) f_n'(m_1/2, m_2/2, m_3/2) \cdot \\ & \cos 2\pi[m_1(x_j - x_n) + m_2(y_j - y_n) + m_3(z_j - z_n)] \Big] \cdot \\ & \cos[\pi(m_1 x + m_2 y + m_3 z)] \Pi(x/2)\Pi(y/2)\Pi(z/2) \Big\} \end{aligned} \tag{42}$$

Remove all the known pairs of Patterson peaks.

Then apply Sect. 2.1 - 2.2 to the final expression obtained. A lowest Patterson function is obtained.

One variance is to remove the peak at the origin and the known pair of Patterson peaks by combining expressions (39) and (41).

$$\begin{aligned} p_c(\mathbf{r}) = \frac{1}{V}\int & \left\{ |F(\mathbf{h})|^2 - \sum_{n=1}^{N'} [f_n'(\mathbf{h})]^2 - 2 f_j'(\mathbf{h}) f_n'(\mathbf{h}) \cos[2\pi\mathbf{h}\cdot(\mathbf{r}_j - \mathbf{r}_n)] \right\} \cdot \\ & \exp(-2\pi i\mathbf{h}\cdot\mathbf{r}) d\mathbf{h} \end{aligned}$$

Use the Shannon's sampling theory by combining expressions (40) and (42).

$$\begin{aligned} p_c(\mathbf{r}) = \frac{1}{8V} \sum_{m_1,m_2,m_3} \Big\{ \Big[& |F(m_1/2, m_2/2, m_3/2)|^2 \\ & - \sum_{n=1}^{N'} [f_n'(m_1/2, m_2/2, m_3/2)]^2 \\ & - 2 f_j'(m_1/2, m_2/2, m_3/2) f_n'(m_1/2, m_2/2, m_3/2) \cdot \\ & \cos 2\pi[m_1(x_j - x_n) + m_2(y_j - y_n) + m_3(z_j - z_n)] \Big] \cdot \\ & \cos[\pi(m_1 x + m_2 y + m_3 z)] \Pi(x/2)\Pi(y/2)\Pi(z/2) \Big\} \end{aligned}$$

Remove all the known pairs of Patterson peaks.

Then apply Sect. 2.1 - 2.2 to the final expression obtained. Use the height of the highest peaks. A lowest Patterson function is obtained.

Add the peaks obtained in these variances to the lowest Patterson function obtained in Sect. 2.2. Then apply Sect. 2.3 to the resultant lowest Patterson function of the unit cell.

2.5. Demonstrations with crystal structures reported in the literature

Choose a large number of crystal structures reported in the literature, of various sizes, small, mediate, or macromolecular, and with or without heavy atoms. As in Yuen (2011e), Sect. 2.3, the calculated Fourier transforms of a unit cell at $(m_1/4, m_2/4, m_3/4)$ are good for determination of atomic coordinates, including the Patterson peaks. Apply Sect. 2.1, 2.2 and 2.3. Find all non-negative sharpening functions. Obtain the lowest Patterson function. Examine application of iterative algorithm with oversampling, and superresolution by extrapolation to deal with the problem of Fourier series truncation. Examine the result of resolving all Patterson peaks. The result of division of atoms near a boundary surface, an edge or a corner, can be examined.

3. Discussions and conclusion

Sect 2.2 presents an analytical method of sharpening the Patterson function of a unit cell. We have an important conclusion: Investigation of sharpening is for all crystals. The result can then be used for any specific crystal, as needed.

The sharpened peak ${}^{s}G_c(\mathbf{r})$ (see (20)) and the sharpened Patterson function of the unit cell ${}^{s}p_c(\mathbf{r})$ (see (15)) have size (support) $[-2,2]$, $[-2,2]$, $[-2,2]$ (in fractional units). The Patterson peaks are centered in the region, $[-1,1]$, $[-1,1]$, $[-1,1]$. The sharpened peak ${}^{s}G_c(\mathbf{r})$ has a sharp peak at the origin with ripples. It is reasonable to expect that the sharpened Patterson function of the unit cell ${}^{s}p_c(\mathbf{r})$ has essentially a support slightly larger than the region, $[-1,1]$, $[-1,1]$, $[-1,1]$ with ripples outside this region. When all the non-negative sharpened Patterson functions are used and the lowest Patterson function obtained, the ripples are wiped out (Sect. 2.1, 2.2).

Sect. 2.3 then deals with the effect due to Fourier series truncation: large peak half-widths. Iterative algorithms with oversampling is employed. We start from the lowest Patterson function of a unit cell, and apply iterative algorithms. There is no initial phase problem.

Number of Fourier transforms at $(m_1/4, m_2/4, m_3/4)$ is 4^3 = 64-times the number of Fourier coefficients. For the iterative algorithms, this provides more accurate results for extrapolation. The oversampling ratio 4^3 = 64 is larger than the oversampling ratio $2^3 = 8$ used in articles in above, for examples, Yuen (2011a, 2011b, 2011c). Zayed (1993), p. 22 states that oversampling greater than the Nyquist rate can reduce interpolation noise level due to noisy data, the sampled values are dependent and the signal reconstruction is not affected by losing an arbitrarily large but finite number of samples values (Marks (1991), Sect. 4.1.1, 5,1,1). (Along one axis, oversampling ratio corresponding to the Nyquist rate of sampling is 2).

Using point atom-pairs, all the peaks of the Patterson function of a unit cell may be resolved.

In Yuen (2011c), we have *ab initio* determination of crystal electron density and atomic coordinates via point-atom model. Here we only need the positions of the Patterson peaks or point atom-pairs, hence it is reasonable to expect that iterative algorithms with oversampling will be able to determine the positions of these peaks; superresolution by extrapolation will work.

Because of (7), the treatment presented is an approximation.

The peak at the origin and all known Patterson peaks may be removed (Sect. 2.4). As the Fourier transform expressed in (6) is an approximation, the removal of the peaks is not complete.

The sharpening method can be applied to a large number of crystals of known structures, including macromolecular crystals, to find the effectiveness of the method and to improve the method (Sect. 2.5).

The sharpening of the Patterson function of a crystal (Yuen (2011d)), and the sharpening of the Patterson function of a unit cell are complementary to each other. These, together, can be used for crystal atomic-coordinate determination.

Caliandro et al. (2008) has determined the structure of a crystal of 7890 non-H atoms by means of Patterson method. With the sharpening of the Patterson functions of a crystal, and of a unit cell, crystals atomic coordinates of more atoms may be determined.

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39

Dual-real-spaces iterative algorithms with oversampling applied to electron density function and Patterson function of a unit cell

July 8, 2011

Yuen (2011c), dual-real-spaces iterative algorithms applied to electron density function and Patterson function of a crystal is adopted, except that this time, we use Patterson functions of a unit cell (Yuen (2011d), Sect. 2.2) and sharpening of Patterson function of a unit cell (Yuen (2011e), Sect. 2.1, 2.2).

Fourier transform is used.

Oversampling is used (Yuen (2011a, 2011b). The oversampling ratio = $4^3 = 64$.

In Yuen (2011b), we have *ab initio* determination of crystal electron density and atomic coordinates via point-atom model. Here we only need to determine the crystal atomic coordinates, hence it is reasonable to expect that dual-real-spaces iterative algorithms with large oversampling ratio, applied to electron density function and Patterson function of a unit cell will be able to determine these coordinates.

Dual-real-spaces iterative algorithms applied to electron density function and Patterson function of a crystal (Yuen (2011c)), and dual-real-spaces iterative algorithms with oversampling applied to electron density function and Patterson function of a unit cell are complementary to each other. These, together, can be used for crystal atomic-coordinate determination.

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40

Sayre’s equation and tangent formula for equal-atom crystal. Extension to include the in-between Fourier coefficients

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Abstract

This article extends the Sayre’s equation to include the in-between Fourier coefficients. The in-between Fourier coefficients are included by dealing with a unit cell. The result is $F(\mathbf{h}) = \dfrac{f(\mathbf{h})}{f^{sq}(\mathbf{h})V}\sum_{\mathbf{m}} F(\mathbf{m}/2)F(\mathbf{h}-(\mathbf{m}/2))$. The form of this equation resembles the form of the Sayre’s equation for crystal. The tangent formula,

$$\tan\varphi_{\mathbf{h}} = \frac{\sum_{\mathbf{m}} |F(\mathbf{m}/2)||F(\mathbf{h}-(\mathbf{m}/2))|\sin(\varphi_{\mathbf{m}/2}+\varphi_{\mathbf{h}-(\mathbf{m}/2)})}{\sum_{\mathbf{m}} |F(\mathbf{m}/2)||F(\mathbf{h}-(\mathbf{m}/2))|\cos(\varphi_{\mathbf{m}/2}+\varphi_{\mathbf{h}-(\mathbf{m}/2)})}.$$

The equation obtained and the Sayre’s equation of crystal are complementary to each other. They are separate conditions; one cannot replace the other. The equation obtained, the Sayre’s equation of crystal and the tangent formulae, together, can be used for crystal atomic-coordinate determination.

Keywords: Point-atom; in-between structure factors; Fourier transform; Shannon sampling; cardinal series.

1. Introduction

Squaring the magnitude of the Fourier coefficients gives the important and useful

Patterson function. How about if the electron density is squared? Sayre (1952) presents an ingenious squaring method for phase determination, which employs like atoms, and obtains the important and useful Sayre's equation (see also Rothbauer (1980)).

Woofson (1958a, 1958b) present an equation between structure factors for structures containing unequal or overlapped atoms. Barrett & Zwick (1971) presents a method for the extension and refinement of crystallographic protein phases. Krabbendam & Kroon (1971) presents an algorithm for phase refinement with the aid of Sayre's equation. Sayre (1972, 1974) presents a least-squares phase refinement employing the Sayre's equation. Rothbauer (1976, 1977) presents structure-factor equations. Lunin (1985) presents fast differentiation algorithm and Sayre's equation. Harrison (1987) shows that Sayre's equation is a Chernov bound to maximum entropy. Hou et al. (1989) presents a trial and error method for crystal structure determination. Main (1990) and Zhang & Main (1990) discuss the use of Sayre's equation with constraints in real space. Cowtan (1991) presents a procedure for phase refinement and extension. Fan (1991) presents direct methods for modulated structures. Mishnev (1991) provides physical interpretation of Sayre's equation. Woolfson (1991a, 1991b) presents discussions. Cowtan & Main (1993) presents simultaneous application of real and reciprocal space constraints. Fan et al. (1993), Sha et al. (1994) and de Gelder et al. (1996) present direct methods for incommensurate intergrowth compounds. Lunin (1993) uses electron-density histograms. Zhang (1993) combines constraints for macromolecular phase refinement and extension. Sato (1994) and Refaat et al. (1995) present direct-space methods using Sayre's equation. Sato (1994) discusses use of the tangent formula. Dorset et al. (1995) discusses the Sayre equation in electron crystallography. Tivol (1995) discusses solution of the phase problem and Sayre equation. Schuller (1996) presents *MAGICSQUASH*. Chen & Su (2000) solves the Sayre equation by simulated annealing. Roach et al. (2001) uses Sayre-type equation for complex electron density. Use of Sayre equation for binary images is discussed in the review of Thumiger & Zanotti (2009).

Sayre-equation tangent formula and SAYTAN are presented in Debaerdemaeker et al. (1985, 1988). Woolfson & Yao (1990) presents an *ab initio* solution of a small protein.

Shiono & Woolfson (1991) discusses properties and limitations of Sayre's equation. A detailed treatment of *SAYTAN* is presented in Tate (1991). Woolfson (1991c) presents discussion of tangent formula. Mukherjee & Woolfson (1993) investigates a small protein at low resolution. Woolfson (1993) investigates the problems with large structures and low resolution. Hubbard et al. (1994) applies *SAYTAN* to three oligonucleotides. Mishnev & Woolfson (1994) presents a new figure of merit for protein phase sets at moderate resolution. Mukherjee & Woolfson (1995) applies *SAYTAN* to 2-Zn insulin. Mo et al. (1996) uses known phases from three-beam diffraction experiments to re-determine the crystal structure of rubredoxin. Mukherjee (1999) presents an a*b initio* structure determination of rubredoxin. Guo et al. (2000) presents bulk-solvent-corrected Sayre equation.

Woolfson (1987) presents a review of direct methods. Sayre equation, tangent formula and *SAYTAN* are discussed in Woolfson & Fan (1995), Sect. 3.4.1, 3.5.5, 7.5.1, 7.6.1, 7.6.3 and Fan (1998)). Giacovazzo (1998), Sect. 4.3, 4.4, 14.5, 14.6 presents discussions of Sayre's equation. Sayre (1980) presents a general discussion of phase extension and refinement using convolutional equation systems.

This article extends the Sayre's equation for equal-atom crystal (Sect. 2.1, 2.2) to include the in-between Fourier coefficients (in-between structure factors). The in-between Fourier coefficients are included by dealing with a unit cell. Equal point-atoms, and equal atoms with atomic scattering factors are dealt with in Sect. 2.3 and 2.4 respectively. The equations obtained are exact equations. The form of the equations obtained resembles the forms of the Sayre's equations for crystal. The equations obtained and the Sayre's equations for crystal are complementary to each other.

2.1. Sayre's equation for equal-point-atom crystal

Consider a crystal of equal point-atoms (Yuen (2011c), (7)).

$$F'(\mathbf{h}) = F(\mathbf{h}) / f(\mathbf{h}) = \sum \delta(\mathbf{r}' - \mathbf{r}_j) \exp[2\pi i(\mathbf{h} \cdot \mathbf{r}_j)] \tag{1}$$

A point atom is represented by a point with value H centered at a sphere with radius R. The value of the atom in this sphere, except the center, is equal to zero. Let the atoms be real and $H = 1$. The crystal has N unit cells. N is finite. The electron density of the crystal is $\rho(\mathbf{r})$. To obtain the Sayre's equation, we first consider equal-point-atom crystal.

The Fourier transform of the crystal of N unit cells,

$$F_N(\mathbf{h}) = V \int_{NV} \rho(\mathbf{r}) \exp(2\pi i \mathbf{h} \cdot \mathbf{r}) d\mathbf{r} \tag{2}$$

where V is the volume of one unit cell. NV is the volume of the crystal. The components of $\mathbf{h}$ may or may not be integers. The factor V is used in (2) because the electron density is in electron per unit volume, while $\mathbf{r}$ is in fractional coordinates.

Now consider the squared crystal which consists of the square of the electron density, $\rho^2(\mathbf{r})$. Since $\rho(\mathbf{r})$ consists of real point atoms of value = 1, the crystal is identical to the squared crystal.

$$\rho^2(\mathbf{r}) = \rho(\mathbf{r}) = 1$$

The Fourier transform of the squared crystal of N unit cells,

$$F_N(\mathbf{h}) = V \int_{NV} \rho^2(\mathbf{r}) \exp(2\pi i \mathbf{h} \cdot \mathbf{r}) d\mathbf{r}$$

$$= V \left[\int_{NV} \rho(\mathbf{r}) \exp(2\pi i \mathbf{h} \cdot \mathbf{r}) d\mathbf{r} \right] \otimes \left[\int_{NV} \rho(\mathbf{r}) \exp(2\pi i \mathbf{h} \cdot \mathbf{r}) d\mathbf{r} \right],$$

by the frequency convolution theorem (Campbell & Foster (1948), p. 39, pair 202).

$$= V \left[\frac{1}{V} F_N(\mathbf{h}) \right] \otimes \left[\frac{1}{V} F_N(\mathbf{h}) \right] \tag{3}$$

from (2).

$$= \frac{1}{V} \int F_N(\mathbf{k}) F_N(\mathbf{h} - \mathbf{k}) d\mathbf{k} \tag{4}$$

The integration is over the whole reciprocal space. The components of $\mathbf{h}$ and $\mathbf{k}$ may or may not be integers.

$\otimes$ denotes convolution.

Consider large N. $F_N(\mathbf{h}) = NF'(\mathbf{h})$ when the components of $\mathbf{h}$ are integers, otherwise negligible.

$F'(\mathbf{h})$ is the structure factor which is the Fourier transform of a unit cell of point atoms.

Therefore we only consider (3), hence (4), for $\mathbf{h}$, the components of which are integers.

$F_N(\mathbf{k}) = NF'(\mathbf{k})$ when the components of $\mathbf{k}$ are integers, otherwise negligible.

Hence we only consider (4) for $\mathbf{k}$, the components of which are integers.

Therefore, from (4),

$$NF'(\mathbf{h}) = \frac{N}{V}\sum_{\mathbf{k}} F'(\mathbf{k})F'(\mathbf{h}-\mathbf{k}) \tag{5}$$

$F'(\mathbf{h})$ with $\mathbf{h}$ a reciprocal lattice vector is well defined for an infinite crystal. When the integration in (4) is changed to summation in (5), the result must be independent of N, the number of unit cells. Hence the factor N in the right-hand side of (5) is used.

Hence $F'(\mathbf{h}) = \frac{1}{V}\sum_{\mathbf{k}} F'(\mathbf{k})F'(\mathbf{h}-\mathbf{k})$ (6)

This is the Sayre's equation for equal point-atoms (Sayre (1952), (1.3), Fan (1998), (5)). This is an exact equation.

From (6), the Sayre-equation tangent formula (Fan (1998), (21)),

$$\tan\varphi_{\mathbf{h}} = \frac{\sum_{\mathbf{k}} |F'(\mathbf{k})||F'(\mathbf{h}-\mathbf{k})|\sin(\varphi_{\mathbf{k}} + \varphi_{\mathbf{h}-\mathbf{k}})}{\sum_{\mathbf{k}} |F'(\mathbf{k})||F'(\mathbf{h}-\mathbf{k})|\cos(\varphi_{\mathbf{k}} + \varphi_{\mathbf{h}-\mathbf{k}})} \tag{7}$$

Remark: If the crystal contains more than one species of atoms, let the value of the point atom = the atomic number of the species of atoms. Simply squaring the electron density cannot get the same crystal. Hence the Sayre's equation is not obtained.

2.2. Sayre's equation for equal-atom crystal

(1) becomes

$$F(\mathbf{h}) = f(\mathbf{h})\sum \exp[2\pi i(\mathbf{h}\cdot\mathbf{r}_j)] \tag{8}$$

When the conditions of non-negativity and atomicity are satisfied, the structure factor of the squared crystal (Fan (1998), (4))

$$F^{sq}(\mathbf{h}) = f^{sq}(\mathbf{h})\sum \exp[2\pi i(\mathbf{h}\cdot\mathbf{r}_j)] \tag{9}$$

$f^{sq}(\mathbf{h})$ is the scattering factor of the squared atom.

(5) becomes (see Fan (1998), (2))

$$NF^{sq}(\mathbf{h}) = \frac{N}{V}\sum_{\mathbf{k}} F(\mathbf{k})F(\mathbf{h}-\mathbf{k}) \tag{10}$$

The components of $\mathbf{h}$ are integers. $\mathbf{h}$ is a reciprocal lattice vector.
When the condition of equal-atom structure is satisfied (Fan (1998), p. 80)

$$F(\mathbf{h}) = [f(\mathbf{h})/f^{sq}(\mathbf{h})]F^{sq}(\mathbf{h})\,. \tag{11}$$

From (10) and (11), we have the Sayre's equation for equal atoms (Fan (1998), (5))

$$F(\mathbf{h}) = \frac{f(\mathbf{h})}{f^{sq}(\mathbf{h})V}\sum_{\mathbf{k}} F(\mathbf{k})F(\mathbf{h}-\mathbf{k}) \tag{12}$$

This is an exact equation.
The Sayre-equation tangent formula is

$$\tan\varphi_{\mathbf{h}} = \frac{\sum_{\mathbf{k}} |F(\mathbf{k})||F(\mathbf{h}-\mathbf{k})|\sin(\varphi_{\mathbf{k}}+\varphi_{\mathbf{h}-\mathbf{k}})}{\sum_{\mathbf{k}} |F(\mathbf{k})||F(\mathbf{h}-\mathbf{k})|\cos(\varphi_{\mathbf{k}}+\varphi_{\mathbf{h}-\mathbf{k}})} \tag{13}$$

2.3. Extension of Sayre's equation for equal-point-atom crystal to include the in-between Fourier coefficients

Consider a unit cell of the crystal as an object.

The Fourier transform of the unit cell (see (2)),

$$F'(\mathbf{h}) = V\int_V \rho(\mathbf{r})\exp(2\pi i\mathbf{h}\cdot\mathbf{r})d\mathbf{r}$$

$\rho(\mathbf{r})$ are point atoms of unit value. The squared unit cell is identical to the unit cell.

From (3) and (4),

$$F'(\mathbf{h}) = \frac{1}{V}F'(\mathbf{h})\otimes F'(\mathbf{h})$$

$$= \frac{1}{V}\int F'(\mathbf{k})F'(\mathbf{h}-\mathbf{k})d\mathbf{k} \tag{14}$$

The integration is over the whole reciprocal space. The components of $\mathbf{h}$ and $\mathbf{k}$ may or may not be integers.

To simplify the treatment, we treat the one-dimensional case.

$$F'(h) = \frac{1}{a}\int F'(k)F'(h-k)dk \tag{15}$$

a is the length of the unit cell which is the object. Consider the object in the interval [0,1]. The electron density $\rho(x) = 0$ for $x \notin [0,1]$. x is in fractional unit. Use the Shannon's sampling theory (Marks (1991), p. 1),

$$F'(k) = \sum_{m=-\infty}^{\infty} F'(m/2)\sin c(2k-m) \tag{16}$$

The $\sin c$ function is defined as $\sin c(x) = \dfrac{\sin(\pi x)}{\pi x}$.

For even m, $F'(m/2)$ are the Fourier coefficients.

For odd m, $F'(m/2)$ are the in-between Fourier coefficients.

Substitute (16) into (15),

$$F'(h) = \frac{1}{a}\int\left\{\sum_{m=-\infty}^{\infty} F'(m/2)\sin c(2k-m)\right\}\cdot$$

$$\left\{\sum_{n=-\infty}^{\infty} F'(n/2)\sin c[2(h-k)-n]\right\}dk \tag{17}$$

We treat the cardinal series as absolutely convergent (Mark (1991), Sect. 4.4, Higgins (1985), p. 70, Th. 2). From Knopp (1971), p. 146,

$$F'(h)=\frac{1}{a}\int\left\{\sum_{m=-\infty}^{\infty}\sum_{n=-\infty}^{\infty}F'(m/2)F'(n/2)\sin c[2k-m]\sin c[2(h-k)-n]\right\}dk \tag{18}$$

We treat the cardinal series as uniformly convergent (Marks (1991), Sect. 3.3.1, Higgins (1985), p. 70, Th. 1). The double series in (18) can be treated as the special case $(x=y)$ of the two-dimensional cardinal series in Goodman (2005), (2-57). It is taken as uniformly convergent. The integration and the summation in (18) can be interchanged (Titchmarsh (1968), Sect. 1.71).

$$F'(h)=\frac{1}{a}\sum_{m=-\infty}^{\infty}\sum_{n=-\infty}^{\infty}F'(m/2)F'(n/2)\int\sin c[2k-m]\sin c[2(h-k)-n]dk \tag{19}$$

The integral can be denoted by $D(h,m,n)$.

$$D(h,m,n)=\int\sin c[2k-m]\sin c[2k-(2h-n)]dk \tag{20}$$

The set of functions $\sin c[2k-m]$, $m=0,\pm1,+2,\ldots$, forms an orthonormal set (Marks (1991), p.13, Higgins (1985), p. 56, Th. H1).

$$\int\sin c[2k-m]\sin c[2k-n]dk=\delta(n-m) \tag{21}$$

where the Knronecker delta,

$\delta(m)=1$ when $m=0$

and $=0$ when $m\neq 0$

Hence in (20) $D(h,m,n)$ =1 when $m=2h-n$, i.e. $n=2h-m$.

Hence $n+m=2h$.

h is an integer or half-integer.

(19) becomes

$$F'(h)=\frac{1}{a}\sum_{m=-\infty}^{\infty}F'(m/2)F'(h-(m/2)) \tag{22}$$

Return to the three-dimensional case (14). The object is given by the unit cell with $x\in[0,1]$, $y\in[0,1]$, $z\in[0,1]$. Use the three-dimensional Shannon's sampling theory (Goodman (2005), (2-57)),

$$F'(\mathbf{k}) = \sum_{m_1,m_2,m_3} F'(m_1/2, m_2/2, m_3/2)) \sin c[2k_1 - m_1] \sin c[2k_2 - m_2] \cdot \sin c[2k_3 - m_3]$$

The summations of m_1, m_2, m_3 are from $-\infty$ to ∞.

Similar to (17) - (22), we obtain

$$F'(\mathbf{h}) = \frac{1}{V} \sum_{m_1,m_2,m_3} F'(m_1/2, m_2/2, m_3/2) \cdot F'(h_1 - (m_1/2), h_2 - (m_2/2), h_3 - (m_3/2)) \tag{23}$$

This is an exact equation.

The components of $\mathbf{h}$ are integers or half-integers.

The components of $\mathbf{m}$ are integers.

In short notation,

$$F'(\mathbf{h}) = \frac{1}{V} \sum_{\mathbf{m}} F'(\mathbf{m}/2) F'(\mathbf{h} - (\mathbf{m}/2)) \tag{24}$$

(24) is the extension of the Sayre’s equation for equal-point-atom crystal to include the in-between Fourier coefficients.

The tangent formula,

$$\tan \varphi_{\mathbf{h}} = \frac{\sum_{\mathbf{m}} |F'(\mathbf{m}/2)| |F'(\mathbf{h} - (\mathbf{m}/2))| \sin(\varphi_{\mathbf{m}/2} + \varphi_{\mathbf{h}-(\mathbf{m}/2)})}{\sum_{\mathbf{m}} |F'(\mathbf{m}/2)| |F'(\mathbf{h} - (\mathbf{m}/2))| \cos(\varphi_{\mathbf{m}/2} + \varphi_{\mathbf{h}-(\mathbf{m}/2)})} \tag{25}$$

where φ’s are the phases of the complex numbers.

2.4. Extension of Sayre’s equation for equal-atom crystal to include the in-between Fourier coefficients

Follow Sect. 2.2 and 2.3.

$$F^{sq}(\mathbf{h}) = \frac{1}{V} \sum_{\mathbf{m}} F(\mathbf{m}/2) F(\mathbf{h} - (\mathbf{m}/2)) \tag{26}$$

From (11) and (26),

$$F(\mathbf{h}) = \frac{f(\mathbf{h})}{f^{sq}(\mathbf{h}) V} \sum_{\mathbf{m}} F(\mathbf{m}/2) F(\mathbf{h} - (\mathbf{m}/2)) \tag{27}$$

This is an exact equation.

The tangent formula is

$$\tan\varphi_{\mathbf{h}} = \frac{\sum_{\mathbf{m}} |F(\mathbf{m}/2)||F(\mathbf{h}-(\mathbf{m}/2))|\sin(\varphi_{\mathbf{m}/2}+\varphi_{\mathbf{h}-(\mathbf{m}/2)})}{\sum_{\mathbf{m}} |F(\mathbf{m}/2)||F(\mathbf{h}-(\mathbf{m}/2))|\cos(\varphi_{\mathbf{m}/2}+\varphi_{\mathbf{h}-(\mathbf{m}/2)})} \qquad (28)$$

2.5. Demonstrations with crystal structures reported in the literature

Choose a large number of crystal structures reported in the literature, of various sizes, small, mediate, or macromolecular, and with or without heavy atoms. As stated in Yuen (2011f), Sect. 2.3, the calculated in-between Fourier coefficients are good for determination of atomic coordinates. Accurate atomic coordinates are determined by conventional method in X-ray crystallography, including the Sayre’s equation. Hence the calculated in-between Fourier coefficients should be good for the extension of the Sayre’s equation to include the in-between Fourier coefficients. Check whether Eq. (24) and (27) are satisfied, in particular for crystals containing more than one real or complex atom species. See if there is any trend that can be used for determination of the phases of the observed structure factors.

3. Discussions and conclusion

The in-between Fourier coefficients are included by dealing with a unit cell. The form of (24) resembles the form of (6), and the form of (27) resembles the form of (12). As expected, the extension (27) reduces to (12) when all the in-between Fourier coefficients are equal to zero. As the in-between Fourier coefficients are, in fact, not equal to zero, (27) does not include (12). (27) provides additional relation between the Fourier coefficients to that provided by (12). As the number of Fourier coefficients and in-between Fourier coefficients are eight times the number of Fourier coefficients, this is helpful in solving the phase problem. Eq. (27) and the Sayre’s equation for crystal (12) are complementary to each other. They are separate conditions; one cannot replace the other. The Sayre’s equation (12), the extension (27), and the two tangent formulae (13) and (28), together, can be used for crystal atomic-coordinate determination including macromolecular crystals.

The next step is to extend Sect. 2.2 and 2.4 to crystals containing more than one real or complex atom species. Because of the difference in atomic scattering factors, the crystal is only an approximate point-atom crystal. Choose a large number of crystal structures reported in the literature, of various sizes, small, mediate, or macromolecular, and with or without heavy atoms (Sect. 2.5). Check whether Eq. (27) is satisfied. See if there is any trend that can be used for determination of the phases of the observed structure factors. In Fan (1998), p. 81, it is stated that ‘in many cases Sayre’s equation may still be applicable even when the three conditions (positivity, atomicity and equal-atom structure) are not completely satisfied’.

Dual-real-spaces procedures can be used employing the point-atom models of crystals and iterative algorithms with oversampling (Yuen (2011c, 2011d)), sharpening of Patterson function of a crystal (Yuen (2011e)), sharpening of Patterson function of a unit cell (Yuen (2011g)), and the Sayre’ equation (12), the extension (27), and the two tangent formulae (13) and (28).

It will be useful to extend direct methods to include the in-between Fourier coefficients. Giacovazzo et al. (1998, 1999a, 1999b, 1999c) presents the first step for development of direct-methods procedures applied to rational index reflections.

As shown in Yuen (2011a – 2011g), the in-between Fourier coefficients are very important quantities. They may be included in the forthcoming articles in X-ray crystallography.

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41

Sayre’s equation applied to the Patterson function of equal-atom unit cell. Extension of Bragg intensities and mid-Bragg intensities of a unit cell

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Abstract

Sayre’s equation is applied to the Patterson function of equal-atom unit cell, which is used for extension of Bragg intensities and mid-Bragg intensities of a unit cell. An exact equation is obtained. $I(\mathbf{h}) = \dfrac{g(\mathbf{h})}{g^{sq}(\mathbf{h})V}\sum_{\mathbf{m}} I(\mathbf{m}/2)I(\mathbf{h}-(\mathbf{m}/2))$

Keywords: Sayre equation; point-atom; Fourier transform.

1. Introduction

This is a sequel article of Yuen (2011a). Sayre’s equation is applied to the Patterson function of equal-atom unit cell, which leads to extension of Bragg intensities and mid-Bragg intensities of a unit cell (Sect. 2.1, 2.2). An exact equation is obtained.

The mid-Bragg intensities are obtained by interpolation in multidimensions (Yuen (2011b), Sect. 2.2) or by experimental measurement.

Sayre’s equation applied to the Patterson function of equal-atom crystal and Sayre’s equation applied to the Patterson function of equal-atom unit cell are complementary to each other. The Bragg intensities are first extended by application of Sayre’s equation to the Patterson function of a crystal (Sect. 2.3). A consistent set of $g(h)/g^{sq}(h)$, extended Bragg intensities, extended mid-Bragg intensities, and improved interpolated mid-Bragg intensities is then obtained by iterative process (Sect. 2.4).

2.1. Sayre’s equation applied to the Patterson function of an equal-point-atom unit cell

Consider a unit cell of the crystal as an object.

For a unit cell of equal point-atoms with height = 1 (Yuen (2011c), (1)), the square of electron density of the unit cell, the squared unit cell is identical to the unit cell.

$$F'(\mathbf{h}) = \frac{1}{V}\sum_{\mathbf{m}} F'(\mathbf{m}/2)F'(\mathbf{h}-(\mathbf{m}/2)) \tag{1}$$

is the extension of the Sayre’s equation for equal-point-atom crystal to include the in-between Fourier coefficients (Yuen (2011c), (24)).

$F'(\mathbf{h})$ is the Fourier transform of electron density of the unit cell of equal point-atoms.

The components of $\mathbf{h}$ are integers or half-integers.

The components of $\mathbf{m}$ are integers.

Now consider the Patterson function of this unit cell of equal-point-atom crystal with the large peak at the origin removed (Yuen (2011b), (12)). All the Patterson peaks are point atom-pairs of height = 1 (Yuen (2011b), (25)). The square of the Patterson function is identical to the Patterson function.

The Fourier transform of the Patterson function of the unit cell is the diffraction intensity $I'(\mathbf{h})$ of the unit cell (Yuen (2011b), (14)). From (1),

$$I'(\mathbf{h}) = \frac{1}{V}\sum_{\mathbf{m}} I'(\mathbf{m}/2)I'(\mathbf{h}-(\mathbf{m}/2)) \tag{2}$$

The components of $\mathbf{h}$ are integers or half-integers.

The components of $\mathbf{m}$ are integers.

(2) is an exact equation.

When $\mathbf{h}$ is a reciprocal lattice vector, the Fourier transform of the Patterson function of the unit cell is the Bragg intensity $I'(\mathbf{h})$ of the crystal.

When one or more components of $\mathbf{h}$ are half-integers, $I'(\mathbf{h})$ is the mid-Bragg intensity.

2.2. Sayre's equation applied to the Patterson function of an equal-atom unit cell

Consider a crystal of real electron density. Let $p_c(\mathbf{r})$ be the Patterson function of an equal-atom unit cell with the large peak at the origin removed. The effect of division of atoms near a boundary surface, an edge or a corner, is neglected, as in macromolecular crystal. $p_c(\mathbf{r})$ has support in $[-1,1]$, $[-1,1]$, $[-1,1]$ (in fractional units). $p_c(\mathbf{r})$ consists of equal atom-pairs.

For a unit cell containing N atoms, the Fourier transform of the unit cell

$$F(\mathbf{h}) = f(\mathbf{h})\sum_{n=1}^{N}\exp(2\pi i\mathbf{h}\cdot\mathbf{r}_n)\,, \tag{3}$$

$f(\mathbf{h})$ is the atomic scattering factor

The components of $\mathbf{h}$ may or may not be integers.

There are $N(N-1)$, interatomic vector, or $N(N-1)$ Patterson peaks.

From Yuen (2011b), (25) and (26),

$$p_c(\mathbf{r}) = \frac{1}{V}\sum_{m=1}^{N(N-1)}G_c(\mathbf{r}-\mathbf{r}_m) \tag{4}$$

where $$G_c(\mathbf{r}) = \int\left|f(\mathbf{h})\right|^2\exp[-2\pi i\mathbf{h}\cdot\mathbf{r}]d\mathbf{h} \tag{5}$$

$\mathbf{r}_m$ is an interatomic vector.

Let $g(\mathbf{h})$ be the scattering factor of one Patterson peak (an atom-pair).

$g(\mathbf{h})$ = the Fourier transform of $G_c(\mathbf{r})$ in (5)

$$= \iint\left|f(\mathbf{h}')\right|^2\exp[2\pi i(\mathbf{h}-\mathbf{h}')\cdot\mathbf{r}]d\mathbf{h}'d\mathbf{r} \tag{6}$$

The integration of $\mathbf{h}'$ is over the reciprocal space.

The integration of $\mathbf{r}$ is over the support of $p_c(\mathbf{r})$.

The Fourier transform of $p_c(\mathbf{r})$

= the diffraction intensity of the unit cell, $I(\mathbf{h})$ (Yuen (2011b), (14)).

$$I(\mathbf{h}) = g(\mathbf{h})\sum_{m=1}^{N(N-1)}\exp(2\pi i\mathbf{h}\cdot\mathbf{r}_m) \tag{7}$$

The squared Patterson function of the unit cell

$$p_c^{\,2}(\mathbf{r}) = p_c(\mathbf{r})\,p_c(\mathbf{r}) \tag{8}$$

The Fourier transform of the squared Patterson function

$$P_c^{sq}(\mathbf{h}) = g^{sq}(\mathbf{h}) \sum_{m=1}^{N(N-1)} \exp(2\pi i \mathbf{h}\cdot\mathbf{r}_m) \tag{9}$$

$g^{sq}(\mathbf{h})$ is the scattering factor of the squared Patterson peak.

From Yuen (2011c), (26),

$$P_c^{sq}(\mathbf{h}) = \frac{1}{V}\sum_{\mathbf{m}} I(\mathbf{m}/2)I(\mathbf{h}-(\mathbf{m}/2)) \tag{10}$$

From Yuen (2011a), (12),

$$I(\mathbf{h}) = [g(\mathbf{h})/g^{sq}(\mathbf{h})]P_c^{sq}(\mathbf{h})\,. \tag{11}$$

Since $I(\mathbf{h})$, $g(\mathbf{h})$ and $g^{sq}(\mathbf{h})$ are non-negative, $P_c^{sq}(\mathbf{h})$ is non-negative. We may denote $P_c^{sq}(\mathbf{h})$ by $I^{sq}(\mathbf{h})$. (10) becomes

$$I^{sq}(\mathbf{h}) = \frac{1}{V}\sum_{\mathbf{m}} I(\mathbf{m}/2)I(\mathbf{h}-(\mathbf{m}/2)) \tag{12}$$

From (11) and (12), or Yuen (2011c), (27),

$$I(\mathbf{h}) = \frac{g(\mathbf{h})}{g^{sq}(\mathbf{h})V}\sum_{\mathbf{m}} I(\mathbf{m}/2)I(\mathbf{h}-(\mathbf{m}/2)) \tag{13}$$

The components of $\mathbf{h}$ are integers or half-integers.

The components of $\mathbf{m}$ are integers.

(13) is an exact equation.

2.3. Bragg intensities are first extended by application of Sayre's equation to the Patterson function of a crystal

When the mid-Bragg intensities are obtained by interpolation in multidimensions, they have larger uncertainties. In Yuen (2011a), (14), we have extension of Bragg intensities by application of Sayre's equation to the Patterson function of a crystal

$$I(\mathbf{h}) = \frac{g(\mathbf{h})}{g^{sq}(\mathbf{h})V} \sum_{\mathbf{k}} I(\mathbf{k}) I(\mathbf{h}-\mathbf{k}) \tag{14}$$

We can first extend the Bragg intensities by means of (14). Employing the mid-Bragg intensities obtained by interpolation in multidimensions, (13) is then used to extend the mid-Bragg intensities.

Because of atomicity, the factor $g(\mathbf{h})/g^{sq}(\mathbf{h})$ are the same in (14) and (13). Evaluation of $g(\mathbf{h})/g^{sq}(\mathbf{h})$ in Yuen (2011a), Sect. 2.3 can be employed.

2.4. A consistent set of $g(h)/g^{sq}(h)$, extended Bragg intensities, extended mid-Bragg intensities, and improved interpolated mid-Bragg intensities

From (13) and follows Yuen (2011a), (15),

$$\frac{g(h)}{g^{sq}(h)} = V \left\langle \frac{I(\mathbf{h})}{\sum_{\mathbf{m}} I(\mathbf{m}/2) I(\mathbf{h}-(\mathbf{m}/2))} \right\rangle_{h} \tag{15}$$

We have another expression for empirically evaluating $g(h)/g^{sq}(h)$ from the Bragg intensities and mid-Bragg intensities.

Use, say, the average of $g(h)/g^{sq}(h)$ obtained from (14) and (15), and iterations involving Sect. 2.3, (13) and (15), a consistent sets of $g(h)/g^{sq}(h)$, extended Bragg intensities, and improved interpolated mid-Bragg intensities can be obtained. If the mid-Bragg intensities are obtained by interpolation in multidimensions, they can be improved within their uncertainties in the final stage of the iterations. Hence we have a consistent set including improved interpolated mid-Bragg intensities,

2.5. Verification with crystal structures reported in the literature

Choose a large number of crystal structures with high resolutions, reported in the literature, of various sizes, small, mediate, or macromolecular, and with or without heavy atoms. Apply Sect. 2.2 and 2.4 to a unit cell, and Sect. 2.3 to the crystal. Examine the verification of Eq. (13).

3. Discussions and conclusion

Sayre's equation is applied to the Patterson function of equal-point-atom unit cell (Sect. 2.1). An exact equation (2) is obtained.

Sayre's equation is applied to the Patterson function of equal-atom unit cell. An exact equation (13) is obtained. It is used for extension of the Bragg intensities and mid-Bragg intensities. The extended Bragg intensities and mid-Bragg intensities obtained can then be used in various methods for crystal structure determination.

The Bragg intensities are first extended by application of Sayre's equation to the Patterson function of a crystal (Sect. 2.3). A consistent set of $g(h)/g^{sq}(h)$, extended Bragg intensities, extended mid-Bragg intensities, and improved interpolated mid-Bragg intensities is then obtained by iterative process (Sect. 2.4).

It is useful to investigate the applicability of (13) when the three conditions (non-negativity, atomicity and equal-atom structure (Fan (1998), p. 79)) are not satisfied (Shiono & Woolfson (1991), Fan (1998), p. 81), notably the overlapping of the Patterson peaks. The result of applying (13) to a unit cell of large number of crystal structures of high resolutions, reported in the literature can be examined (Sect. 2.5).

The equation obtained, (13) can be combined with the Sayre's equation applied to the Patterson function of equal-atom crystal (Yuen (2011a), (14)), the Sayre's equation for crystal (Fan (1998), (5), Yuen (2011c), (12)), the tangent formula (Fan (1998), (21), Yuen (1911c), (7)), the Sayre's equation for unit cell (Yuen (2011c), (27)) or the tangent formula (Yuen (1911c), (25)), for crystal atomic-coordinate determination.

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42

Accelerated convergence of extended Bragg intensities and mid-Bragg intensities obtained by applying Sayre's equation to the Patterson function of an equal-atom unit cell

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Abstract

Modification of the shape of Patterson peak of a unit cell is used to accelerate convergence of extended Bragg and mid-Bragg intensities obtained by applying Sayre's equation to the Patterson function of an equal-atom unit cell. The modified Patterson function has same peak coordinates as the Patterson function. An effective method is obtained: The problem of accelerated convergence of extended Bragg and mid-Bragg intensities reduces to the problem of choosing $c(\mathbf{h})$ such that $c(\mathbf{h})/[c(\mathbf{h})\otimes c(\mathbf{h})]$ is large for the $I''(\mathbf{h})$ from the experimentally observed $\left|F(\mathbf{h})\right|^2$, and small otherwise. $c(\mathbf{h})/[c(\mathbf{h})\otimes c(\mathbf{h})]$ tends to 0 as $h\rightarrow\infty$. It is found that the accelerated convergence of extension of the Bragg intensities and mid-Bragg intensities by means of the Sayre's equation, is, in fact, for crystals in general. Moreover, it provides a method of evaluating $g(\mathbf{h})/g^{sq}(\mathbf{h})$. For simple equal-atom crystals, the atomic coordinates of which are determined by symmetry, the Bragg intensities and mid-Bragg intensities with extension, the *ab initio* electron densities and the $g(\mathbf{h})/g^{sq}(\mathbf{h})$ obtained can be regarded as fundamental data in solid state physics. These will be useful in studies of solid state physics.

Keywords: Most general modified Patterson peak; most general modified Patterson function; point-atom unit cell; $g(\mathbf{h})/g^{sq}(\mathbf{h})$; $c(\mathbf{h})/[c(\mathbf{h})\otimes c(\mathbf{h})]$; macromolecular crystal; fundamental data; solid state physics.

1. Introduction

This is a sequel article of Yuen (2011d). It deals with the accelerated convergence of extended Bragg intensities and mid-Bragg intensities obtained by applying Sayre's equation (Sayre (1952)) to the Patterson function of an equal-atom unit cell.

In Yuen (2011g), (13), we have presented an exact equation for extension of Bragg intensities and mid-Bragg intensities by applying Sayre's equation to the Patterson function of an equal-atom unit cell.

$$I(\mathbf{h}) = \frac{g(\mathbf{h})}{g^{sq}(\mathbf{h})V}\sum_{\mathbf{m}} I(\mathbf{m}/2)I(\mathbf{h}-(\mathbf{m}/2)) \tag{1}$$

The components of $\mathbf{h}$ are integers or half-integers.

The components of $\mathbf{m}$ are integers.

Starting with experimentally observed $I(\mathbf{m}/2)$ and $I(\mathbf{h}-(\mathbf{m}/2))$, $I(\mathbf{h})$ for all $\mathbf{h}$ are built up.

It would be ideal if $g(\mathbf{h})/g^{sq}(\mathbf{h})$ is large for the experimentally observed $I(\mathbf{h})$, and then drops to almost zero for other values of $\mathbf{h}$. Then the calculation of the extension by means of (1) will converge fast, and an accurate collection of $I(\mathbf{h})$ for all $\mathbf{h}$ will be obtained. In order to achieve this purpose, modification of Patterson function of a unit cell, leading to accelerated convergence of extension of Bragg intensities and mid-Bragg intensities, is studied in this article. The Patterson peak is modified to other shapes, while the peak coordinates remain unchanged. An effective method is presented.

In the accelerated convergence of extension of Bragg intensities and mid-Bragg intensities, $g(\mathbf{h})/g^{sq}(\mathbf{h})$ is not used. It is found that accelerated convergence of the extension of the Bragg intensities and mid-Bragg intensities by means of the Sayre's equation, is, in fact, for crystals in general. Moreover, it provides a method of evaluating $g(\mathbf{h})/g^{sq}(\mathbf{h})$ (Sect. 2.3).

For simple equal-atom crystals, the atomic coordinates of which are determined by symmetry, the Bragg intensities and mid-Bragg intensities with extension, the *ab initio* electron densities and the $g(\mathbf{h})/g^{sq}(\mathbf{h})$ obtained can be regarded as fundamental data in solid state physics (Sect. 2.5). They will be useful in studies of solid state physics.

2.1. Accelerated convergence of extended Bragg intensities and mid-Bragg intensities

Consider a crystal of equal atoms. Let $p_c(\mathbf{r})$ be the Patterson function of an equal-atom unit cell. The effect of division of atoms near a boundary surface, an edge or a corner will be treated later in this section. $p_c(\mathbf{r})$ has support in $[-1,1]$, $[-1,1]$, $[-1,1]$ (in fractional units). $p_c(\mathbf{r})$ consists of equal atom-pairs.

For a unit cell containing N atoms, the Fourier transform of the unit cell

$$F(\mathbf{h}) = f(\mathbf{h})\sum_{n=1}^{N}\exp(2\pi i\mathbf{h}\cdot\mathbf{r}_n)\,, \tag{2}$$

$f(\mathbf{h})$ is the atomic scattering factor.

The components of $\mathbf{h}$ may or may not be integers.

The Patterson function of a unit cell

$$p_c(\mathbf{r}) = \frac{1}{V}\int |F(\mathbf{h})|^2 \exp(-2\pi i\mathbf{h}\cdot\mathbf{r})d\mathbf{h} \tag{3}$$

The Fourier transform of $p_c(\mathbf{r})$ in (3)

$$= \frac{I(\mathbf{h})}{V}$$

$$= \frac{|F(\mathbf{h})|^2}{V} \tag{4}$$

$I(\mathbf{h})$ is the intensity

The Patterson function of the unit cell,

$$p_c(\mathbf{r}) = \frac{1}{V}\sum_{n=1}^{N}\sum_{m=1}^{N}\int \left| f(\mathbf{h})^2 \right| \exp[-2\pi i\mathbf{h}\cdot(\mathbf{r}-(\mathbf{r}_n-\mathbf{r}_m))]d\mathbf{h}$$

$$= \frac{1}{V}\sum_{n=1}^{N}\sum_{m=1}^{N} G_c[\mathbf{r}-(\mathbf{r}_n-\mathbf{r}_m)] \qquad (5)$$

where $G_c(\mathbf{r}) = \int \left| f(\mathbf{h}) \right|^2 \exp[-2\pi i\mathbf{h}\cdot\mathbf{r}]d\mathbf{h}$ (6)

(see Yuen (2011e), (9) and (10)). $G_c(\mathbf{r})$ has the form of a peak at the origin.

By applying Sayre's equation to the Patterson function of a unit cell (3), we obtain

$$I(\mathbf{h}) = \frac{g(\mathbf{h})}{g^{sq}(\mathbf{h})V}\sum_{\mathbf{m}} I(\mathbf{m}/2)I(\mathbf{h}-(\mathbf{m}/2)) \qquad (1)$$

where $g(\mathbf{h})$ is the scattering factor of one Patterson peak (an atom-pair).

$g^{sq}(\mathbf{h})$ is the scattering factor of the squared Patterson peak.

The components of $\mathbf{h}$ are integers or half-integers.

The components of $\mathbf{m}$ are integers.

From (2) and Yuen (2011f), (1),

$$F(\mathbf{h})/f(\mathbf{h}) = \sum \delta(\mathbf{r}'-\mathbf{r}_n)\exp[2\pi i(\mathbf{h}\cdot\mathbf{r}_n)] \qquad (7)$$

This represents an equal-point-atom unit cell.

From (2011e), (24), the Patterson function of a unit cell consisting of equal point-atom-pairs, is given by

$$^{s}p_c(\mathbf{r}) = \frac{1}{V}\int \frac{|F(\mathbf{h})|^2}{[\hat{f}(\mathbf{h})]^2}\exp(-2\pi i\mathbf{h}\cdot\mathbf{r})d\mathbf{h} \qquad (8)$$

For a point-atom unit cell, there is no division of atoms near a boundary surface, an edge or a corner. The point-atom on a boundary surface, an edge or a corner, either belongs to one unit cell or another.

The Fourier transform of $^{s}p_c(\mathbf{r})$ in (8)

$$= \frac{I'(\mathbf{h})}{V}$$

$$= \frac{|F(\mathbf{h})|^2}{V[f(\mathbf{h})]^2} \tag{9}$$

Corresponding to (1), we have

$$I'(\mathbf{h}) = \frac{1}{V} \sum_{\mathbf{m}} I'(\mathbf{m}/2) I'(\mathbf{h} - (\mathbf{m}/2)) \tag{10}$$

from Yuen (2011g), (2).

The most general modified Patterson peak is given by Yuen (2011e), (26)

$$^{s}G_{c}(\mathbf{r}) = \int c(\mathbf{h}) \exp(-2\pi i \mathbf{h} \cdot \mathbf{r}) d\mathbf{h} \tag{11}$$

$c(\mathbf{h})$ is the scattering factor of the modified Patterson peak $^{s}G_{c}(\mathbf{r})$.

$c(\mathbf{h})$ = Fourier transform of $^{s}G(\mathbf{r})$. (12)

$c(\mathbf{h})$ is chosen such that $^{s}G_{c}(\mathbf{r})$ is non-negative.

For $\mathbf{h}$ a reciprocal lattice vector, $c(\mathbf{h})$ is chosen positive.

The most general modified Patterson function is given by Yuen (2011e), (27).

$$^{s}p_{c}(\mathbf{r}) = \frac{1}{64V} \int \frac{|F(\mathbf{h})|^2}{[\hat{f}(\mathbf{h})]^2} c(\mathbf{h}) \exp(-2\pi i \mathbf{h} \cdot \mathbf{r}) d\mathbf{h} \tag{13}$$

The Fourier transform of $^{s}p_{c}(\mathbf{r})$ in (13)

$$= \frac{I''(\mathbf{h})}{64V}$$

$$= \frac{|F(\mathbf{h})|^2}{64V[f(\mathbf{h})]^2} c(\mathbf{h}) \tag{14}$$

The most general modified Patterson function is given by Yuen (2011e), (17)

$$^{s}p_{c}(\mathbf{r}) = \frac{1}{V} \sum_{n=1}^{N} \sum_{m=1}^{N} {}^{s}G_{c}[\mathbf{r} - (\mathbf{r}_{m} - \mathbf{r}_{n})] \tag{15}$$

with ${}^{s}G_{c}(\mathbf{r})$ given by (11) (see Yuen (2011e), (17), (26)).

The squared modified-Patterson-peak

$$\left[{}^{s}G_{c}(\mathbf{r})\right]^{2} = {}^{s}G_{c}(\mathbf{r})\,{}^{s}G_{c}(\mathbf{r}) \tag{16}$$

The squared modified-Patterson-function

$$\left[{}^{s}p_{c}(\mathbf{r})\right]^{2} = {}^{s}p_{c}(\mathbf{r})\,{}^{s}p_{c}(\mathbf{r}) \tag{17}$$

The structure factor of the squared modified-Patterson-function

$$= c^{sq}(\mathbf{h}) \sum_{m=1}^{N(N-1)} \exp(2\pi i \mathbf{h}\cdot\mathbf{r}_{m}) \tag{18}$$

$c^{sq}(\mathbf{h})$ = the scattering factor of the squared modified-Patterson-peak $\left[{}^{s}G_{c}(\mathbf{r})\right]^{2}$.

(19)

$c^{sq}(\mathbf{h})$ = Fourier transform of $\left[{}^{s}G_{c}(\mathbf{r})\right]^{2}$. (20)

Regard $\mathbf{h}$ is any vector in the reciprocal space and use the convolution theorem. From (16), $c^{sq}(\mathbf{h}) = c(\mathbf{h}) \otimes c(\mathbf{h})$ (21)

$\otimes$ denotes convolution.

From Yuen (2011g), (13), we have

$$I''(\mathbf{h}) = \frac{c(\mathbf{h})}{c^{sq}(\mathbf{h})V} \sum_{\mathbf{m}} I''(\mathbf{m}/2)\, I''(\mathbf{h}-(\mathbf{m}/2)) \tag{22}$$

The components of $\mathbf{h}$ are integers or half-integers.

The components of $\mathbf{m}$ are integers.

The problem of accelerated convergence of extended Bragg intensities and mid-Bragg intensities reduces to the problem of choosing $c(\mathbf{h})$ such that $c(\mathbf{h})/[c(\mathbf{h})\otimes c(\mathbf{h})]$ is large for the $I''(\mathbf{h})$ from the experimentally observed $\left|F(\mathbf{h})\right|^{2}$ (see (14)), and small otherwise. $c(\mathbf{h})/[c(\mathbf{h})\otimes c(\mathbf{h})]$ tends to 0 as $h \to \infty$. This is necessary for the convergence of the summation in (22). There may be more than one choice.

After the $c(\mathbf{h})$ is chosen, calculate all $I''(\mathbf{h})$ from the experimentally observed $|F(\mathbf{h})|^2$ (see (14)) and the chosen $c(\mathbf{h})$. Then use (22) to build up $I''(\mathbf{h})$ for all $\mathbf{h}$. For the $I''(\mathbf{h})$ which is not yet obtained by extension, use zero. Because of the choice of $c(\mathbf{h})$, there is accelerated convergence for evaluation of the extended $I''(\mathbf{h})$. Then use $I''(\mathbf{h})$ for all $\mathbf{h}$, and obtain $\dfrac{|F(\mathbf{h})|^2}{V[f(\mathbf{h})]^2}$ from (14). That is, we obtain the Patterson function consisting of equal point-atom-pairs ((8) and (9)). Since $f(\mathbf{h})$ is known, $I(\mathbf{h}) = |F(\mathbf{h})|^2$ for all $\mathbf{h}$ (see (4)), is found. This is the result that we are seeking. We have achieved the aim of accelerated convergence in the evaluation of the extended Bragg intensities and mid-Bragg intensities.

2.2 Choices of $c(\mathbf{h})$

Choices of $c(\mathbf{h})$ in Yuen (2011d), Sect. 2.2 are adopted.

2.3. Evaluation of $g(\mathbf{h})/g^{sq}(\mathbf{h})$

In the evaluation of $I(\mathbf{h})$ for all $\mathbf{h}$ in Sect. 2.1, we have not used (1) and $g(\mathbf{h})/g^{sq}(\mathbf{h})$. This is because we have used $F(\mathbf{h})/f(\mathbf{h})$ in (7) to transform the unit cell to an equal-point-atom unit cell. Here the property of original crystal is not used. This means that **accelerated convergence of the extension of the Bragg intensities and mid-Bragg intensities by means of the Sayre's equation, is, in fact, for crystals in general.** A similar conclusion is found in the sharpening of Patterson function of a unit cell (Yuen (2011e), Sect. 2.2). From the equal-point-atom unit cell, we can then treat the most general modified Patterson peak in (11) and the most general modified Patterson function in (13). $c(\mathbf{h})$ chosen in Sect. 2.1 and 2.2 has no physical meaning. It is just a mathematical technique for accelerating the convergence. There can be more than one optimum choices of $c(\mathbf{h})$. Transform the crystal to equal-point-atom unit cell avoids the problem of evaluating $g(\mathbf{h})/g^{sq}(\mathbf{h})$. In fact, from $I(\mathbf{h})$ for all $\mathbf{h}$, $\mathbf{h}$ being integers or half-integers, we can use (1) to obtain $g(\mathbf{h})/g^{sq}(\mathbf{h})$. This is another achievement of acceleration of the convergence of extension of Bragg intensities and mid- Bragg intensities.

2.4. Bragg intensities are first extended with acceleration

When the mid-Bragg intensities are obtained by interpolation in multidimensions, they have larger uncertainties. Use Yuen (2011d); the Bragg intensities are first extended with acceleration. Employing the mid-Bragg intensities obtained by interpolation in multidimensions, Sect. 2.1 and 2.2 are then used to extend the mid-Bragg intensities with acceleration. The scheme of extension in Yuen (2011d), Sect. 2.3 is used; the Bragg intensities and mid-Bragg intensities are extended one by one:

2.5. Application to simple equal-atom crystal. Fundamental data in solid state physics

Yuen (2011d), Sect. 2.2 is adopted. Here, the mid-Bragg intensities are also extended.

The Bragg intensities and mid-Bragg intensities with extension, the *ab initio* electron densities and the $g(\mathbf{h})/g^{sq}(\mathbf{h})$ obtained can be regarded as fundamental data in solid state physics. They will be useful in studies of solid state physics. The $g(\mathbf{h})/g^{sq}(\mathbf{h})$ obtained provides more understanding of atomic properties.

3. Discussions and conclusion

When Sayre's equation is applied to the Patterson function of an equal-atom unit cell, an exact equation for extension of Bragg intensities and mid-Bragg intensities has been obtained. By means of modifying the shape of the Patterson peak, we obtain accelerated convergence of extension of Bragg intensities and mid-Bragg intensities (Sect. 2.1). The modified Patterson function of the unit cell has same peak coordinates as the Patterson function. An effective method is obtained: The problem of accelerated convergence of extended Bragg intensities and mid-Bragg intensities reduces to the problem of choosing $c(\mathbf{h})$ such that $c(\mathbf{h})/[c(\mathbf{h})\otimes c(\mathbf{h})]$ is large for the $I''(\mathbf{h})$

from the experimentally observed $|F(\mathbf{h})|^2$, and small otherwise. $c(\mathbf{h})/[c(\mathbf{h})\otimes c(\mathbf{h})]$ tends to 0 as $h\rightarrow\infty$. This is necessary for the convergence of the summation in (22). The method is applicable to macromolecular crystal.

Choices of $c(\mathbf{h})$ are suggested in Yuen (2011g), Sect. 2.2. These have to be studied by numerical calculations. Further investigations are needed to have optimum choices of $c(\mathbf{h})$ for three-dimensional crystal, and that the modified Patterson peaks satisfy the conditions of non-negativity and atomicity (Fan (1998), p. 79)). Different optimum choices should lead to the same extended Bragg intensities and mid-Bragg intensities. In fact, different optimum choices leading to the same result indicates that the result is correct.

Sayre's equation applied to the Patterson function of equal-atom crystal and Sayre's equation applied to the Patterson function of equal-atom unit cell are complementary to each other. The Bragg intensities are first extended with acceleration (Sect. 2.4). Sect. 2.1 and 2.2 are then used to extend the mid-Bragg intensities with acceleration.

$g(\mathbf{h})/g^{sq}(\mathbf{h})$ is not used in the accelerated convergence of extension of Bragg intensities and mid-Bragg intensities. It is found that, like Yuen 2011d, accelerated convergence of the extension of the Bragg and mid-Bragg intensities by means of the Sayre's equation is, in fact, for crystals in general. Moreover, it provides a method of evaluating $g(\mathbf{h})/g^{sq}(\mathbf{h})$ (Sect. 2.3). It can be compared with the results obtained by other method of evaluating $g(\mathbf{h})/g^{sq}(\mathbf{h})$ in Yuen (2011c), Sect. 2.3, Yuen (2011d), Sect. 2.4, and Yuen (2011g), Sect. 2.4.

It is useful to investigate the applicability of (22) and the chosen $c(\mathbf{h})$ when the three conditions (non-negativity, atomicity and equal-atom structure (Fan (1998), p. 79) are not satisfied (Shiono & Woolfson (1991), Fan (1998), p. 81), notably if there is overlapping of the peaks in the modified Patterson function (13). We may choose an optimum $c(\mathbf{h})$, while the non-negativity condition of the modified Patterson function is violated. It is mentioned in Woolfson (1991), p. 44 that, in terms of the normalized structure factors, 'Sayre equation holds reasonably well over a very large range of conditions'.

(8) represents the Patterson function of a unit cell consisting of equal point-atom-pairs. For a point-atom unit cell, there is no division of atoms near a boundary surface, an edge or a corner. The point-atom on a boundary surface, an edge or a corner, either belongs to one unit cell or another. Hence the acceleration of convergence of extension of Bragg intensities and mid-Bragg intensities presented shows that (13) of Yuen (2011g) does apply to crystal containing a small number of atoms per unit cell. This is one more achievement of acceleration of the convergence of extension of Bragg and mid-Bragg intensities.

The higher the resolution of the experimental reflections collected, the less will be the overlapping of the peaks of the modified Patterson function. Hence the resolution of the experimental reflections collected should be as high as possible. The result of applying the accelerated convergence of extension of Bragg intensities and mid-Bragg intensities (Sect. 2.1) to a large number of crystal structures of high resolutions, reported in the literature, can be examined.

Extension of the Bragg intensities and mid-Bragg intensities is very useful. This is of great help in crystal structure determination. The number of Bragg intensities and mid-Bragg intensities is increased, while the number of atomic coordinates to be determined remains the same. It reduces the Fourier series truncation error, producing sharper peaks and reducing the ripples. This helps to identify the atoms in the process of solving the structure, for examples, in sharpened Patterson functions (Yuen (2011e)). The atomic coordinates and electron densities obtained will be more accurate. More accurate knowledge of electron density will be helpful in understanding of chemical and biological properties of molecules.

For crystals containing a very small number of atoms, the atomic coordinates can be determined by symmetry (Sect. 2.5). If the Bragg intensities and mid-Bragg intensities are extended, the more accurate electron density obtained will be very useful in studies of solid state physics. In this case, the three conditions of the applicability of the Sayre's equation (non-negativity, atomicity and equal-atom structure (Fan (1998), p. 79)) are satisfied. The Bragg intensities and mid-Bragg intensities with extension, the *ab initio* electron densities and the $g(\mathbf{h})/g^{sq}(\mathbf{h})$ obtained can be regarded as fundamental data in solid state physics. The results obtained should be identical to those obtained in Yuen (2011g), Sect. 2.4.

The accelerated convergence of extended Bragg intensities and mid-Bragg intensities can be applied to the many crystal structures reported in the literature. Iterative algorithms with oversampling may then be applied to refine the electron densities (Yuen (2011a), Sect. 2.3, Yuen (2011b), Sect. 2.2).

Extension of Bragg intensities and mid-Bragg intensities may be regarded as the first step in crystal structure analysis. The Bragg intensities and mid-Bragg intensities are first extended, and then various methods can be applied to solve the structure.

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43

Dual-real-spaces iterative algorithms with oversampling applied to electron density function and Patterson function of a unit cell, plus extension of Bragg intensities and mid-Bragg intensities from application of Sayre’s equation to the Patterson function of a unit cell

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By applying Sayre’s equation to the Patterson function of a unit cell, the Bragg and mid-Bragg intensity can be extended (Yuen (2011d). The convergence of the extension can be accelerated (Yuen (2011e)), and all extended intensities are obtained. This means that there is no Fourier series truncation error. These extended intensities can then be used in dual-real-spaces iterative algorithms applied to electron density function and Patterson function of a unit cell (Yuen (2011c)). Unlike the superresolution by extrapolation employed in Yuen (2011c), these extended intensities are fixed during the iterations. The only quantities to be determined are the phases of the Bragg and mid-Bragg intensities. This may be an improvement.

As in Yuen (2011a), dual-real-spaces iterative algorithms with oversampling, plus extension of Bragg and mid-Bragg intensities, can be used for systematic investigation of equal point-atom crystals. The purpose is to find out the extent of the shapes and size of the unit cells, and the number of atoms per unit cell, that the iterative algorithms will lead to the solution of the crystal structure. The iterative algorithms can then be applied to crystals of various sizes, with or without heavy atoms, structure solved or not yet solved, to determine the a*b initio* electron density and atomic coordinates (Yuen (2011b).

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44

Accelerated convergence of extension of Fourier coefficients and in-between Fourier coefficients by means of Sayre's equation for equal-atom unit cell

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Abstract

Modification of the shape of the atoms is used to accelerate convergence of extended Fourier coefficients and in-between Fourier coefficients obtained by applying Sayre's equation to an equal-atom unit cell. The modified electron density has same atomic coordinates as the original unit cell. An effective method is obtained: The problem of accelerated convergence of extended Fourier coefficients and in-between Fourier coefficients reduces to the problem of choosing $c(\mathbf{h})$ such that $c(\mathbf{h})/[c(\mathbf{h})\otimes c(\mathbf{h})]$ is large for the modified Fourier coefficients and in-between Fourier coefficients $F''(\mathbf{h})$ from the known $F(\mathbf{h})$, and small otherwise. $c(\mathbf{h})/[c(\mathbf{h})\otimes c(\mathbf{h})]$ tends to 0 as $h \to \infty$. The method is applicable to macromolecular crystal. It is found that accelerated convergence of the extension of the Fourier coefficients and in-between Fourier coefficients by means of the Sayre's equation, is, in fact, for crystals in general. Moreover, it provides a method of evaluating $f(\mathbf{h})/f^{sq}(\mathbf{h})$.

Keywords: Point-atom crystal; most general modified atom; most general modified electron density; $f(\mathbf{h})/f^{sq}(\mathbf{h})$; $c(\mathbf{h})/[c(\mathbf{h})\otimes c(\mathbf{h})]$; Gaussian atom; macromolecular crystal.

1. Introduction

This is a sequel article of Yuen (2011d, 2011c). Sayre (1952) has presented a very useful and important exact equation for extension of structure factors for equal-atom crystal. Sayre's equation extended to include the in-between Fourier coefficients (Yuen (2011d), (27)) is

$$F(\mathbf{h}) = \frac{f(\mathbf{h})}{f^{sq}(\mathbf{h})V} \sum_{\mathbf{m}} F(\mathbf{m}/2) F(\mathbf{h} - (\mathbf{m}/2))$$

The components of $\mathbf{h}$ are integers or half-integers.
The components of $\mathbf{m}$ are integers.

It would be ideal if $f(\mathbf{h})/f^{sq}(\mathbf{h})$ is large for the known $F(\mathbf{h})$, and then drops to almost zero for other values of $\mathbf{h}$. Then the calculation of the extension by means of this equation will converge fast, and an accurate collection of $F(\mathbf{h})$ for all $\mathbf{h}$ will be obtained. In general, this is not the case. In order to achieve this purpose, modification of the electron density, leading to accelerated convergence of extension of Fourier coefficients and in-between Fourier coefficients, is studied in this article (Sect. 2.1). The development follows that of Yuen (2011d).

Choices of $c(\mathbf{h})$ are suggested in Sect. 2.2.

The Fourier coefficients and in-between Fourier coefficients are extended one by one (Sect. 2.3).

In the accelerated convergence of extension of Fourier coefficients and in-between Fourier coefficients, $f(\mathbf{h})/f^{sq}(\mathbf{h})$ is not used. It is found that accelerated convergence of the extension of the Fourier coefficients and in-between Fourier coefficients by means of the Sayre's equation, is, in fact, for crystals in general. Moreover, it provides a method of evaluating $f(\mathbf{h})/f^{sq}(\mathbf{h})$ (Sect. 2.4).

2.1. Accelerated convergence of extended Fourier coefficients and in-between Fourier coefficients

Consider a real or complex crystal of equal atoms. For a unit cell containing N atoms, the Fourier transform of the unit cell

$$F(\mathbf{h}) = f(\mathbf{h}) \sum_{n=1}^{N} \exp(2\pi i \mathbf{h} \cdot \mathbf{r}_n), \tag{1}$$

$f(\mathbf{h})$ is the atomic scattering factor

The components of $\mathbf{h}$ may or may not be integers.

The electron density of the unit cell

$$\rho(\mathbf{r}) = \frac{1}{V}\int F(\mathbf{h})\exp(-2\pi i\mathbf{h}\cdot\mathbf{r})d\mathbf{h} \tag{2}$$

V the volume of one unit cell.

The coefficient in (2)

$$= \frac{F(\mathbf{h})}{V} \tag{3}$$

Sayre's equation extended to include the in-between Fourier coefficients (Yuen (2011d), (27)) is

$$F(\mathbf{h}) = \frac{f(\mathbf{h})}{f^{sq}(\mathbf{h})V}\sum_{\mathbf{m}} F(\mathbf{m}/2)F(\mathbf{h}-(\mathbf{m}/2)) \tag{4}$$

where $f^{sq}(\mathbf{h})$ is the scattering factor of the squared atom.

The components of $\mathbf{h}$ are integers or half-integers.

The components of $\mathbf{m}$ are integers.

From (1) and Yuen (2011d), (1),

$$F'(\mathbf{h}) = F(\mathbf{h})/f(\mathbf{h}) = \sum \delta(\mathbf{r}'-\mathbf{r}_n)\exp[2\pi i(\mathbf{h}\cdot\mathbf{r}_n)] \tag{5}$$

This represents an equal-point-atom unit cell.

From (2) and (5), the electron density of this equal-point-atom unit cell

$$\rho'(\mathbf{r}) = \frac{1}{V}\int F'(\mathbf{h})\exp(-2\pi i\mathbf{h}\cdot\mathbf{r})d\mathbf{h}$$

$$= \frac{1}{V}\int \frac{F(\mathbf{h})}{f(\mathbf{h})}\exp(-2\pi i\mathbf{h}\cdot\mathbf{r})d\mathbf{h} \tag{6}$$

The coefficients in (6)

$$= \frac{F'(\mathbf{h})}{V}$$

$$= \frac{F(\mathbf{h})}{Vf(\mathbf{h})} \tag{7}$$

Corresponding to (4), we have

$$F'(\mathbf{h}) = \frac{1}{V}\sum_{\mathbf{m}} F'(\mathbf{m}/2)F'(\mathbf{h}-(\mathbf{m}/2)) \tag{8}$$

from Yuen (2011d), (24).

From (5), the most general modified Fourier transform of the unit cell is given by

$$F''(\mathbf{h}) = \frac{F(\mathbf{h})}{f(\mathbf{h})}c(\mathbf{h}) = c(\mathbf{h})\sum \delta(\mathbf{r}'-\mathbf{r}_n)\exp[2\pi i(\mathbf{h}\cdot\mathbf{r}_n)] \tag{9}$$

Note that the atomic coordinates are the same as those of the unit cell (see (1)).

$c(\mathbf{h})$ is the scattering factor (that is, the Fourier transform) of the modified atom $\xi(\mathbf{r})$. $\xi(\mathbf{r})$ is the most general modified atom. $c(\mathbf{h})$ and $\xi(\mathbf{r})$ have moderate shapes.

$c(\mathbf{h})$ is chosen such that the modified atom $\xi(\mathbf{r})$ is non-negative and confined to a radius of about 0.65Å, and, for Gaussian atom, $c(\mathbf{h})$ has a radius of 1.3Å^{-1} (Sayre (1952), p. 61).

For $\mathbf{h}$ the components of which are integers or half-integers, $c(\mathbf{h})$ is chosen positive.

The modified electron density of the unit cell of modified atoms,

$$\rho''(\mathbf{r}) = \frac{1}{V}\int F''(\mathbf{h})\exp(-2\pi i\mathbf{h}\cdot\mathbf{r})d\mathbf{h}$$

$$= \frac{1}{V}\int \frac{F(\mathbf{h})}{f(\mathbf{h})}c(\mathbf{h})\exp(-2\pi i\mathbf{h}\cdot\mathbf{r})d\mathbf{h} \tag{10}$$

$\rho''(\mathbf{r})$ is the most general modified electron density.

The coefficient in (10)

$$= \frac{F''(\mathbf{h})}{V} \tag{11}$$

$$= \frac{F(\mathbf{h})}{Vf(\mathbf{h})}c(\mathbf{h}) \tag{12}$$

The squared modified-atom

$$[\xi(\mathbf{r})]^2 = \xi(\mathbf{r})\xi(\mathbf{r}) \tag{13}$$

The electron density of the squared modified-unit-cell

$$[\rho''(\mathbf{r})]^2 = \rho''(\mathbf{r})\rho''(\mathbf{r}) \tag{14}$$

The Fourier transform of the squared modified-unit-cell

$$= c^{sq}(\mathbf{h})\sum_n \exp(2\pi i\mathbf{h}\cdot\mathbf{r}_n) \tag{15}$$

$c^{sq}(\mathbf{h})$ is the scattering factor of the squared modified-atom $[\xi(\mathbf{r})]^2$.

$$c^{sq}(\mathbf{h}) = \text{Fourier transform of } [\xi(\mathbf{r})]^2. \tag{16}$$

Use the convolution theorem. From (13),

$$c^{sq}(\mathbf{h}) = c(\mathbf{h})\otimes c(\mathbf{h}) \tag{17}$$

$\otimes$ denotes convolution.

From Yuen (2011d), (27), we have

$$F''(\mathbf{h}) = \frac{c(\mathbf{h})}{c^{sq}(\mathbf{h})V}\sum_{\mathbf{m}} F''(\mathbf{m}/2)F''(\mathbf{h}-(\mathbf{m}/2)) \tag{18}$$

The problem of accelerated convergence of extended Fourier coefficients and in-between Fourier coefficients reduces to the problem of choosing $c(\mathbf{h})$ such that $c(\mathbf{h})/[c(\mathbf{h})\otimes c(\mathbf{h})]$ is large for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$ (magnitude and phase) (see (12)), and small otherwise. The components of $\mathbf{h}$ are integers or half-integers. $c(\mathbf{h})/[c(\mathbf{h})\otimes c(\mathbf{h})]$ tends to 0 as $h\to\infty$. This is necessary for the convergence of the summation in (18). There may be more than one choice.

After the $c(\mathbf{h})$ is chosen, calculate all $F''(\mathbf{h})$ from the known $F(\mathbf{h})$ (see (12)) and the chosen $c(\mathbf{h})$. Then use (18) to build up $F''(\mathbf{h})$ for all $\mathbf{h}$. For the $F''(\mathbf{h})$ which is not yet obtained by extension, use zero. Because of the choice of $c(\mathbf{h})$, there is accelerated convergence for evaluation of the extended $F''(\mathbf{h})$. Then use $F''(\mathbf{h})$ for all $\mathbf{h}$, and obtain $\frac{F(\mathbf{h})}{Vf(\mathbf{h})}$ from (12). That is, we obtain the electron density consisting of equal point atoms ((6) and (7)). Since $f(\mathbf{h})$ is known, $F(\mathbf{h})$ for all

$\mathbf{h}$, is found. This is the result that we are seeking. We have achieved the aim of accelerated convergence in the evaluation of the extended Fourier coefficients and in-between Fourier coefficients. The method is applicable to macromolecular crystal.

2.2 Choices of $c(\mathbf{h})$

Choices of $c(\mathbf{h})$ in Yuen (2011c), Sect. 2.2 are adopted.

2.3. Scheme of extension

The scheme of extension in Yuen (2011b), Sect. 2.3 is used; the Fourier coefficients and in-between Fourier coefficients are extended one by one until all Fourier coefficients and in-between Fourier coefficients for the intended resolution are obtained.

To refine the Fourier coefficients and in-between Fourier coefficients determined by extension, arrange the extended Fourier coefficients and in-between Fourier coefficients in descending order according to their magnitudes. Start from the largest magnitude, repeat the extension of the Fourier coefficients and in-between Fourier coefficients one by one.

2.4. Evaluation of $f(\mathbf{h})/f^{sq}(\mathbf{h})$

See Yuen (2011c), Sect. 2.4.

Accelerated convergence of the extension of the Fourier coefficients and in-between Fourier coefficients by means of the Sayre's equation is, in fact, for crystals in general. From $F(\mathbf{h})$ for all $\mathbf{h}$, we can use (4) to obtain $f(\mathbf{h})/f^{sq}(\mathbf{h})$. This is another achievement of acceleration of the convergence of extension of Fourier coefficients and in-between Fourier coefficients.

3. Discussions and conclusion

By means of modifying the shape of the atoms, we obtain accelerated convergence of extension of Fourier coefficients and in-between Fourier coefficients (Sect. 2.1). The modified electron density has same atomic coordinates as the original unit cell. An effective method is obtained: The problem of accelerated convergence of extended

Fourier coefficients and in-between Fourier coefficients reduces to the problem of choosing $c(\mathbf{h})$ such that $c(\mathbf{h})/[c(\mathbf{h})\otimes c(\mathbf{h})]$ is large for the modified Fourier coefficients and in-between Fourier coefficients $F''(\mathbf{h})$ from the known $F(\mathbf{h})$, and small otherwise. The components of $\mathbf{h}$ are integers or half-integers. $c(\mathbf{h})/[c(\mathbf{h})\otimes c(\mathbf{h})]$ tends to 0 as $h \to \infty$. This is necessary for the convergence of the summation in (18). The method is applicable to macromolecular crystal. Extension of Fourier coefficients and in-between Fourier coefficients may be applicable to low-resolution phasing.

$f(\mathbf{h})/f^{sq}(\mathbf{h})$ is not used in the accelerated convergence of extension of Fourier coefficients and in-between Fourier coefficients. It is found that accelerated convergence of the extension of the Fourier coefficients and in-between Fourier coefficients by means of the Sayre's equation, is, in fact, for crystals in general. Moreover, it provides a method of evaluating $f(\mathbf{h})/f^{sq}(\mathbf{h})$ (Sect. 2.4). It can be compared with the results obtained by other method of determining the scattering factor of the 'squared atom' in the references in Yuen (2011a), Sect. 1, and the result of Yuen (2011c), Sect. 2.4.

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45

Extension of Woolfson's modifications of Sayre's equation to include the in-between Fourier coefficients for crystals containing unequal atoms

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Abstract
Woolfson's modifications of Sayre's equation are extended to include the in-between Fourier coefficients for crystals containing unequal atoms.

Keywords: In-between Fourier coefficients; unequal atoms.

1. Introduction

Woolfson (1958) has presented two very useful and important exact equations for extension of structure factors for crystal containing two or more than two types of atoms, respectively:

$$F(\mathbf{h}) = A(\mathbf{h})V^{-1}\sum_{\mathbf{k}} F(\mathbf{k})F(\mathbf{h}-\mathbf{k}) + B(\mathbf{h})V^{-2}\sum_{\mathbf{k}}\sum_{\mathbf{l}} F(\mathbf{k})F(\mathbf{l})F(\mathbf{h}-\mathbf{k}-\mathbf{l}) \tag{1}$$

$$F(\mathbf{h}) = A(\mathbf{h})V^{-1}\sum_{\mathbf{k}} F(\mathbf{k})F(\mathbf{h}-\mathbf{k}) + B(\mathbf{h})V^{-2}\sum_{\mathbf{k}}\sum_{\mathbf{l}} F(\mathbf{k})F(\mathbf{l})F(\mathbf{h}-\mathbf{k}-\mathbf{l}) + C(\mathbf{h})V^{-3}\sum_{\mathbf{k}}\sum_{\mathbf{l}}\sum_{\mathbf{m}} F(\mathbf{k})F(\mathbf{l})F(\mathbf{m})F(\mathbf{h}-\mathbf{k}-\mathbf{l}-\mathbf{m}) + \ldots \tag{2}$$

These are modifications of Sayre's equation (Sayre (1952)).

This article extends these equations to include the in-between Fourier coefficients for crystals.

2.1. Extension to include the in-between Fourier coefficients for crystals containing two types of atoms

Consider a real or complex, centrosymmetric or non-centrosymmetric crystal of two types of atoms, P and Q. Follow Woolfson (1958), Sect. 2. If we use (1) and Yuen (2011), (24), we will obtain the equation for the extension

$$\begin{aligned} F(\mathbf{h}) = {} & A_{PQ}(\mathbf{h})V^{-1}\sum_{\mathbf{k}} F(\mathbf{k}/2)F(\mathbf{h}-(\mathbf{k}/2)) \\ & + B_{PQ}(\mathbf{h})V^{-2}\sum_{\mathbf{k}}\sum_{\mathbf{l}} F(\mathbf{k}/2)F(\mathbf{l}/2)F(\mathbf{h}-(\mathbf{k}/2)-(\mathbf{l}/2)) \end{aligned} \tag{3}$$

The components of $\mathbf{h}$ are integers or half-integers. $F(\mathbf{h})$ is Fourier coefficient or in-between Fourier coefficient.

The components of $\mathbf{k}$ and $\mathbf{l}$ are integers.

2.2. Extension to include the in-between Fourier coefficients for crystals containing three types of atoms

Consider a real or complex, centrosymmetric or non-centrosymmetric crystal of three types of atoms P, Q and R. Follow Woolfson (1958), Sect. 2, 6. If we use (2) and Yuen (2011), (24), we will obtain the equation for the extension

$$\begin{aligned} F(\mathbf{h}) = {} & A_{PQR}(\mathbf{h})V^{-1}\sum_{\mathbf{k}} F(\mathbf{k}/2)F(\mathbf{h}-(\mathbf{k}/2)) \\ & + B_{PQR}(\mathbf{h})V^{-2}\sum_{\mathbf{k}}\sum_{\mathbf{l}} F(\mathbf{k}/2)F(\mathbf{l}/2)F(\mathbf{h}-(\mathbf{k}/2)-(\mathbf{l}/2)) \\ & + C_{PQR}(\mathbf{h})V^{-3}\sum_{\mathbf{k}}\sum_{\mathbf{l}}\sum_{\mathbf{m}} F(\mathbf{k}/2)F(\mathbf{l}/2)F(\mathbf{m}/2)\cdot \\ & \qquad F(\mathbf{h}-(\mathbf{k}/2)-(\mathbf{l}/2)-(\mathbf{m}/2)) \end{aligned} \tag{4}$$

The components of $\mathbf{h}$ are integers or half-integers. $F(\mathbf{h})$ is Fourier coefficient or in-between Fourier coefficient.

The components of $\mathbf{k}$, $\mathbf{l}$ and $\mathbf{m}$ are integers.

2.3. Extension to include the in-between Fourier coefficients for crystals containing more than three types of atoms

Similar treatment can be used, employing

$$\begin{aligned} F(\mathbf{h}) = {} & A(\mathbf{h})V^{-1}\sum_{\mathbf{k}} F(\mathbf{k}/2)F(\mathbf{h}-(\mathbf{k}/2)) \\ & + B(\mathbf{h})V^{-2}\sum_{\mathbf{k}}\sum_{\mathbf{l}} F(\mathbf{k}/2)F(\mathbf{l}/2)F(\mathbf{h}-(\mathbf{k}/2)-(\mathbf{l}/2)) \\ & + C(\mathbf{h})V^{-3}\sum_{\mathbf{k}}\sum_{\mathbf{l}}\sum_{\mathbf{m}} F(\mathbf{k}/2)F(\mathbf{l}/2)F(\mathbf{m}/2)\cdot \\ & \qquad F(\mathbf{h}-(\mathbf{k}/2)-(\mathbf{l}/2)-(\mathbf{m}/2)) \\ & + \ldots \end{aligned} \tag{5}$$

3. Discussions and conclusion

Woolfson’s modifications of Sayre’s equation are extended to include the in-between Fourier coefficients for crystals containing two, three or more than three types of atoms.

References

Sayre, D. (1952). The squaring method: a new method for phase determination. *Acta Cryst.* **5,** 60-65.

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46

Accelerated convergence of extension of Fourier coefficients and in-between Fourier coefficients by means of modifications of Sayre's equation for crystal of unequal atoms

November 29, 2011

Abstract

Woolfson's modifications of Sayre's equation have been extended to include the in-between Fourier coefficients for crystals containing unequal atoms. Modification of the shape of the atoms is used to accelerate convergence of extended Fourier coefficients and in-between Fourier coefficients for crystal containing two, three, or more than three types of atoms. An effective method is obtained: For crystal containing three types of atoms, the problem of accelerated convergence of extended Fourier coefficients and in-between Fourier coefficients reduces to the problem of choosing $c(\mathbf{h})$ such that $A''_{PQR}(\mathbf{h})$, $B''_{PQR}(\mathbf{h})$ and $C''_{PQR}(\mathbf{h})$ are large for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$, and small otherwise. $A''_{PQR}(\mathbf{h})$, $B''_{PQR}(\mathbf{h})$ and $C''_{PQR}(\mathbf{h})$ tend to 0 as $h \to \infty$. The method is applicable to macromolecular crystal. It is found that accelerated convergence of the extension of the Fourier coefficients and in-between Fourier coefficients by means of modifications of the Sayre's equation, is, in fact, for crystals in general. Moreover, it provides relations between $A_{PQ}(\mathbf{h})$ and $B_{PQ}(\mathbf{h})$, and between $A_{PQR}(\mathbf{h})$, $B_{PQR}(\mathbf{h})$ and $C_{PQR}(\mathbf{h})$. These are useful in evaluation of

$A_{PQ}(\mathbf{h})$ and $B_{PQ}(\mathbf{h})$, $A_{PQR}(\mathbf{h})$, $B_{PQR}(\mathbf{h})$ and $C_{PQR}(\mathbf{h})$.

Keywords: Point-atom crystal; most general modified atom; most general modified electron density; Gaussian atom; macromolecular crystal.

1. Introduction

This is a sequel article of Yuen (2011b) which deals accelerated convergence of extension of structure factors by means of modifications of Sayre's equation for crystal of unequal atoms, and Yuen (2011c) which deals with accelerated convergence of extension of Fourier coefficients and in-between Fourier coefficients for equal-atom unit cell. In Yuen, P. S. (2011d), Woolfson's modifications of Sayre's equation have been extended to include the in-between Fourier coefficients for crystals containing unequal atoms. This article deals with accelerated convergence of extension of Fourier coefficients and in-between Fourier coefficients by means of modifications of Sayre's equation for crystal of unequal atoms. Accelerated convergence is achieved by modifying the electron density.

2.1. Accelerated convergence of extended Fourier coefficients and in-between Fourier coefficients of crystal containing two types of atoms

Consider a real or complex, centrosymmetric or non-centrosymmetric crystal of two types of atoms, P and Q. The Fourier transform of a unit cell

$$F(\mathbf{h}) = \sum_n f_n(\mathbf{h}) \exp[2\pi i(\mathbf{h} \cdot \mathbf{r}_n)] \tag{1}$$

$f_n(\mathbf{h})$ is the atomic scattering factor

The components of $\mathbf{h}$ may or may not be integers.

The electron density of the unit cell

$$\rho(\mathbf{r}) = \frac{1}{V} \int F(\mathbf{h}) \exp(-2\pi i \mathbf{h} \cdot \mathbf{r}) d\mathbf{h} \tag{2}$$

V is the volume of one unit cell.

The coefficient in (2)

$$= \frac{F(\mathbf{h})}{V} \tag{3}$$

Extension of Woolfson’s modification of Sayre’s equation to include the in-between Fourier coefficients (Yuen (2011d), (3)) is

$$F(\mathbf{h}) = A_{PQ}(\mathbf{h})V^{-1}\sum_{\mathbf{k}} F(\mathbf{k}/2)F(\mathbf{h}-(\mathbf{k}/2))$$

$$+ B_{PQ}(\mathbf{h})V^{-2}\sum_{\mathbf{k}}\sum_{\mathbf{l}} F(\mathbf{k}/2)F(\mathbf{l}/2)F(\mathbf{h}-(\mathbf{k}/2)-(\mathbf{l}/2)) \tag{4}$$

The components of $\mathbf{h}$ are integers or half-integers. $F(\mathbf{h})$ is Fourier coefficient or in-between Fourier coefficient.

The components of $\mathbf{k}$ and $\mathbf{l}$ are integers.

$A_{PQ}(\mathbf{h})$ and $B_{PQ}(\mathbf{h})$ can be determined from the scattering factors of the electron densities and their squares and cubes, of the two types of atoms (Woolfson (1958), Sect. 2, 3).

Let Z_n be the atomic number of the n-th atom. The atomic scattering factors have approximately same shapes. Let

$$\hat{f}(\mathbf{h}) = (\sum f_n(\mathbf{h}))/(\sum Z_n) \tag{5}$$

$\hat{f}(\mathbf{h})$ is the atomic scattering factor of a hypothetical atom with $Z = 1$.

$$f_n(\mathbf{h}) \approx Z_n \hat{f}(\mathbf{h}) \tag{6}$$

$$F'(\mathbf{h}) = \frac{F(\mathbf{h})}{\hat{f}(\mathbf{h})} = \sum_n Z_n \exp[2\pi i(\mathbf{h}\cdot\mathbf{r}_n)] \tag{7}$$

This represents a point-atom unit cell.

From (2) and (7), the electron density of this point-atom unit cell

$$\rho'(\mathbf{r}) = \frac{1}{V}\int F'(\mathbf{h})\exp(-2\pi i\mathbf{h}\cdot\mathbf{r})d\mathbf{h}$$

$$= \frac{1}{V}\int \frac{F(\mathbf{h})}{\hat{f}(\mathbf{h})}\exp(-2\pi i\mathbf{h}\cdot\mathbf{r})d\mathbf{h} \tag{8}$$

The coefficient in (8)

$$= \frac{F'(\mathbf{h})}{V} \tag{9}$$

$$= \frac{F(\mathbf{h})}{V\hat{f}(\mathbf{h})}c(\mathbf{h}) \tag{10}$$

Corresponding to (4), we have

$$\begin{aligned} F'(\mathbf{h}) = {} & A'_{PQ}(\mathbf{h})V^{-1}\sum_{\mathbf{k}} F'(\mathbf{k}/2)F'(\mathbf{h}-(\mathbf{k}/2)) \\ & + B'_{PQ}(\mathbf{h})V^{-2}\sum_{\mathbf{k}}\sum_{\mathbf{l}} F'(\mathbf{k}/2)F'(\mathbf{l}/2)F'(\mathbf{h}-(\mathbf{k}/2)-(\mathbf{l}/2)) \end{aligned} \tag{11}$$

Let Z_P and Z_Q be the atomic numbers of atoms of types P and Q respectively.

From (6), Woolfson (1958), (5) and (6),

$$A'_{PQ}(\mathbf{h})Z_P{}^2 + B'_{PQ}(\mathbf{h})Z_P{}^3 = Z_P \tag{12}$$

$$A'_{PQ}(\mathbf{h})Z_Q{}^2 + B'_{PQ}(\mathbf{h})Z_Q{}^3 = Z_Q \tag{13}$$

From (12) and (13), $A'_{PQ}(\mathbf{h})$ and $B'_{PQ}(\mathbf{h})$ are determined. For point-atom crystal, $A'_{PQ}(\mathbf{h})$ and $B'_{PQ}(\mathbf{h})$ are constants, independent of $\mathbf{h}$.

From (7), the most general modified Fourier transform of the unit cell is given by

$$F''(\mathbf{h}) = \frac{F(\mathbf{h})}{\hat{f}(\mathbf{h})}c(\mathbf{h}) = c(\mathbf{h})\sum_n Z_n \exp[2\pi i(\mathbf{h}\cdot\mathbf{r}_n)] \tag{14}$$

Note that the atomic coordinates are the same as those of the unit cell (see (1)).

$c(\mathbf{h})$ is the scattering factor (that is, the Fourier transform) of the modified atom $\xi(\mathbf{r})$. $\xi(\mathbf{r})$ is the most general modified atom. $c(\mathbf{h})$ and $\xi(\mathbf{r})$ have moderate shapes.

$c(\mathbf{h})$ is chosen such that the modified atom $\xi(\mathbf{r})$ is non-negative and confined to a radius of about 0.65Å, and, for Gaussian atom, $c(\mathbf{h})$ has a radius of 1.3Å^{-1} (Sayre (1952), p. 61).

For $\mathbf{h}$ the components of which are integers or half-integers, $c(\mathbf{h})$ is chosen positive.

The modified electron density of the unit cell of modified atoms,

$$\begin{aligned} \rho''(\mathbf{r}) &= \frac{1}{V}\int F''(\mathbf{h})\exp(-2\pi i\mathbf{h}\cdot\mathbf{r})d\mathbf{h} \\ &= \frac{1}{V}\int \frac{F(\mathbf{h})}{\hat{f}(\mathbf{h})}c(\mathbf{h})\exp(-2\pi i\mathbf{h}\cdot\mathbf{r})d\mathbf{h} \qquad (15) \end{aligned}$$

$\rho''(\mathbf{r})$ is the most general modified electron density.

The coefficient in (15)

$$= \frac{F''(\mathbf{h})}{V} \qquad (16)$$

$$= \frac{F(\mathbf{h})}{V\hat{f}(\mathbf{h})}c(\mathbf{h}) \qquad (17)$$

Corresponding to (4), we have

$$\begin{aligned} F''(\mathbf{h}) &= A''_{PQ}(\mathbf{h})V^{-1}\sum_{\mathbf{k}} F''\mathbf{k}/2)F''(\mathbf{h}-(\mathbf{k}/2)) \\ &\quad + B''_{PQ}(\mathbf{h})V^{-2}\sum_{\mathbf{k}}\sum_{\mathbf{l}} F''(\mathbf{k}/2)F''(\mathbf{l}/2)F''(\mathbf{h}-(\mathbf{k}/2)-(\mathbf{l}/2)) \end{aligned} \qquad (18)$$

	P	Q	
The modified atom	$\xi(\mathbf{r})Z_P$	$\xi(\mathbf{r})Z_Q$	(19)

Square of the modified atom $[\xi(\mathbf{r})]^2 Z_P{}^2$ $[\xi(\mathbf{r})]^2 Z_Q{}^2$

Cube of the modified atom $[\xi(\mathbf{r})]^3 Z_P{}^3$ $[\xi(\mathbf{r})]^3 Z_Q{}^3$

The scattering factor of the modified atoms and their squares and cubes

P Q

$$c(\mathbf{h})Z_P \qquad c(\mathbf{h})Z_Q \tag{20}$$

$$c(\mathbf{h})\otimes c(\mathbf{h})Z_P{}^2 . \qquad c(\mathbf{h})\otimes c(\mathbf{h})Z_Q{}^2 \tag{21}$$

$$c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})Z_P{}^3 \qquad c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})Z_Q{}^3 \tag{22}$$

$\mathbf{h}$ is any vector in the reciprocal space.

From (20), (21), (22), Woolfson (1958), (5) and (6),

$$A''_{PQ}(\mathbf{h})c(\mathbf{h})\otimes c(\mathbf{h})Z_P{}^2 + B''_{PQ}(\mathbf{h})c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})Z_P{}^3 = c(\mathbf{h})Z_P \tag{23}$$

$$A''_{PQ}(\mathbf{h})c(\mathbf{h})\otimes c(\mathbf{h})Z_Q{}^2 + B''_{PQ}(\mathbf{h})c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})Z_Q{}^3 = c(\mathbf{h})Z_Q \tag{24}$$

From (23) and (24), $A''_{PQ}(\mathbf{h})$ and $B''_{PQ}(\mathbf{h})$ are determined.

Starting with known Fourier coefficients and in-between Fourier coefficients, $F''(\mathbf{h})$ for all $\mathbf{h}$ are built up by means of (18). **The problem of accelerated convergence of extended Fourier coefficients and in-between Fourier coefficients reduces to the problem of choosing $c(\mathbf{h})$ such that $A''_{PQ}(\mathbf{h})$ and $B''_{PQ}(\mathbf{h})$ are large for the**

$F''(\mathbf{h})$ from the known $F(\mathbf{h})$ (magnitude and phase) (see (17)), and small otherwise. The components of $\mathbf{h}$ are integers or half-integers. $A''_{PQ}(\mathbf{h})$ and $B''_{PQ}(\mathbf{h})$ **tend to 0 as $h \to \infty$.** This is necessary for the convergence of the summation in (18). There may be more than one choice.

After the $c(\mathbf{h})$ is chosen, with $A''_{PQ}(\mathbf{h})$ and $B''_{PQ}(\mathbf{h})$ determined, calculate all $F''(\mathbf{h})$ from the known $F(\mathbf{h})$ (see (17)) and the chosen $c(\mathbf{h})$. Then use (18) to build up $F''(\mathbf{h})$ for all $\mathbf{h}$. For the $F''(\mathbf{h})$ which is not yet obtained by extension, use zero. Because of the choice of $c(\mathbf{h})$, there is accelerated convergence for evaluation of the extended $F''(\mathbf{h})$. Then use $F''(\mathbf{h})$ for all $\mathbf{h}$, and obtain $\dfrac{F(\mathbf{h})}{V\hat{f}(\mathbf{h})}$ from (17). That is, we obtain the electron density consisting of point atoms ((8) and (10)). Since $\hat{f}(\mathbf{h})$ is known, $F(\mathbf{h})$ for all $\mathbf{h}$, is found. This is the result that we are seeking. We have achieved the aim of accelerated convergence in the evaluation of the extended Fourier coefficients and in-between Fourier coefficients. The method is applicable to macromolecular crystal.

2.2. Choices of $c(\mathbf{h})$

Choices of $c(\mathbf{h})$ in Yuen (2011b), Sect. 2.2 are adopted.

2.3. Scheme of extension

The scheme of extension in Yuen (2011a), Sect. 2.3 is used; the Fourier coefficients and in-between Fourier coefficients are extended one by one until all Fourier coefficients and in-between Fourier coefficients for the intended resolution are obtained.

To refine the Fourier coefficients and in-between Fourier coefficients determined by extension, arrange these Fourier coefficients and in-between Fourier coefficients in descending order according to their magnitudes. Start from the largest magnitude, repeat the extension of the Fourier coefficients and in-between Fourier coefficients one by one.

2.4. Evaluation of $A_{PQ}(\mathbf{h})$ and $B_{PQ}(\mathbf{h})$

Follow Yuen (2011b), Sect. 2.4.

Accelerated convergence of the extension of the Fourier coefficients and in-between Fourier coefficients by means of the modified Sayre equation is, in fact, for crystals in general. From $F(\mathbf{h})$ for all $\mathbf{h}$, we can use (4) to obtain a relation between $A_{PQ}(\mathbf{h})$ and $B_{PQ}(\mathbf{h})$, which can be used in the evaluation of $A_{PQ}(\mathbf{h})$ and $B_{PQ}(\mathbf{h})$. This is another achievement of acceleration of the convergence of extension of Fourier coefficients and in-between Fourier coefficients.

2.5. Accelerated convergence of extended Fourier coefficients and in-between Fourier coefficients of crystal containing three types of atoms

Follow Sect. 2.1. Consider a real or complex, centrosymmetric or non-centrosymmetric crystal of three types of atoms P, Q and R. The Fourier transform of the unit cell

$$F(\mathbf{h}) = \sum_n f_n(\mathbf{h}) \exp[2\pi i(\mathbf{h} \cdot \mathbf{r}_n)] \tag{25}$$

The electron density of the unit cell

$$\rho(\mathbf{r}) = \frac{1}{V} \int F(\mathbf{h}) \exp(-2\pi i \mathbf{h} \cdot \mathbf{r}) d\mathbf{h} \tag{26}$$

The coefficient in (26)

$$= \frac{F(\mathbf{h})}{V}$$

Extension of Woolfson’s modification of Sayre’s equation to include the in-between

Fourier coefficients (Yuen (2011d), (4)) is

$$\begin{aligned}F(\mathbf{h}) = {} & A_{PQR}(\mathbf{h})V^{-1}\sum_{\mathbf{k}} F(\mathbf{k}/2)F(\mathbf{h}-(\mathbf{k}/2)) \\ & + B_{PQR}(\mathbf{h})V^{-2}\sum_{\mathbf{k}}\sum_{\mathbf{l}} F(\mathbf{k}/2)F(\mathbf{l}/2)F(\mathbf{h}-(\mathbf{k}/2)-(\mathbf{l}/2)) \\ & + C_{PQR}(\mathbf{h})V^{-3}\sum_{\mathbf{k}}\sum_{\mathbf{l}}\sum_{\mathbf{m}} F(\mathbf{k}/2)F(\mathbf{l}/2)F(\mathbf{m}/2)\cdot \\ & \qquad F(\mathbf{h}-(\mathbf{k}/2)-(\mathbf{l}/2)-(\mathbf{m}/2)) \end{aligned} \tag{27}$$

The components of $\mathbf{h}$ are integers or half-integers. $F(\mathbf{h})$ is Fourier coefficient or in-between Fourier coefficient.

The components of $\mathbf{k}$, $\mathbf{l}$ and $\mathbf{m}$ are integers.

$A_{PQR}(\mathbf{h})$, $B_{PQR}(\mathbf{h})$ and $C_{PQR}(\mathbf{h})$ can be determined from the scattering factors of the electron densities and their squares, cubes and quadruples, of the three types of atoms (Woolfson (1958), Sect. 2, 3).

Note that $A_{PQR}(\mathbf{h})$ and $B_{PQR}(\mathbf{h})$ are different from $A_{PQ}(\mathbf{h})$ and $B_{PQ}(\mathbf{h})$ for two types of atoms (Sect. 2.1).

$$F'(\mathbf{h}) = \frac{F(\mathbf{h})}{\hat{f}(\mathbf{h})} = \sum_n Z_n \exp[2\pi i(\mathbf{h}\cdot\mathbf{r}_n)] \tag{28}$$

represents a point-atom unit cell.

From (26) and (28), the electron density of this point-atom unit cell

$$\begin{aligned}\rho'(\mathbf{r}) &= \frac{1}{V}\int F'(\mathbf{h})\exp(-2\pi i\mathbf{h}\cdot\mathbf{r})d\mathbf{h} \\ &= \frac{1}{V}\int \frac{F(\mathbf{h})}{\hat{f}(\mathbf{h})}\exp(-2\pi i\mathbf{h}\cdot\mathbf{r})d\mathbf{h}\end{aligned} \tag{29}$$

The coefficient in (29)

$$= \frac{F'(\mathbf{h})}{V} \tag{30}$$

$$= \frac{F(\mathbf{h})}{V\hat{f}(\mathbf{h})} c(\mathbf{h}) \tag{31}$$

Corresponding to (27), we have

$$\begin{aligned} F'(\mathbf{h}) = A'_{PQR}(\mathbf{h}) V^{-1} \sum_{\mathbf{k}} F'(\mathbf{k}/2) F'(\mathbf{h} - (\mathbf{k}/2)) \\ + B'_{PQR}(\mathbf{h}) V^{-2} \sum_{\mathbf{k}} \sum_{\mathbf{l}} F'(\mathbf{k}/2) F'(\mathbf{l}/2) F'(\mathbf{h} - (\mathbf{k}/2) - (\mathbf{l}/2)) \\ + C'_{PQR}(\mathbf{h}) V^{-3} \sum_{\mathbf{k}} \sum_{\mathbf{l}} \sum_{\mathbf{m}} F'(\mathbf{k}/2) F'(\mathbf{l}/2) F'(\mathbf{m}/2) \cdot \\ F'(\mathbf{h} - (\mathbf{k}/2) - (\mathbf{l}/2) - (\mathbf{m}/2)) \end{aligned} \tag{32}$$

Let Z_R the atomic numbers of atoms of type R. From Woolfson (1958), (5) and (6),

$$A'_{PQR}(\mathbf{h}) Z_P{}^2 + B'_{PQR}(\mathbf{h}) Z_P{}^3 + C'_{PQR}(\mathbf{h}) Z_P{}^4 = Z_P \tag{33}$$

$$A'_{PQR}(\mathbf{h}) Z_Q{}^2 + B'_{PQR}(\mathbf{h}) Z_Q{}^3 + C'_{PQR}(\mathbf{h}) Z_Q{}^4 = Z_Q \tag{34}$$

$$A'_{PQR}(\mathbf{h}) Z_R{}^2 + B'_{PQR}(\mathbf{h}) Z_R{}^3 + C'_{PQR}(\mathbf{h}) Z_R{}^4 = Z_R \tag{35}$$

From (33), (34) and (35), $A'_{PQR}(\mathbf{h})$, $B'_{PQR}(\mathbf{h})$ and $C'_{PQR}(\mathbf{h})$ are determined. For point-atom crystal, $A'_{PQR}(\mathbf{h})$, $B'_{PQR}(\mathbf{h})$ and $C'_{PQR}(\mathbf{h})$ are constants, independent of $\mathbf{h}$.

From (28), the most general modified Fourier transform of the unit cell is given by

$$F''(\mathbf{h}) = \frac{F(\mathbf{h})}{\hat{f}(\mathbf{h})} c(\mathbf{h}) = c(\mathbf{h}) \sum_{n} Z_n \exp[2\pi i(\mathbf{h} \cdot \mathbf{r}_n)] \tag{36}$$

Note that the atomic coordinates are the same as those of the unit cell (see (25)). $c(\mathbf{h})$ is the scattering factor (that is, the Fourier transform) of the modified atom $\xi(\mathbf{r})$. $\xi(\mathbf{r})$ is the most general modified atom. $c(\mathbf{h})$ and $\xi(\mathbf{r})$ have moderate shapes.

$c(\mathbf{h})$ is chosen such that the modified atom $\xi(\mathbf{r})$ is non-negative and confined to a radius of about 0.65Å, and, for Gaussian atom, $c(\mathbf{h})$ has a radius of 1.3Å^{-1} (Sayre (1952), p. 61).

For $\mathbf{h}$ the components of which are integers or half-integers, $c(\mathbf{h})$ is chosen positive.

The modified electron density of the unit cell of modified atoms,

$$\rho''(\mathbf{r}) = \frac{1}{V}\int F''(\mathbf{h})\exp(-2\pi i\mathbf{h}\cdot\mathbf{r})d\mathbf{h}$$

$$= \frac{1}{V}\int \frac{F(\mathbf{h})}{\hat{f}(\mathbf{h})}c(\mathbf{h})\exp(-2\pi i\mathbf{h}\cdot\mathbf{r})d\mathbf{h} \qquad (37)$$

$\rho''(\mathbf{r})$ is the most general modified electron density.

The coefficient in (37)

$$= \frac{F''(\mathbf{h})}{V} \qquad (38)$$

$$= \frac{F(\mathbf{h})}{V\hat{f}(\mathbf{h})}c(\mathbf{h}) \qquad (39)$$

Corresponding to (27), we have

$$F''(\mathbf{h}) = A''_{PQR}(\mathbf{h})V^{-1}\sum_{\mathbf{k}} F''\mathbf{k}/2)F''(\mathbf{h}-(\mathbf{k}/2))$$

$$+ B''_{PQR}(\mathbf{h})V^{-2}\sum_{\mathbf{k}}\sum_{\mathbf{l}} F''(\mathbf{k}/2)F''(\mathbf{l}/2)F''(\mathbf{h}-(\mathbf{k}/2)-(\mathbf{l}/2))$$

$$+ C''_{PQR}(\mathbf{h})V^{-3}\sum_{\mathbf{k}}\sum_{\mathbf{l}}\sum_{\mathbf{m}} F''(\mathbf{k}/2)F''(\mathbf{l}/2)F''(\mathbf{m}/2)\cdot$$

$$F''(\mathbf{h}-(\mathbf{k}/2)-(\mathbf{l}/2)-(\mathbf{m}/2))$$

$$(40)$$

	P	Q	R
The modified atom	$\xi(\mathbf{r})Z_P$	$\xi(\mathbf{r})Z_Q$	$\xi(\mathbf{r})Z_R$

Square of the modified atom $[\xi(\mathbf{r})]^2 Z_P{}^2$ $[\xi(\mathbf{r})]^2 Z_Q{}^2$ $[\xi(\mathbf{r})]^2 Z_R{}^2$

Cube of the modified atom $[\xi(\mathbf{r})]^3 Z_P{}^3$ $[\xi(\mathbf{r})]^3 Z_Q{}^3$ $[\xi(\mathbf{r})]^3 Z_R{}^3$

Quadruple of the modified atom $[\xi(\mathbf{r})]^4 Z_P{}^4$ $[\xi(\mathbf{r})]^4 Z_Q{}^4$ $[\xi(\mathbf{r})]^4 Z_R{}^4$

(41)

The scattering factors of the modified atoms and their squares and cubes

$$P \qquad Q \qquad R$$

$$c(\mathbf{h})Z_P \qquad c(\mathbf{h})Z_Q \qquad c(\mathbf{h})Z_R$$

$$c(\mathbf{h})\otimes c(\mathbf{h})Z_P{}^2\,. \qquad c(\mathbf{h})\otimes c(\mathbf{h})Z_Q{}^2 \qquad c(\mathbf{h})\otimes c(\mathbf{h})Z_R{}^2$$

$$c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})Z_P{}^3 \qquad c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})Z_Q{}^3$$

$$c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})Z_R{}^3$$

$$c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})Z_P{}^4$$

$$c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})Z_Q{}^4$$

$$c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})Z_R{}^4$$

$\mathbf{h}$ is any vector in the reciprocal space.

(42)

From these scattering factors, and Woolfson (1958), (5) and (6),

$$A''_{PQR}(\mathbf{h})c(\mathbf{h})\otimes c(\mathbf{h})Z_P{}^2 + B''_{PQR}(\mathbf{h})c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})Z_P{}^3$$

$$+C''_{PQR}(\mathbf{h})c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h}){Z_P}^4=c(\mathbf{h})Z_P \qquad (43)$$

$$A''_{PQR}(\mathbf{h})c(\mathbf{h})\otimes c(\mathbf{h}){Z_Q}^2+B''_{PQR}(\mathbf{h})c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h}){Z_Q}^3$$

$$+C''_{PQR}(\mathbf{h})c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h}){Z_Q}^4=c(\mathbf{h})Z_Q \qquad (44)$$

$$A''_{PQR}(\mathbf{h})c(\mathbf{h})\otimes c(\mathbf{h}){Z_R}^2+B''_{PQR}(\mathbf{h})c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h}){Z_R}^3$$

$$+C''_{PQR}(\mathbf{h})c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h})\otimes c(\mathbf{h}){Z_R}^4=c(\mathbf{h})Z_R \qquad (45)$$

From (43), (44) and (45), $A''_{PQR}(\mathbf{h})$, $B''_{PQR}(\mathbf{h})$ and $C''_{PQR}(\mathbf{h})$ are determined.

Starting with known Fourier coefficients and in-between Fourier coefficients, $F''(\mathbf{h})$ for all $\mathbf{h}$ are built up by means of (40). **The problem of accelerated convergence of extended Fourier coefficients and in-between Fourier coefficients reduces to the problem of choosing $c(\mathbf{h})$ such that $A''_{PQR}(\mathbf{h})$, $B''_{PQR}(\mathbf{h})$ and $C''_{PQR}(\mathbf{h})$ are large for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$ (magnitude and phase) (see (39)), and small otherwise. The components of $\mathbf{h}$ are integers or half-integers. $A''_{PQR}(\mathbf{h})$, $B''_{PQR}(\mathbf{h})$ and $C''_{PQR}(\mathbf{h})$ tend to 0 as $h\to\infty$.** This is necessary for the convergence of the summation in (40). There may be more than one choice.

After the $c(\mathbf{h})$ is chosen, with $A''_{PQR}(\mathbf{h})$, $B''_{PQR}(\mathbf{h})$ and $C''_{PQR}(\mathbf{h})$ determined, calculate all $F''(\mathbf{h})$ from the known $F(\mathbf{h})$ (see (39)) and the chosen $c(\mathbf{h})$. Then use (40) to build up $F''(\mathbf{h})$ for all $\mathbf{h}$. For the $F''(\mathbf{h})$ which is not yet obtained by extension, use zero. Because of the choice of $c(\mathbf{h})$, there is accelerated convergence for evaluation of the extended $F''(\mathbf{h})$. Then use $F''(\mathbf{h})$ for all $\mathbf{h}$, and obtain $\dfrac{F(\mathbf{h})}{V\hat{f}(\mathbf{h})}$ from (39). That is, we obtain the electron density consisting of point atoms

((29) and (31)). Since $\hat{f}(\mathbf{h})$ is known, $F(\mathbf{h})$ for all $\mathbf{h}$, is found. This is the result that we are seeking. We have achieved the aim of accelerated convergence in the evaluation of the extended Fourier coefficients and in-between Fourier coefficients. The method is applicable to macromolecular crystal.

2.6. Choices of $c(\mathbf{h})$

Choices of $c(\mathbf{h})$ in Yuen (2011b), Sect. 2.2 are adopted.

2.7. Scheme of extension

Same as Sect. 2.3.

2.8. Evaluation of $A_{PQR}(\mathbf{h})$, $B_{PQR}(\mathbf{h})$ and $C_{PQR}(\mathbf{h})$

Follow Yuen (2011b), Sect. 2.4.

Accelerated convergence of the extension of the Fourier coefficients and in-between Fourier coefficients by means of the modified Sayre equation is, in fact, for crystals in general. From $F(\mathbf{h})$ for all $\mathbf{h}$, we can use (27) to obtain we a relation between $A_{PQR}(\mathbf{h})$, $B_{PQR}(\mathbf{h})$ and $C_{PQR}(\mathbf{h})$, which can be used in the evaluation of $A_{PQR}(\mathbf{h})$, $B_{PQR}(\mathbf{h})$ and $C_{PQR}(\mathbf{h})$. This is another achievement of acceleration of the convergence of extension of Fourier coefficients and in-between Fourier coefficients.

2.9. Accelerated convergence of extended Fourier coefficients and in-between Fourier coefficients of crystal containing more than three types of atoms

Similar treatment can be used, employing Yuen (2011d), (5),

$$F(\mathbf{h}) = A(\mathbf{h})V^{-1}\sum_{\mathbf{k}} F(\mathbf{k}/2)F(\mathbf{h}-(\mathbf{k}/2))$$

$$
\begin{aligned}
&+B(\mathbf{h})V^{-2}\sum_{\mathbf{k}}\sum_{\mathbf{l}}F(\mathbf{k}/2)F(\mathbf{l}/2)F(\mathbf{h}-(\mathbf{k}/2)-(\mathbf{l}/2)) \\
&+C(\mathbf{h})V^{-3}\sum_{\mathbf{k}}\sum_{\mathbf{l}}\sum_{\mathbf{m}}F(\mathbf{k}/2)F(\mathbf{l}/2)F(\mathbf{m}/2)\cdot \\
&\qquad\qquad F(\mathbf{h}-(\mathbf{k}/2)-(\mathbf{l}/2)-(\mathbf{m}/2)) \\
&+\ldots \qquad (46)
\end{aligned}
$$

3. Discussions and conclusion

By means of modifying the shape of the atoms, we obtain accelerated convergence of extension of Fourier coefficients and in-between Fourier coefficients for crystals containing two, three or more than three types of atoms (Sect. 2.1, 2.5, 2.9). The modified electron density has same atomic coordinates as the original unit cell. An effective method is obtained: For crystal containing three types of atoms, the problem of accelerated convergence of extended Fourier coefficients and in-between Fourier coefficients reduces to the problem of choosing $c(\mathbf{h})$ such that $A''_{PQR}(\mathbf{h})$, $B''_{PQR}(\mathbf{h})$ and $C''_{PQR}(\mathbf{h})$ are large for the $F''(\mathbf{h})$ from the known $F(\mathbf{h})$ (magnitude and phase) (see (39)), and small otherwise. The components of $\mathbf{h}$ are integers or half-integers. $A''_{PQR}(\mathbf{h})$, $B''_{PQR}(\mathbf{h})$ and $C''_{PQR}(\mathbf{h})$ tend to 0 as $h \to \infty$. This is necessary for the convergence of the summation in (40). Results for crystal containing two types of atoms are similar. The method is applicable to macromolecular crystal. Extension of Fourier coefficients and in-between Fourier coefficients may be applicable to low- resolution phasing.

$A_{PQ}(\mathbf{h})$ and $B_{PQ}(\mathbf{h})$, $A_{PQR}(\mathbf{h})$, $B_{PQR}(\mathbf{h})$ and $C_{PQR}(\mathbf{h})$ are not used in the accelerated convergence of extension of Fourier coefficients and in-between Fourier coefficients. It is found that accelerated convergence of the extension of the Fourier coefficients and in-between Fourier coefficients by means of the modifications of Sayre's equation, is, in fact, for crystals in general. Moreover, it provides relations between $A_{PQ}(\mathbf{h})$ and $B_{PQ}(\mathbf{h})$, and between $A_{PQR}(\mathbf{h})$, $B_{PQR}(\mathbf{h})$ and $C_{PQR}(\mathbf{h})$. Combing with the relations in Yuen (2011b), Sect. 2.4, 2.8, these are useful in evaluation of $A_{PQ}(\mathbf{h})$ and $B_{PQ}(\mathbf{h})$, $A_{PQR}(\mathbf{h})$, $B_{PQR}(\mathbf{h})$ and $C_{PQR}(\mathbf{h})$ (Sect. 2.4, 2.8).

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47

Sharpening in reciprocal space.

A set of minimum number of Fourier coefficients and in-between Fourier coefficients for generating all the Fourier coefficients and in-between Fourier coefficients for crystals of equal and unequal atoms

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Abstract

By means of sharpening in reciprocal space, a set of minimum number of Fourier coefficients and in-between Fourier coefficients for generating all the Fourier coefficients and in-between Fourier coefficients for a crystal of equal or unequal atoms can be obtained empirically. $c(\mathbf{h})$ is sharp. The modified atom $\xi(\mathbf{r})$ has broad shape.

Keywords: Empirical; non-negativity; atomicity.

1. Introduction

Employing accelerated convergence of extension of Fourier coefficients and in-between Fourier coefficients by means of modifications of Sayre's equation (Yuen (2011d, 2011e)), all Fourier coefficients and in-between Fourier coefficients can be generated by a set of Fourier coefficients and in-between Fourier coefficients. If a set of minimum number of Fourier coefficients and in-between Fourier coefficients for generating all the Fourier coefficients and in-between Fourier coefficients can be determined, the other Fourier coefficients and in-between Fourier coefficients can be obtained by extension with acceleration. This is important and useful in X-ray crystallography, in particular, protein crystals which do not diffract to atomic resolution, or sensitive to radiations.

Methods in crystallography, for example, Yuen (2011a, 2011b), may be applied to the set of minimum number of Fourier coefficients and in-between Fourier coefficients. The amount of computation is much reduced. This is means that applicability of these methods is extended to larger crystals. In Yuen (2011a, 2011b), Sect. 3, it is mentioned that 'sharpening' of $c(\mathbf{h})$ in the reciprocal space may be applied. The radius of the sphere of reflections is reduced. Hence smaller number of reflections is used. $c(\mathbf{h})$ is the scattering factor of the modified atom $\xi(\mathbf{r})$. $c(\mathbf{h})$ is sharp. $\xi(\mathbf{r})$ has broad shape. There are large regions of overlapping of the modified atoms. $\xi(\mathbf{r})$ may have negative ripples. The three conditions (non-negativity, atomicity and equal-atom structure) for the application of Sayre's equation (Fan (1998), p. 79) may not be satisfied. This article suggests that we determine empirically how sharp $c(\mathbf{h})$ can be, or how severe such violations can be tolerated for a crystal containing two types of atoms (Sect. 2.1) or three types of atoms (Sect. 2.2).

2.1. A set of minimum number of Fourier coefficients and in-between Fourier coefficients for generating all the Fourier coefficients and in-between Fourier coefficients for a crystal of equal atoms

Similar to Yuen (2011c), Sect. 2.1. Use Yuen (2011d), Sect 2.1.

We intend to investigate how sharp $c(\mathbf{h})$ can be, i.e. what is the minimum number of Fourier coefficients and in-between Fourier coefficients needed for generating all the Fourier coefficients and in-between Fourier coefficients? For a number of crystals of known structures, we can use $c(\mathbf{h})$ as sharp as possible, accelerated convergence of extension of structure factors is then applied. $\xi(\mathbf{r})$ has broad shape. There will be large regions of overlapping of the modified atoms $\xi(\mathbf{r})$. $\xi(\mathbf{r})$ may have negative regions. We want to see how severe violations of the conditions of atomicity and non-negativity can be tolerated. The admissible sharpness of $c(\mathbf{h})$ can be established empirically. In Yuen (2011c), Sect. 2.1, the admissible sharpness of $c(\mathbf{h})$ has been established for Fourier coefficients. It is reasonable to expect that the admissible sharpness of $c(\mathbf{h})$ for Fourier coefficients and in-between Fourier coefficients should be equal to or sharper than that of Yuen (2011c), Sect. 2.1 for Fourier coefficients.

2.2. A set of minimum number of Fourier coefficients and in-between Fourier coefficients for generating all the Fourier coefficients and in-between Fourier coefficients for a crystal of unequal atoms

Similar to Yuen (2011c), Sect. 2.2. Use Yuen (2011e), Sect 2.1, 2.5, 2.9.

The admissible sharpness of $c(\mathbf{h})$ is established empirically. $c(\mathbf{h})$ is sharp. $\xi(\mathbf{r})$ has broad shape. The admissible sharpness of $c(\mathbf{h})$ for Fourier coefficients and in-between Fourier coefficients should be equal to or sharper than that of Yuen (2011c), Sect. 2.2 for Fourier coefficients.

3. Discussions and conclusion

For crystals containing equal or unequal atoms, with optimum choice of $c(\mathbf{h})$, we can obtain empirically a set of minimum number of Fourier coefficients and in-between Fourier coefficients or modified Fourier coefficients and in-between Fourier coefficients which generate all the Fourier coefficients and in-between Fourier coefficients. As this set is obtained empirically from a number of crystals of known structures, the environment of atoms, such as distances between nearest-neighbors, next nearest-neighbors, bond lengths and bond angles, etc, are included implicitly.

Methods in crystallography, for example, Yuen (2011a, 2011b), may be applied to the set of minimum number of Fourier coefficients and in-between Fourier coefficients or modified Fourier coefficients and in-between Fourier coefficients. As we need only deal with the set of minimum number of Fourier coefficients and in-between Fourier coefficients or modified Fourier coefficients and in-between Fourier coefficients, this means that applicability of methods in crystallography is extended to larger crystals.

The Fourier coefficients and in-between Fourier coefficients can be extended to atomic resolution. Direct methods can then be applied.

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48

Combination of methods in X-ray crystallography and iterative algorithms with oversampling

November 29, 2011

Abstract

Sayre’s equation and its modifications are incorporated with point-atom model, and sharpening of Patterson function of a unit cell. Iterative algorithms with oversampling are employed to alternate between point atoms, sharp atoms, moderate shape and broad shape of $\xi(\mathbf{r})$. The atomic coordinates are fixed in the Sayre’s equation and its modifications, and sharpening of Patterson function. The main objective is to determine the atomic coordinates.

Keywords: Point atom; sharpening; moderate shape; broad shape.

1. Introduction

This is a sequel article of Yuen (2011i). In the above, we have presented articles about phase-retrieval method, iterative algorithms with oversampling, point-atom model, sharpening of Patterson function, and Sayre’s equation and its modifications for a unit cell. Now we try to combine together these approaches.

2.1. Phase-retrieval method incorporates with Sayre’s equation and its modification for a unit cell

In Yuen (2011a, 2011c, 2011d), solution for the phase problem are obtained by means of phase-retrieval method. In-between | Fourier coefficients | (or mid-Bragg intensities) are employed. In Yuen (2011n, 2011o), we have accelerated convergence of extension of Fourier coefficients and in-between Fourier coefficients by means of Sayre's equation and its modifications for crystals of equal atoms and unequal atoms. As in-between Fourier coefficients are used in Yuen (2011n, 2011o), phase-retrieval method can be applied. As mentioned in Yuen (2011b), Sect. 2.3, Yuen (2011e), Sect. 2.2, the process of phase-retrieval method is time-consuming, iterative algorithms with oversampling will be employed.

2.2. Iterative algorithms with oversampling applied to the most general modified electron density $\rho''(\mathbf{r})$ and the most general modified Fourier transform of the unit cell $F''(\mathbf{h})$

In Yuen (2011n, 2011o), we have accelerated convergence of extension of Fourier coefficients and in-between Fourier coefficients by means of Sayre's equation and its modifications for crystals of equal atoms and unequal atoms. In Yuen (2011n), Sect. 2.1, and Yuen (2011o), Sect. 2.1, 2.5, there are the most general modified Fourier transform of the unit cell $F''(\mathbf{h})$, the most general modified atom $\xi(\mathbf{r})$, and the most general modified electron density of the unit cell $\rho''(\mathbf{r})$. $c(\mathbf{h})$ and $\xi(\mathbf{r})$ have moderate shapes. $F''(\mathbf{h})$ from the known $F(\mathbf{h})$ is large, and small otherwise. Hence $F''(\mathbf{h})$ for all $\mathbf{h}$, the components of which are integers or half-integers, can be used. This means that there is no Fourier series truncation error. Iterative algorithms with oversampling can applied to the most general modified electron density of the unit cell $\rho''(\mathbf{r})$ and the most general modified Fourier transform of the unit cell $F''(\mathbf{h})$ (Yuen (2011b, 2011e)). Initial phases in Yuen (2011f) may be employed.

2.3. Iterative algorithms with oversampling applied to the minimum number of Fourier coefficients and in-between Fourier coefficients

In Yuen (2011p), we have sharpening of $c(\mathbf{h})$ in reciprocal space. The modified atom $\xi(\mathbf{r})$ has broad shape. We obtain a set of minimum number of Fourier coefficients and in-between Fourier coefficients for generating all the Fourier coefficients and in-between Fourier coefficients for crystal of equal or unequal atoms. Iterative algorithms with oversampling can be applied to the modified electron density of the unit cell, and the minimum number of Fourier coefficients and in-between Fourier coefficients. The modified atoms may have large regions of overlap, and there are negative regions. Initial phases in Yuen (2011f) may be employed.

As the number of Fourier coefficients and in-between Fourier coefficients used is small, the amount of computation is reduced, at the expense that the modified atoms may have large regions of overlap, and there are negative regions. As mentioned in Yuen (2011n), Sect. 2.1, and Yuen (2011o), Sect. 2.1, 2.5, the atomic coordinates of the modified atoms $\xi(\mathbf{r})$ are the same as those of the crystal. The gross shapes of the molecules and the atomic coordinates can be determined. The shapes of the modified atoms $\xi(\mathbf{r})$ may be difficult to identify because of the large regions of overlapping. Moreover $\xi(\mathbf{r})$ has broad shape which may be very different from the atoms of the crystal.

2.4. Combination of Sect. 2.2 and 2.3.

Follow Yuen (2011i), Sect. 2.3. Iterative algorithms with oversampling are used.

2.5. Combination of Sect. 2.4 and sharpening of Patterson function of a unit cell

In dual-real-spaces iterative algorithms with oversampling applied to electron density function and Patterson function of a unit cell, plus acceleration of extension of Bragg and mid-Bragg intensities (Yuen (2011k, 2011m),

point-atom model is used in the electron density function, and

sharpened Patterson peaks (hence sharp atoms) are used for the Patterson function (Yuen (2011j).

A large number of Fourier coefficients and in-between Fourier coefficients are used.

In Sect. 2.2, 2.3, and sharpening of Patterson function, the atomic coordinates are the same as those of the crystal.

Hence we can use iterative algorithms with oversampling, which alternate between the processes in Yuen (2011m), and Sect. 2.2, 2.3, i.e. alternate between point atoms, sharp atoms, moderate shape and broad shape of $\xi(\mathbf{r})$. The main objective is to determine the atomic coordinates. The shapes of the atoms are immaterial.

3. Discussions and conclusion

We have incorporate Sayre's equation and its modifications with point-atom model and sharpening of Patterson function of a unit cell. Iterative algorithms with oversampling are employed to alternate between point atoms, sharp atoms, moderate shape and broad shape of $\xi(\mathbf{r})$. The main objective is to determine the atomic coordinates. The basic idea is to use different shapes of the atoms, with the atomic coordinates fixed in the Sayre's equation and its modifications, and sharpening of Patterson function, and try to determine the atomic coordinates.

After the atomic coordinates are determined, follow Yuen (2011b), Sect. 2.3, Yuen (2011e), Sect. 2.2. By means of iterative algorithms with oversampling, refine and obtain the *ab initio* electron density (R -index = 0). Shapes of the atoms are obtained. From this *ab initio* electron density, more accurate atomic coordinates are obtained.

The next step will be to combine these iterative algorithms with other methods in X-ray crystallography, for examples, isomorphous replacement and direct methods extended to include in-between Fourier coefficients.

It will be very useful if we can develop a routine and efficient method of determining atomic coordinates of macromolecular crystal. We may use the method presented in this article and perform systematic studies of artificial crystals, and crystals in reality, of various sizes, small, mediate, or macromolecular, and with or without heavy atoms, as in Yuen (2011g, 2011h).

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49

Determination of structure of nonperiodic object by means of methods in X-ray crystallography

November 29, 2011

This is a sequel of Yuen (2011a, 2011b).

Much work has been done in the structure determination of macromolecules. Perrakis et al. (2011) presents toolkit of methods. Martin et al. (2011) describes single particle imaging. Wilson (2011) describes construction of high-resolution 3D map of electron density of complex biomolecules. Santucci et al. (2011) presents optical tweezers for synchrotron radiation probing of trapped biological and soft matter objects in aqueous environments. Garman & Weik (2011) reviews macromolecular crystallography radiation damage research. Modern electron microscope is reviewed in Thomas & Midgley (2011).

Various methods have been developed in structure determination in X-ray crystallography, such as sharpening of Patterson function, direct methods, point-atom model, accelerated convergence of extension of Bragg intensities and structure factors, and sharpening in reciprocal space. These powerful methods can be applied to structure determination of nonperiodic object. As in Yuen (2011b), Sect. 2.3, use an orthogonal or triclinic container to contain a macromolecule. The container plus solution and the macromolecule forms a basic unit cell, of which the structure will be determined. The experimentally determined dimensions of the container give the unit cell constants. Mathematically add replicas of the unit cell successively to give a three-dimensional crystal. | Fourier coefficients | and in-between | Fourier coefficients | are obtained from the experimentally measured diffraction intensity of the container plus the solution and the macromolecule. To avoid radiation damage, the solution can be frozen. Methods in X-ray crystallography can be applied to this crystal.

If only | Fourier coefficients | are employed, use, for examples, Yuen (2011c – 2011i).

If both | Fourier coefficients | and in-between | Fourier coefficients | are employed, use, for examples, Yuen (2011j -2011s).

As in Yuen (2011b), Sect. 2.4, a macromolecule, can be embedded in a liquid in a rectangular container, which is then solidified.

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50

Determination of structure of fibre by means of methods in X-ray crystallography

November 29, 2011

This is a sequel of Yuen (2011a, 2011d).

Various methods have been developed in structure determination in X-ray crystallography, such as sharpening of Patterson function, direct methods, point-atom model, accelerated convergence of extension of Bragg intensities and structure factors, and sharpening in reciprocal space. These powerful methods can be applied to structure determination of a fibre.

Because of the 1-dimensional periodic structure, the diffracted intensity is greatly enhanced to form strong diffraction planes which can be measured accurately.

For a freely rotating fibre, determine its maximum diameter (Yuen (2011b), (H)). For a fixed fibre, determine its maximum length along any direction normal to the fibre axis (Yuen (2011c), Sect. 2.2). Use an orthogonal unit cell to contain one period of the fibre. Mathematically add replicas of the unit cell successively to give a 3-dimensional crystal.

Alternatively, as in Yuen (2011d), Sect. 3.2, use an orthogonal container to contain the fibre. The fibre axis is along the longest dimension of the container. The solution is frozen at very low temperature. The basic unit cell consists of one period of the fibre plus the frozen solution and the top, bottom and two sides of the container. The molecules of the solution and the walls of the container constitute background of constant electron densities in comparison to the strong diffraction planes of the fibre. The atomic coordinates of the fibre will be determined.

As in Yuen (2011d), Sect. 3.3, the fibre can be embedded in a liquid in a rectangular container, which is then solidified.

$|$ Fourier coefficients $|$ and in-between $|$ Fourier coefficients $|$ for ($l+\frac{1}{2}$, and/or $m+\frac{1}{2}$, n) are obtained from the experimentally measured diffraction intensity of the fibre, and, if appropriate, the container plus the solution. The other in-between $|$ Fourier coefficients $|$ may be obtained by interpolation in multidimensions. Methods in X-ray crystallography can be applied to this crystal.

If only $|$ Fourier coefficients $|$ are employed, use, for examples, Yuen (2011e – 2011k).

If both $|$ Fourier coefficients $|$ and in-between $|$ Fourier coefficients $|$ are employed, use, for examples, Yuen (2011m -2011u).

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51

Determination of structure of membrane by means of methods in X-ray crystallography

November 29, 2011

This is a sequel of Yuen (2011a, 2011c).

Various methods have been developed in structure determination in X-ray crystallography, such as sharpening of Patterson function, direct methods, point-atom model, accelerated convergence of extension of Bragg intensities and structure factors, and sharpening in reciprocal space. These powerful methods can be applied to structure determination of a planar periodic membrane.

Because of the 2-dimensional periodic structure, the diffracted intensity is greatly enhanced to form strong diffraction lines which can be measured accurately.

For a membrane, determine its maximum thickness (Yuen (2011b), (J)). Use an orthogonal unit cell to contain one period of the membrane. Mathematically add replicas of the unit cell successively to give a 3-dimensional crystal.

Alternatively, as in Yuen (2011c), Sect. 2.2, use an orthogonal container to contain the membrane. The solution is frozen at very low temperature. The basic unit cell consists of one period of the membrane plus the frozen solution and the top and bottom of the container. The molecules of the solution and the walls of the container constitute background of constant electron densities in comparison to the strong diffraction lines of the membrane. The atomic coordinates of the membrane will be determined.

As in Yuen (2011c), Sect. 2.3, the membrane can be embedded in a liquid in a rectangular container, which is then solidified.

$|$ Fourier coefficients $|$ and in-between $|$ Fourier coefficients $|$ for $(l,\ m,\ n+\frac{1}{2})$

are obtained from the experimentally measured diffraction intensity of the membrane, and, if appropriate, the container plus the solution. The other in-between | Fourier coefficients | may be obtained by interpolation in multidimensions. Methods in X-ray crystallography can be applied to this crystal.

If only | Fourier coefficients | are employed, use, for examples, Yuen (2011d – 2011j).

If both | Fourier coefficients | and in-between | Fourier coefficients | are employed, use, for examples, Yuen (2011k -2011t).

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Applications.

In the above, the phase of the Fourier transform of an object is found from the magnitude. The waves may be optical wave, acoustic wave and matter wave, including electromagnetic wave, X-ray, electron, ion and neutron waves. The waves may be coherent, incoherent or partially coherent. These will be used in the following.

For optical wave and X-ray, the intensities or magnitude of the mutual intensities of the diffraction points, diffraction rods or diffraction planes are measured.

52

Microcavity arrays. Parallel arrays of macromolecules

November 29, 2011

Abstract
The microcavity arrays of Diaz et al. (2005), Bunk et al. (2007a), Bunk et al. (2007b) and Bunk et al. (2008) may be arranged as one-dimensional or two-dimensional arrays of freely rotating or fixed macromolecules. The shape and dimensions, and constant electron densities of the silicon walls and the glass slides form a very strong container constraint. The region of constant electron density of the solvent provides a solvent constraint. This offers unique opportunities for determination of *ab initio* electron densities of macromolecules in solution. Quasi-monochromatic coherent, incoherent, or partially coherent wave may be used as wave incident on microcavity arrays. A quasi-monochromatic partially coherent radiation may be formed by a combination of a coherent wave and an incoherent wave.

Keywords: Macromolecules; diffraction; one-dimensional periodic object; fibre; two-dimensional periodic object; membrane; diffraction rods; Bragg rods; container constraint; solvent constraint; tight supports; *ab initio* electron densities; very probably unique solution.

1. Introduction

Diaz et al. (2005), Bunk et al. (2007a), Bunk et al. (2007b) and Bunk et al. (2008) have introduced a very useful and important instrument, microcavity arrays. By means of a microcavity array, an one-dimensional periodic object, a fixed fibre, can be provided. The shape and dimensions, and constant electron densities of the silicon walls and the glass slides form a very strong container constraint. The region of constant electron density of the solvent provides a solvent constraint.

In the Summary and Outlook of Bunk et al. (2007b), Bunk et al. (2007b) and Bunk et al. (2008), it is mentioned that ‘Scaling down the channels in these arrays to the range of 10-50 nm will offer unique opportunities for studies of confinement-induced ordering of proteins in solution'. Along the same direction of thinking, this article presents primitive ideas about parallel arrays of macromolecules. This offers unique opportunities for determination of *ab initio* electron densities of macromolecules in solution.

One-dimensional microcavity array of freely rotating macromolecules is presented in Sect. 2.1. Fixed macromolecule is presented in Sect. 2.2. Two-dimensional microcavity array is presented in Sect. 2.3. The diffraction intensity is greatly enhanced to form strong diffraction rods (Bragg rods) and can be measured accurately.

Quasi-monochromatic coherent, incoherent, or partially coherent wave may be used as wave incident on microcavity arrays (Sect. 3). A quasi-monochromatic partially coherent radiation may be formed by a combination of a coherent wave and an incoherent wave.

Partially coherent illumination is used in Bunk et al. (2007a), Bunk et al. (2007b) and Bunk et al. (2008).

Fluorescence imaging is presented in Webb et al. (2006), Chung et al. (2007) and Wu et al. (2008).

2.1. Freely rotating macromolecules

In Diaz et al. (2005), Fig. 1, (b), Fig. 2, (a), and Sect. 2, the silicon plate is tilted at an angle of, say, 10°. Very dilute solution of a rod-like macromolecule is added to the top side of the channel. Very dilute solution is used as we desire to have only one macromolecule in one channel. In this respect, fluorescence imaging may be useful. The rod-like macromolecule will settle at the lower end of the bottom of the channel. If the macromolecule is balance in weight, it will have random orientation. Hence, for all the channels, we have an one-dimensional array of freely rotating macromolecules. Because of the one-dimensional periodic structure, the diffracted intensity is greatly enhanced to form strong diffraction planes which can be measured accurately. The in-between | Fourier coefficients | in the diffraction planes can be measured; no interpolation is needed. The molecules of the solvent have random locations and orientations. It constitutes a background of constant electron density in comparison to the strong diffraction planes of the macromolecules. The region of constant electron density of the solvent provides a solvent constraint. Likewise, the silicon walls and the glass slides (Diaz et al. (2005), Fig. 1, (a)) have constant electron densities. The shape and dimensions, and constant electron densities of the silicon walls and glass plates form a very strong container constraint. The basic unit cell consists of one macromolecule plus the solvent and the silicon walls and the glass slides. The silicon walls and the glass slides provide the six sharp boundary surfaces. These are tight supports. Iterative algorithms with oversampling may then be applied to obtain an *ab initio* electron density of the macromolecules (Yuen (2011c, 2011f)). The tight supports of the sharp boundaries, and the constant electron densities of the solvent and the silicon walls help the iterations to converge to a unique electron density, like the solvent flattening method (Wang (1981, 1985). From this electron density, the atomic species, chemical formula, and atomic coordinates of the macromolecule in the basic unit cell are determined.

If we regard the rod-like macromolecule to have a head and a tail, we need to have the tails of all the macromolecule at the lower end of the bottom of the channel. Fluorescence imaging may be used to identify the head, say, of the macromolecule. Then only macromolecule in a channel with a fluorescence head will be used.

To help the movement of the solvent and the macromolecules, the lower end of the bottom of the channel may be equipped with an adjustable glass plate to control the solution to flow out of the channel.

2.2. Fixed macromolecules

In Diaz et al. (2005), Fig. 1, (b), Fig. 2, (a), and Sect. 2, a channel has a rectangular bottom with two long edges. The orientation of the silicon plate can be adjusted so that a channel is rested upon a long edge tilting at an angle of, say, 10°, that is, the channel appears to have a V-shaped bottom. If the rod-like macromolecule is not balanced in weight, it will rest with its heavier side at the bottom. Fluorescence imaging may be used to identify the heavier side, say, of the macromolecule. Then only macromolecule in a channel with its heavier side at the bottom will be used. Hence we have a fixed macromolecule.

Powerful methods in X-ray crystallography, such as sharpening of Patterson function, direct methods, point-atom model, accelerated convergence of extension of Bragg intensities and structure factors, and sharpening in reciprocal space, can be applied to structure determination of an one-dimensional array of fixed macromolecules (Yuen (2011h, 2011i)).

2.3. Two-dimensional microcavity array

Parallel microcavity arrays can be combined to form a two-dimensional array. Following Sect. 2.1 or 2.2, we have freely rotating macromolecule or fixed macromolecule in two dimensions. The diffraction intensity is greatly enhanced to form strong diffraction rods and can be measured accurately. For the same number of channels, the one-dimensional array and two-dimensional array have the same amount of diffraction energy. For the one-dimensional array, the diffraction energy is concentrated in diffraction plane. For the two-dimensional array, the diffraction energy is concentrated in diffraction rods. The two-dimensional array has more concentration of diffraction intensities than the one-dimension arrays in Sect. 2.2, 2.3. The in-between | Fourier coefficients | along the diffraction rods can be measured; no interpolation is needed. Iterative algorithms with oversampling may then be applied to obtain a*b initio* electron densities of the macromolecules (Yuen (2011e, 2011g)).

Powerful methods in X-ray crystallography, can be applied to structure determination of a two-dimensional array of fixed macromolecule (Yuen (2011j)).

3. Quasi-monochromatic coherent wave

Quasi-monochromatic coherent wave can be used as the wave incident on the microcavity arrays of freely rotating macromolecules.

3.1. One-dimensional microcavity array (a fixed fibre)

We will set x_1 along one channel of the microcavity array. The channels are periodic along x_3. The microcavity array forms a fixed fibre, with x_3 the fibre axis. Each channel is one period of the fibre. x_1, x_2, x_3 forms an orthogonal system.

In the following about projections, we will apply Yuen (2011k), Sect. 2.6.1, 2.6.2, 2.6.3, and 2.6.4 modified for fixed fibre, and the projections may be non-centrosymmetric or complex.

Projection of one period of a fixed fibre to a plane normal to the fibre axis:

(A) Diffraction intensity of the diffraction plane for $\omega_3 = 0$ is used

Ab initio $f_p(x_1, x_2)$ of the projection is determined (Yuen (2011k), Sect. 2.6.1, (A)).

(B) Magnitude of the mutual intensity of the diffraction plane for $\omega_3 = 0$ is used

Ab initio mutual intensity and $|f_p(x_1, x_2)|$ of the projection are determined (Yuen (2011k), Sect. 2.6.1, (B)).

Projection to a plane containing the fibre axis:

(A) Diffraction intensity of the diffraction lines for $\omega_1 = 0$ is used

Ab initio $f_h(x_{o2}, x_{o3})$ of the projection is determined (Yuen (2011k), Sect. 2.6.2, (A)).

(B) Magnitude of the mutual intensity of the diffraction lines for $\omega_1 = 0$ is used

Ab initio mutual intensity and $|f_h(x_{o2}, x_{o3})|$ of the projection are determined (Yuen (2011k), Sect. 2.6.2, (B)).

Projection to the x_1-axis:

(A) Diffraction intensity of the diffraction line for $\omega_2 = \omega_3 = 0$ is used

Ab initio $f_n(x_1)$ of the projection is determined (Yuen (2011k), Sect. 2.6.3, (A)).

(B) Magnitude of the mutual intensity of the diffraction line for $\omega_2 = \omega_3 = 0$ is used

Ab initio mutual intensity and $|f_n(x_1)|$ of the projection are determined (Yuen (2011k), Sect. 2.6.3, (B)).

Projection to the x_3-axis, **the fibre axis:**

This corresponds to the one-dimensional concentration profile in Bunk et al. (2007b) and Bunk et al. (2008), when partially coherent wave is used.

(A) Diffraction intensity of the diffraction line for $\omega_1 = \omega_2 = 0$ is used

Ab initio $f_c(x_3)$ of the projection is determined (Yuen (2011k), Sect. 2.6.4, (A)).

(B) Magnitude of the mutual intensity of the diffraction line for $\omega_1 = \omega_2 = 0$ is used

Ab initio mutual intensity and $\left|f_c(x_3)\right|$ of the projection are determined (Yuen (2011k), Sect. 2.6.4, (B)).

Diffraction of coherent wave by an one-dimensional microcavity array, a fixed fibre:

(A) Diffraction intensity of the diffraction planes is used

A*b initio* $f(\mathbf{x}_o)$ of the fibre is determined (Yuen (2011k), Sect. 2.6.5, (B)).

(B) Magnitude of the mutual intensity of the diffraction planes is used

Ab initio mutual intensity and $\left|f(\mathbf{x}_o)\right|$ of the fibre are determined (Yuen (2011k), Sect. 2.6.5, (C)).

For both (A) and (B), powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011i)).

3.2. Two-dimensional microcavity array (a planar periodic membrane)

In the following about projections, we will apply Yuen (2011k), Sect. 2.7.1 and 2.7.2, modified for projections which may be non-centrosymmetric or complex.

Projection to the x_1-axis, which is normal to the membrane:

(A) Diffraction intensity of the diffraction rod for $\omega_2 = \omega_3 = 0$ is used

Ab initio electron density profile $f_n(x_1)$ of the membrane is determined (Yuen (2011k), Sect. 2.7.1, (A)).

(B) Magnitude of the mutual intensity of the diffraction rod for $\omega_2 = \omega_3 = 0$ is used

Ab initio mutual intensity and $\left|f_n(x_1)\right|$ of the projection is determined (Yuen (2011k), Sect. 2.7.1, (B)).

The results of (A) and (B) should be the same as that in Sect. 3.1, employing one-dimensional microcavity array.

Projection to the plane of the membrane:

(A) Diffraction intensity of the diffraction points for $\omega_1 = 0$ is used

Ab initio $f_h(x_{o2}, x_{o3})$ of the projection is determined (Yuen (2011k), Sect. 2.7.2, (A)).

(B) Magnitude of the mutual intensity of the diffraction points for $\omega_1 = 0$ is used

Ab initio mutual intensity and $|f_h(x_{o2}, x_{o3})|$ of the projection are determined (Yuen (2011k), Sect. 2.7.2, (B)).

The results of (A) and (B) should be the same as that in Sect. 3.1, employing one-dimensional microcavity array.

Diffraction of coherent wave by a two-dimensional microcavity array, a planar periodic membrane:

(A) Diffraction intensity of the diffraction rods is used

Ab initio $f(\mathbf{x}_o)$ of the membrane is determined (Yuen (2011k), Sect. 2.7.3, (B)).

(B) Magnitude of mutual intensity of the diffraction rods is used

Ab initio mutual intensity and $|f(\mathbf{x}_o)|$ of the membrane is determined. (Yuen (2011k), Sect. 2.7.3, (C)).

For both (A) and (B), powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011j)).

Similar treatment for fixed macromolecules.

For quasi-monochromatic incoherent radiation used as the wave incident on the microcavity arrays, see Yuen (2011m), Sect. 2.5, 2.6.

For quasi-monochromatic partially coherent radiation, see Yuen (2011n), Sect. 2.5, 2.6.

For quasi-monochromatic partially coherent radiation formed by a combination of a coherent wave and an incoherent wave, see Yuen (2011o), Sect. 2.6, 2.7.

4. Discussions and conclusion

It is suggested that the microcavity arrays of Diaz et al. (2005), Bunk et al. (2007a), Bunk et al. (2007b) and Bunk et al. (2008) may be arranged as one-dimensional or two-dimensional arrays of freely rotating or fixed macromolecules (Sect. 2.1, 2.2, 2.3). One great advantage of the microcavity arrays is that the in-between | Fourier coefficients | in the diffraction plane for one-dimensional array, or along the diffraction rods for the two-dimensional array, can be measured; no interpolation is needed. The results obtained are exact. The tight supports of the sharp boundaries, and the constant electron densities of the solvent, silicon walls and glass slides help the iterations to converge to a unique electron density. This offers unique opportunities for determination of *ab initio* electron densities of macromolecules in solution. Knowledge of the electron density is important in understanding the active sites and biological activities of the macromolecules. From this electron density, the atomic species, chemical formula, and atomic coordinates of the macromolecule in the basic unit cell are determined.

For the general case of non-centrosymmetric or complex object, the solution of the one-dimensional phase-retrieval problem may not be unique (Yuen (2011a), Sect. 1, Sect. 2.1.1, (B)). Even though it is unique for a centrosymmetric real object, the iterative algorithms may or may not converge to a unique non-negative electron density (Yuen (2011b), Sect. 2.6, 2.8, Yuen (2011d), Sect. 2.2). For centrosymmetric real or the general case of non-centrosymmetric or complex object, to help the iterative algorithms to converge to a unique non-negative electron density, very strong container constraint and solvent constraint are used, as in microcavity arrays.

Powerful methods in X-ray crystallography, such as sharpening of Patterson function, direct methods, point-atom model, accelerated convergence of extension of Bragg intensities and structure factors, and sharpening in reciprocal space, can be applied to structure determination of one-dimensional and two-dimensional arrays of fixed macromolecules (Yuen (2011h, 2011i, 2011j)).

Quasi-monochromatic coherent, incoherent, or partially coherent wave may be used as wave incident on microcavity arrays (Sect. 3). A quasi-monochromatic partially coherent radiation may be formed by a combination of a coherent wave and an incoherent wave. Fibre diffraction and membrane diffraction are dealt with.

Before working with macromolecules, and as preparatory studies, it will be very interesting to apply Sect. 3 to study the concentration distribution of the colloid solutions in Bunk et al. (2007b) and Bunk et al. (2008):

One-dimensional microcavity array (a fixed fibre)

One-dimensional projections to fibre axis or to an axis normal to the fibre axis, two-dimensional projections to a plane containing the fibre axis or normal to the fibre axis, and three-dimensional concentration distribution.

Two-dimensional microcavity array (a planar periodic membrane)

One-dimensional projection normal to the membrane, two-dimensional projection to the plane of the membrane, and three-dimensional concentration distribution.

Quasi-monochromatic coherent, incoherent, partially coherent waves, or a combination of a coherent wave and an incoherent wave are used. Diffraction intensity and magnitude of the mutual intensity of the diffraction plane or diffraction rods are measured.

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Yuen, P. S. (2011o). Imaging of object irradiated with a quasi-monochromatic partially coherent wave which is a combination of a coherent wave and an incoherent wave. Direct and inverse problems, lensless imaging, phase-retrieval microscope, phase-retrieval telescope, lenslets microscope and lenslets telescope, crystal diffraction, fibre diffraction, membrane diffraction. (This book, **II**, pp. 137-164).

Index

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PHASE-RETRIEVAL METHOD

AND

APPLICATIONS

Volume II

Applications: Optics and Miscellaneous

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Preface

The magnitude of the wave diffracted from an object can be measured, but not the phase. If the phase is known, the structure of the object can be determined. By applying general and reasonable constraints on the object, the phase can be obtained from the magnitude. The solution is unique or very probably unique. The phase is not lost; it is just hidden in the magnitude.

Phase-retrieval approach has numerous applications. The structure of all molecules in a human body is essential to understanding biological processes, diseases and developing suitable medicine. In fact, many disciplines involve wave or particle scattered from an object, phase-retrieval approach will be useful.

I have completed three articles about basic principles of phase-retrieval method, and four articles about applications to optical scattering, acoustical scattering, thermal image, seismology, earthquake, tsunami and nuclear physics. Instead of publishing them in journals, I choose to publish a book so that the publication will not be delayed. I think the method and the applications are very important. The sooner they are published the better. If they are published one day earlier, many more lives will be saved. These articles are not peer-reviewed. Because of my very limited knowledge, there are bounded to be many errors. I would be very grateful if readers of this book can inform me of the errors (use the Forum in this website).

As a researcher, I hope my discovery will benefit all mankind. In fact, it is The Lord’s grace through an unworthy person.

> I declare that now I give the intellectual property of all my discoveries in this book to all mankind including those born in future. The intellectual properties of other people are not affected.

I hope no one will use my discoveries in this book to apply for any patent.

Pui Sum Yuen
December 19, 2007

Preface to the 2011 Edition

Since the book first published in 2007, it has many revisions. Articles are deleted and added. It is now divided into two volumes.

Volume I. Fundamentals and X-ray Crystallography. A systematic development of fundamentals of phase-retrieval method is presented. It provides the theoretical foundation and gives the conditions that iterative algorithms with oversampling will give a unique or probably unique solution. Methods in X-ray crystallography, with and without using in-between｜Fourier coefficients｜, are presented. These are applied to structure determination of crystals, nonperiodic objects, fibres and planar periodic membranes.

Volume II. Applications: Optics and Miscellaneous. It presents imaging of object irradiated with quasi-monochromatic coherent, incoherent, or partially coherent waves. The direct and inverse problems are dealt with. Unique decomposition of quasi-monochromatic partially coherent waves into coherent and incoherent components is presented. Imaging with partially coherent wave can then be dealt with in terms of the coherent and incoherent components. The direct and inverse problems of imaging of object emitting polychromatic coherent, incoherent or partially coherent waves are presented. For imaging of object emitting polychromatic partially coherent wave, coherent and incoherent components may be employed. Because of the small size of the nuclei, phase-retrieval method is meaningful. An article is presented for determining nuclear structure and force by means of phase retrieval in scattering.

For readers to inform me of the errors of this book, or suggestions, comments and criticisms, please use e-mail address: puisumyuen@netvigator.com . Thanks.

Pui Sum Yuen
November 2011

Contents

Object irradiated with quasi-monochromatic wave

1

Imaging of object irradiated with quasi-monochromatic coherent wave.

Direct and inverse problems, lensless imaging, phase-retrieval microscope, phase-retrieval telescope, transforming a nonperiodic object (a molecule, a cell, an astronomical object) to a two-dimensional crystal, lenslets microscope and lenslets telescope, crystal diffraction, fibre diffraction, membrane diffraction

First published April 18, 2008

Revised November 29, 2011

This article deals with the direct and inverse problems of imaging of object irradiated with quasi-monochromatic coherent light wave. Diffraction intensity, mutual intensity or radiant intensity is dealt with. Phase-retrieval method by means of iterative algorithms with oversampling is applied to the inverse problem. For lensless imaging, use an incident quasi-monochromatic waves with mean angular frequency $\overline{\omega}$. First Born approximation is used. All orientations of the nonperiodic object are used to get all diffraction intensity and magnitude of the mutual intensity in the far field of the object according to the chosen resolution. Using the real image of a nonperiodic object in a lens, superresolution is achieved. Magnitude of the mutual intensity and the radiant intensity in the far field of the image are measured. Use all orientations of the object and use separately all incident quasi-monochromatic waves of same intensity with mean wave number $\overline{k}$, $\overline{k} \leq k_{\max}$. Phase retrieval is applied to the $|G(\overline{k}\mathbf{u})|$ and the magnitude of the mutual intensity. These are phase-retrieval microscope and phase-retrieval telescope. The object can be obtained by means of Fourier optics with the empirical amplitude transfer function or generalized transfer function. Natural resolution of the structure is dealt with. The Sparrow criterion of resolution (or alike) of the image and the corresponding angular resolution are by-passed. The natural resolution may be comparable to the resolution of the image ($=\overline{\lambda}/2$), and is higher or equal to the resolution provided by the Sparrow criterion. The empirical amplitude transfer function includes aberrations of the lens and other

defects of the system. By means of array of lenslets, a nonperiodic object (a cell, a molecule, an astronomical object) is transformed to a crystal. The weak radiant intensity or mutual intensity of each image is collected into strong diffraction rods which can be measured accurately. Short exposure of weak radiation can be used to avoid damage to the cell or molecule. During each short exposure, some diffraction rods are measured. 20 advantages are listed for lenslets telescope employing radiant intensity. These include minimization or elimination of seeing, determination of three-dimensional surface or volume structure of smaller, fainter and more distant astronomical objects, and inexpensive thin lenslets of long focal length for reduced aberrations and high resolution. Advantages for lenslets telescope employing mutual intensity are similar. Most of these advantages are shared by lenslets microscope. Lenslets microscope and lenslets telescope are very useful in single molecule imaging and astronomy. By dealing with the image in a lens or lenslets, phase-retrieval microscope, phase-retrieval telescope, lenslets microscope and lenslets telescope are applicable whether the first Born approximation is applicable or not. Diffraction of quasi-monochromatic coherent wave by a nonperiodic object, or crystal diffraction, fibre diffraction, and membrane diffraction are dealt with. Powerful methods in X-ray crystallography can also be applied. The presentation is based on Fourier optics. It also applies to Fourier acoustics and matter-wave imaging.

Keywords: Phase retrieval; iterative algorithms with oversampling; diffraction intensity; mutual intensity; radiant intensity; nonperiodic object; lensless imaging; first Born approximation; superresolution by means of a lens; natural resolution; Sparrow criterion of resolution; empirical amplitude transfer function; generalized transfer function; aberrations; system defects; single molecule imaging; astronomy; seeing; crystal diffraction; diffraction points; fibre diffraction; diffraction planes; projections; membrane diffraction; diffraction rods; Bragg rods; electron density profile; optical wave; acoustic wave; matter wave; electromagnetic wave; X-ray; electron wave; ion wave; neutron wave; Fourier optics; Fourier acoustics.

1. Introduction

This article deals with the direct and inverse problems of imaging of object irradiated with quasi-monochromatic coherent light wave. Diffraction intensity, mutual intensity and radiant intensity are dealt with. Phase-retrieval method by means of iterative algorithms with oversampling is applied to the inverse problem. Here and in the following articles, iterative algorithms with oversampling in Yuen (2011b - 2011g, 2011j - 2011s) are applied to objects satisfying constraints in Yuen (2011a, 2011h or 2011i).

Lensless imaging is treated in Sect. 2.1. The first Born approximation is used. For the inverse problem, phase retrieval is used for the radiant intensity or mutual intensity in the far field of the object. Lensless imaging is discussed in Spence et al. (2002), Azaña (2003), Miao et al. (2004), Chapman et al. (2006), Rodenburg et al. (2007), Vartanyants et al. (2007) and Rodenburg (2008).

Diffracted wave from a distant object can only be measured within a small cone. The problem is to recover the other diffracted waves for this fixed wavelength and to determine the structure of the object. This problem is approached via the real image of the nonperiodic object in a lens, which leads to superresolution (Sect. 2.2). The first Born approximation is not used. A lens is simple in design, but it is very useful (see, for example, Zalevsky & Mendlovic (2004), p. 35).

The basic concepts of superresolution are presented in Bertero & Boccacci (2003) and Sheppard (2007). Peřina (1971), Peřina et al. (1977) and Peřina (1985), Sect. 9.2 treat superresolution based on Taylor series. Superresolution in coherent X-ray diffraction microscopy is discussed in Takahashi et al. (2007).

For the inverse problem, phase retrieval is used for the radiant intensity or mutual intensity in the far field of the image. The object can be obtained by means of Fourier optics with the empirical amplitude transfer function or generalized transfer function. Natural resolution of the structure is dealt with. The Sparrow criterion of resolution (or alike) and the corresponding angular separation are by-passed. The natural resolution may be comparable to the resolution of the image. The resolution of the image ($=\overline{\lambda}/2$) is equal to or higher than the diffraction limited resolution of conventional microscope and telescope. In conventional microscope and telescope, one of the most important characteristics is the quality of the image formed. The Sparrow criterion of resolution or alike is applied to the image. The corresponding angular separation is then applied to the object. Fourier optics and imaging are presented in Reynolds et al. (1989), Barrett & Myers (2004), Goodman (2005), Ersoy (2007), and Bertero & Boccacci (2002).

In this article, phase-retrieval method is applied to $\left|G(\mathbf{K})\right|$ or the magnitude of the mutual intensity in the far field of the image in microscope and telescope. These are called phase-retrieval microscope or phase-retrieval telescope (Sect. 2.2). The proposed phase-retrieval microscope or phase-retrieval telescope works. This is first verified by the electron diffraction microscope (Kamimura et al. (2010)). This is different from Coene & Janssen (1992), Thust et al. (1996), Luke et al. (2002), van Dam & Lane (2002), van Noort et al. (2005), Petersen & Keast (2007), Rondeau et al. (2007), Yin et al. (2007), Paganin & Gureyev (2008) and Smith (2008), which deal with the phase retrieval in the stage of the wave exit from the object to the image.

Three-dimensional surface or volume structure of the object including the size (shape and dimensions) is obtained. The diffraction intensity or radiant intensity and mutual intensity can always be measured even if the image is small. An eyepiece is not needed. As mentioned in Paganin & Gureyev (2008), a perfect conventional microscope without aberration cannot be used to measure a phase object. A phase-contrast microscope is used for this purpose (see, for example, Goodman (2005), Sect. 8.1.2, Kawata et al. (1996), Liang et al. (2000) and Kohmura et al. (2001)). The phase-retrieval microscope and phase-retrieval telescope applies to both real and complex objects, including phase object.

The configuration of a molecule in solution is different from that in a crystal. The biological or chemical properties of a molecule depend on its configuration. It is important to determine the structure of a molecule in solution, as mentioned in Yuen (2011a). The scattered intensity in diffraction studies is weak. If a strong laser beam is used, the molecule may be damaged. The exposure time needs to be short. Ram et al. (2008) has developed multifocal plane microscopy for 3D single molecule tracking. Webb et al. (2006) presents a wide-field total-internal-reflection fluorescence microscope for single molecule imaging, while Chung et al. (2007) uses standing wave total internal reflection fluorescence microscopy, and Michalet et al. (2007) presents investigation of detectors. A chemically modified tip of a scanning tunneling microscope is presented in Hahn & Ho (2001). A fundamental resolution measure is presented in Ram et al. (2007). Moerner (2007) presents a review.

By means of a lenslets microscope, a molecule can be transformed to a two-dimensional crystal (Sect. 2.3). In the direct problem, relative to the background, the radiant intensity and the magnitude of the mutual intensity of the diffraction rods (Bragg rods) for the crystal can be measured accurately in a short time. Short exposures of a molecule to weak incident radiation can be used in order not to damage the molecule. During each short exposure, some of the diffraction rods are measured. Not the radiant intensity or magnitude of the mutual intensity in all directions, only the diffraction rods need to be measured. If the aberrations of the lenslets are random, not systematic, the effect of the aberrations of the lenslets upon the values of the diffraction rods may be negligible. In the inverse problem, phase-retrieval method is then used to obtain the phase from the magnitude of the diffraction rods. From the radiant intensity, the object is determined. Natural resolution of the object is used. The image is not used explicitly. The Sparrow criterion of resolution (or alike) of the image and the corresponding angular resolution are by-passed. In the inverse problem using the magnitude of the mutual intensity of the diffraction rods, approximate value of the intensity of the image in one lenslet is obtained. The Sparrow criterion of resolution (or alike) of the approximate intensity of the image and the corresponding angular resolution are applied. Natural resolution of the object is not dealt with. We use the advantage of measuring diffraction rods from a two-dimensional periodic structure to determine the structure of a nonperiodic object. This also applies to an astronomical object in a lenslets telescope.

Ultraviolet microscope is discussed in Heimann & Urstadt (1990) and Minkel (2006).

X-ray microscope is discussed in Hambach et al. (2001), Matsuyama et al. (2006), Dudchik et al. (2007), Reznikova et al. (2007), Sakdinawat & Liu (2007), Kirz & Jacobsen (2009), and Sakdinawat & Attwood (2010).

A general discussion of conventional telescopes in presented in Davies (1997).

Radio telescope is discussed in Ilyasov (2006), Ellingson et al. (2007) and Olmi & Bolli (2007).

Infrared telescope is discussed in Busso et al. (2002), Werner et al. (2004), Gehrz et al. (2007) and Clampin (2008).

Optical telescope is discussed in Clampin (2008) and Benn (2001).

Ultraviolet telescope is discussed in Kruk et al. (1999), Korendyke et al. (2001) and Osterman et al. (2003).

Focusing X-ray and gamma-ray telescopes are discussed in Courtois et al. (2005), Krizmanic et al. (2005) and Knödlseder et al. (2007).

Focusing soft-X-ray telescope is discussed in Weisskopf et al. (2000), Jansen et al. (2001), Kitamoto et al. (2004) and DeLuca et al. (2005).

Focusing hard-X-ray telescope is discussed in Ramsey et al. (2002), Tueller et al. (2005) and Spiga (2007).

Focusing gamma-ray telescope is discussed in Smither et al. (2005), Barrière et al. (2007) and Barrière et al. (2007).

For astronomical imaging see, for example, Renaud et al. (2006) and Monnier et al. (2007).

Microlens arrays are employed in Bewersdorf et al. (1998) and Arai et al. (2006).

Diffraction with a coherent X-ray beam is reviewed in Livet (2007).

Matter wave is presented in Lenz et al. (1993), Nicklaus & Hasselbach (1993), Berman (1997), Nowak et al. (1997), Goldstein et al. (1998), Miesner et al. (1998), Silverman & Strange (1998), Goldstein et al, (1999), Barnett et al. (2000), Bloch et al. (2000), Carusotto et al. (2000), Denschlag et al. (2000), Esslinger et al. (2000), Nugent & Paganin (2000), Prataviera et al. (2000), Franke-Arnold et al. (2001), Köhl et al. (2001), Meystre (2001), Rohwedder (2001), Ketterle (2002), Anderson & Meystre (2003), Herbig et al. (2003), Bongs & Sengstock (2004), Arndt et al. (2005), Hornberger et al. (2005), Vainio et al. (2006), Chen et al. (2008), Koch et al. (2008) and Steuernagel (2009).

Diffraction and scattering of matter wave is presented in Faubel (1984), Liu (1989), Brukner & Zeilinger (1999), Oberthaler et al. (1999), van der Poel et al. (2001), Gammal & Kamchatnov (2004), Goldfarb et al. (2005), Kouznetsov & Oberst (2005), Stibor et al. (2005), Glionna et al. (2008), Lemeshko & Friedrich (2008), Lemeshko &

Friedrich (2009a, 2009b).

Lensless matter wave imaging is presented in Zarubin (1993, 1996a, 1996b).

The most important tool in astronomy is telescope. A conventional telescope employs a large lens to form a real image. Detector is placed at the image, or in optical telescope, an eyepiece is used to view the image. It is difficult to make very large optical lens, and very expensive (Buckley (2001)). Polishing the surfaces is difficult. The weight of the heavy lens may cause deformation. For a very distant astronomical object, the real image may be a bright spot. The resolution of the image $=\overline{\lambda}/2$. For very small object and very small wavelength $\overline{\lambda}$, it may be difficult to place a detector at the image to find the details of the image. The angular resolution is limited by $\overline{\lambda}$, and the diameter of the lens D. The effect of seeing (atmospheric turbulence) greatly reduces the quality of the image. Various approaches has been used to deal with this problem such as choosing appropriate site (Ardeberg & Andersen (1998)), seeing prediction (Vernin et al. (1998)), quantifying man-made seeing (Nicolas et al. (1998)) and speckle imaging (for example, Rondeau et al. (2007), Dainty & Fienup (1987), Roddier (1988), Horch (1995) and Gonsalves & Tolls (2005)). Adaptive telescope has sophisticated movement. The image is recorded as a whole. Infrared telescope has to be cooled to near absolute zero. Much cooling is needed for telescope with large lens.

The phase-retrieval telescope or lenslets telescope (Sect. 2.4) minimizes or avoids the problems in conventional telescope. The radiant intensity or the magnitude of the mutual intensity for the diffraction rods of the image in all the lenslets is measured. 20 advantages are listed for lenslets telescope using the radiant intensity. These include: natural resolution may be comparable to the resolution of the image ($=\overline{\lambda}/2$), and is equal to or higher than the resolution provided by the Sparrow criterion. The Sparrow criterion of resolution and the corresponding angular resolution are by-passed, the lenslets and the real images may not have good quality, while the structure of the object obtained may still have good quality, the effect of seeing is minimized or eliminated, gives three-dimensional surface or volume structure including shape and dimensions, structures of smaller, fainter and more distant astronomical objects can be determined, measurement of the diffraction rods is always possible, which can be measured one after another, data collected by different observatories can be combined, light-weighted, simple operation, inexpensive, etc. The advantages of lenslets telescope employing mutual intensity are similar, except that we have to deal with the Sparrow criterion of resolution of the approximate intensity of the image. Most of these advantages are shared by lenslets microscope.

By employing a lens or a number of lenslets in phase-retrieval microscope, phase-retrieval telescope, lenslets microscope or lenslets telescope, the image of a nonperiodic object is a secondary source, these microscopes and telescopes are applicable whether the first Born approximation is applicable or not.

The direct and inverse problems of crystal diffraction for quasi-monochromatic coherent wave are dealt with in Sect. 2.5. Diffraction points of the diffraction intensity or mutual intensity are used. $f(\mathbf{x}_o)$, mutual intensity and $|f(\mathbf{x}_o)|$ of the crystal are determined.

Determination of *ab initio* electron density, or *ab initio* mutual intensity and magnitude of the electron density of the projection of one period of a fibre (freely rotating) to a plane normal to the fibre axis is presented in Sect. 2.6.1. Sect. 2.6.2 deals with the projection to a plane containing the fibre axis. Sect. 2.6.3 deals with the projection to an axis normal to the fibre axis**.** Sect. 2.6.4 deals with the projection to the fibre axis. The direct and inverse problems of diffraction of coherent wave by a fibre (freely rotating) or a fixed fibre are dealt with in Sect. 2.6.5. *Ab initio* electron density, or *ab initio* mutual intensity and magnitude of the electron density of the fibre are determined. Diffraction planes of the diffraction intensity or mutual intensity are used.

Determination of *ab initio* one-dimensional centrosymmetric electron density profile of a membrane, or *ab initio* mutual intensity and magnitude of the electron density of the projection upon an axis normal to the membrane is presented in Sect. 2.7.1. Sect. 2.7.2 deals with the projection to the plane of the membrane. The direct and inverse problems of diffraction for coherent wave by a planar periodic membrane are dealt with in Sect. 2.7.3. *Ab initio* electron density, or *ab initio* mutual intensity and magnitude of the electron density of the membrane is determined. Diffraction rods of the diffraction intensity or mutual intensity are used.

For diffraction of coherent wave by a nonperiodic object, or crystal diffraction, fibre diffraction, and membrane diffraction, powerful methods in structure determination in X-ray crystallography, such as sharpening of Patterson function, direct methods, point-atom model, accelerated convergence of extension of Bragg intensities and structure factors, and sharpening in reciprocal space can also be applied.

This article may be applied to acoustic wave and matter wave, including electromagnetic wave, X-ray, electron, ion and neutron waves. For optical wave, acoustic wave and matter wave, including electromagnetic wave, X-ray, electron, ion and neutron waves, use Zarubin (1993), (4.2) for three-dimensional object and (5.9) for planar object.

2. Method

The temporal Fourier transform of a function $f(t)$ is given by

$$\int_{-\infty}^{\infty} f(t)\exp(-i\omega t)dt\,.$$

The spatial Fourier transform of a function $f(\mathbf{x})$ is given by

$$\int f(\mathbf{x})\exp(-i\mathbf{K}\cdot\mathbf{x})d\mathbf{x}\,.$$

Consider a quasi-monochromatic partially coherent scalar wave of mean angular frequency $\overline{\omega}$. $\overline{\omega}=c\overline{k}$. c is the velocity of light. $\overline{k}$ is the mean wave number. From Beran & Parrent (1964), (7-49) and (7-50), the mutual coherence function

$$\Gamma(\varsigma_1,\varsigma_2,\tau)\approx\Gamma(\varsigma_1,\varsigma_2,0)\exp(-i\overline{\omega}\tau) \tag{1}$$

with its temporal transform, the cross-spectral density function

$$W(\varsigma_1,\varsigma_2,\omega)\approx\Gamma(\varsigma_1,\varsigma_2,0)\delta(\omega-\overline{\omega}) \tag{2}$$

$$|\tau|<<2\pi/(\Delta\omega)$$

The mutual intensity $J(\varsigma_1,\varsigma_2)\equiv\Gamma(\varsigma_1,\varsigma_2,0)$

2.1. Lensless imaging. Direct and inverse problems

(A) Direct problem

Consider a quasi-monochromatic coherent scalar wave of mean wavelength $\overline{\lambda}$ incident on a real or complex finite-size planar or three-dimensional nonperiodic object $f(\mathbf{x}_o)$. $f(\mathbf{x}_o)$ satisfies the constraints in Yuen (2011a, 2011h or 2011i). The first Born approximation is used. The different waves can be treated on a common basis (Cowley (1990), Sect. 1.2, Zarubin (1993), p. 492).

Intensity of the object $I_o(\mathbf{x}_o) = \left| f(\mathbf{x}_o) \right|^2$ (3)

The mutual intensity of the object

$$J_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}) = f(\mathbf{x}_{o1}) f^*(\mathbf{x}_{o2}) \quad (4)$$

(Beran & Parrent (1964), (4-25), Mehta et al. (1966), (4.17), Peřina (1985), (4.14) and Mandel & Wolf (1995), (4.5-54)).

The diffraction pattern is given by the Fourier transform $F(\mathbf{K})$ of $f(\mathbf{x}_o)$.

$\mathbf{K}$ = wavevector of scattered wave – wavevector of incident wave

The diffraction intensity in the far field of the object,

$$I(\mathbf{K}) = \left| F(\mathbf{K}) \right|^2 \quad (5)$$

Use $\varsigma_1 = r\mathbf{u}_1$, $\varsigma_2 = r\mathbf{u}_2$ where $\mathbf{u}_1$, $\mathbf{u}_2$ are unit vectors, and $\mathbf{K}_1 = \overline{k}(\mathbf{u}_1 - \mathbf{u}_0)$, $\mathbf{K}_2 = \overline{k}(\mathbf{u}_2 - \mathbf{u}_0)$. $\overline{k}\mathbf{u}_0$ is the mean wavevector of the incident wave and $\overline{k}\mathbf{u}_1$, $\overline{k}\mathbf{u}_2$ are mean wavevectors of scattered waves. From Wolf (2007), p.118, (19), the mutual intensity in the far field of the object,

$$J(\varsigma_1, \varsigma_2) = (1/r^2) F^*(\mathbf{K}_1) F(\mathbf{K}_2) \quad (6)$$

The scattered wave is coherent (Wolf (2007), p. 117). The magnitude of the equal-time complex degree of coherence ≡ 1.

Neglecting the factor $1/r^2$, the intensity of the wave in the far field

$$J(\varsigma, \varsigma) = \left| F(\mathbf{K}) \right|^2 \quad (7)$$

which is (5).

The mean wavelength $\overline{\lambda}$ of the incident wave is chosen according to the resolution needed (Yuen (2011h, Sect. 2.3). A three-dimensional object can be rotated so that all diffraction intensity and the magnitude of all mutual intensity in the far field $|J(\varsigma_1,\varsigma_2)|$ for the chosen resolution can be measured

The inverse problem is a continuation of the direct problem.

(B) Inverse problem using the diffraction intensity in the far field of the object

The Fourier transform of a finite-size object extended to the complex plane is an entire function of exponential type. If the magnitude and phase of $F(\mathbf{K})$ in a small region of $\mathbf{K}$ is known, then, by analytic continuation, $F(\mathbf{K})$ for all $\mathbf{K}$, and hence the object $f(\mathbf{x}_o)$ are obtained. This is known as superresolution (Goodman (2005), pp. 162-167).

Iterative algorithms with oversampling in Yuen (2011c or 2011k) are applied to obtain the phase of $F(\mathbf{K})$ from the magnitude. Powerful methods in structure determination in X-ray crystallography, such as sharpening of Patterson function, direct methods, point-atom model, accelerated convergence of extension of Bragg intensities and structure factors, and sharpening in reciprocal space can also be applied (Yuen (2011q)). The solution $f(\mathbf{x}_o)$ is obtained.

The resolution of the object $=\overline{\lambda}/2$, (8)
(Zarubin (1993), p. 502, Totzeck & Krumbügel (1994), p. 189, Goodman (2005), (3-74)).

This lensless imaging method is called coherent diffraction imaging or diffraction microscopy (see, for example, Chapman (2006)).

(C) Inverse problem using the mutual intensity in the far field of the object

Fourier transform of $f^*(-\mathbf{x}_o)=F^*(\mathbf{K})$ (9)

$f(\mathbf{x}_o)$ satisfies the constraint of sharp boundary. From (6), $J(\varsigma_1,\varsigma_2)$ in the far field of the object, is a six-dimensional Fourier transform of $(1/r^2)f^*(-\mathbf{x}_{o1})f(\mathbf{x}_{o2})$. $f^*(-\mathbf{x}_{o1})f(\mathbf{x}_{o2})$ is a six-dimensional complex body (or a four-dimensional complex body for a planar object) with each of its real and imaginary components satisfying the constraints (Yuen (2011i), (C)):

(i) finite size
(ii) non-negative
(iii) bounded
(iv) sectionally continuous
(v) sharp boundary. The component function $\neq 0$ almost everywhere at the boundary surface of the six-dimensional body (or a four-dimensional complex body for a planar object)

The real and imaginary components satisfy the constraint

(vi) The size (shape and dimensions), and the regions of zeros (not isolated points) of the real part and the imaginary part are the same.

Iterative algorithms with oversampling in Yuen (2011k) are applied to obtain the phase of $J(\varsigma_1,\varsigma_2)$ from the magnitude. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011q)). The solution $f^*(-\mathbf{x}_{o1})f(\mathbf{x}_{o2})$ is obtained. Replace $-\mathbf{x}_{o1}$ by $\mathbf{x}_{o1}$. The mutual intensity of the object

$$J_o(\mathbf{x}_{o1},\mathbf{x}_{o2}) = [f^*(\mathbf{x}_{o1})f(\mathbf{x}_{o2})]^* = f(\mathbf{x}_{o1})f^*(\mathbf{x}_{o2}) \qquad (10)$$

is obtained.

For $\mathbf{x}_{o1} = \mathbf{x}_{o2} = \mathbf{x}_o$, intensity of the object

$$I_o(\mathbf{x}_o) = \left|f(\mathbf{x}_o)\right|^2 \qquad (11)$$

is obtained. This agrees with (3). $\left|f(\mathbf{x}_o)\right|$ is determined. This should agree with the $f(\mathbf{x}_o)$ obtained in (B), using the intensity of the scattered wave in the far field.

2.2. Image of object in a lens. Direct and inverse problems. Phase-retrieval

microscope, phase-retrieval telescope

Consider a quasi-monochromatic coherent scalar wave of mean angular frequency $\overline{\omega}$ incident on a real or complex finite-size planar or three-dimensional object $f(\mathbf{x}_o)$. For a distant object $f(\mathbf{x}_o)$, we can only measure $|F(\mathbf{K})|$ in a small region of $\mathbf{K}$. The phase cannot be measured. Hence analytic continuation cannot be applied to get $F(\mathbf{K})$ for all $\mathbf{K}$. The problem is to recover the other diffracted waves for this fixed wavelength and to determine the structure of the object. One approach is to use a physical method, a lens, to collect all the diffracted waves in this small region of $\mathbf{K}$ (see, for example, Beran & Parrent (1964), Ch. 7).

(A) Direct problem

This is a continuation of Sect. 2.1, Lensless imaging, (A) Direct problem.

The incident wave upon the object is along the axis of the lens. The wave (magnitude and phase) incident on the lens is transformed to a real image $g(\mathbf{x})$. Locate the origin of the co-ordinate system at the centre of the lens, with the object $f(\mathbf{x}_o) \equiv f(x_{o1}, x_{o2}, x_{o3})$ on the negative-x_3 axis. Follow Beran & Parrent (1964), p. 101 and (7-5), and Born & Wolf (1989), p. 481. The coordinates in the image plane are normalized so that an object point and its Gaussian image point have the same x_1, x_2 coordinate numbers.

The image $g(\mathbf{x})$ of the coherent object is coherent (Beran & Parrent (1964), p. 109, (7-33)). The mutual intensity of the image

$$J_i(\mathbf{x}_1, \mathbf{x}_2) = g(\mathbf{x}_1) g^*(\mathbf{x}_2) \tag{12}$$

(Beran & Parrent (1964), (4-25), Mehta et al. (1966), Carter & Wolf (1981), (4.1), Peřina (1985), (4.14) and Mandel & Wolf (1995), (4.5-54)).

The impulse response $h(\mathbf{x})$ is proportional to the Fourier transform of the pupil function (Goodman (2005), (5-40), (6-20)). To produce high-quality image, the impulse response is close to a δ function (Goodman (2005), (5-24)). The pupil function is a two-dimensional function.

The image

$$g(\mathbf{x}) \equiv g(x_1, x_2, x_3) = f(\mathbf{x}) \otimes h(\mathbf{x}) = f(x_1, x_2, x_3) \otimes h(x_1, x_2)$$

$$= \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} f(t_1, t_2, x_3) h(x_1 - t_1, x_2 - t_2) dt_1 dt_2 \tag{13}$$

(Goodman (2005), (5-38)).

$$G(\mathbf{K}) \equiv G(K_1, K_2, K_3) = F(\mathbf{K}) H(\mathbf{K}) = F(K_1, K_2, K_3) H(K_1, K_2) \tag{14}$$

where $G(\mathbf{K})$ is the Fourier transform of $g(\mathbf{x})$.

The amplitude transfer function $H(\mathbf{K}) \equiv H(K_1, K_2)$ is the Fourier transform of $h(\mathbf{x})$

(Beran & Parrent (1964), p. 111).

The cutoff frequency of the lens $= D/(2\bar{\lambda} x_3)$ (15)

(Ersoy (2007), p. 165)

The diameter of the lens D and the image distance x_3 are chosen so that $2\pi D/(2\bar{\lambda} x_3) > \bar{k}$.

The frequency-dependence of $h(\mathbf{x})$ can be omitted.

From (12), the intensity of the image, $I_i(\mathbf{x}) = J_i(\mathbf{x}, \mathbf{x}) = |g(\mathbf{x})|^2$ (16)

If the image is large enough, the intensity and the magnitude of the mutual intensity of the image can be measured.

As the lens has finite size, $h(\mathbf{x})$ has infinite extent. Hence because of diffraction, the real image $g(\mathbf{x})$ actually covers the whole image plane. (Note that the image of a three-dimensional centrosymmetric object in a lens is not strictly centrosymmetric). The image $g(\mathbf{x})$ of the coherent object acts as a secondary source (Wolf & Carter (1978), p. 953). The first Born approximation is not used. In the far field of the image, the cross-spectral density function is given in Carter & Wolf (1981), (4.8) for three-dimensional object (or Wolf & Carter (1978), (3.16) for planar object),

$$W(\varsigma_1 \mathbf{u}_1, \varsigma_2 \mathbf{u}_2, \bar{\omega}) = (2\pi)^6 G(\bar{k}\mathbf{u}_1) G^*(\bar{k}\mathbf{u}_2) \{\exp[i\bar{k}(\varsigma_1 - \varsigma_2)]\} / (\varsigma_1 \varsigma_2)$$

(17)

$\mathbf{u}_1$, $\mathbf{u}_2$ are unit vectors.

$\varsigma_1\mathbf{u}_1$ and $\varsigma_2\mathbf{u}_2$ denote points in the far field.

The radiant intensity in the far field of the image,

$$\Lambda(\mathbf{u},\overline{\omega}) = (2\pi)^6 \left|G(\bar{k}\mathbf{u})\right|^2, \tag{18}$$

(Carter & Wolf (1981), (4.9) for three-dimensional object, or Wolf & Carter (1978), (3.17) for planar object). $\mathbf{u}$ is a unit vector. The radiant intensity and the magnitude of the mutual intensity in the far field can be measured. In (18), the region of $\mathbf{u}$ of the image predicted by geometrical optics $f(\mathbf{x})$ is larger than the small region of $\mathbf{u}$ when the diffracted wave from the object is measured directly. This is one kind of superresolution. It is different from the superresolution by extrapolation in Yuen (2011h), Sect. 2.3. Similarly, there is superresolution for the mutual intensity. Moreover, the real image $g(\mathbf{x})$ covers the whole image plane. The information of the object is contained in the real image. In a sense, we may deal with the real image, instead of the object.

For a three-dimensional object, use all orientations of the object. Depending on the chosen resolution, we use a maximum mean wave number $\bar{k}_{\max}$. For a planar or three-dimensional object, use separately all incident quasi-monochromatic waves of same intensity with mean wave number $\bar{k}$, $\bar{k} \le \bar{k}_{\max}$. The radiant intensity and the magnitude of the mutual intensity in the far field of the image are measured.

(B) Inverse problem using the radiant intensity in the far field of the image

From (18), $\left|G(\bar{k}\mathbf{u})\right|$ can be determined.

In practice, only a truncated portion of the real image $g(\mathbf{x})$ can be dealt with. It has sharp boundary

Iterative algorithms with oversampling in Yuen (2011c or 2011k) are applied to obtain the phase of $G(\bar{k}\mathbf{u})$ from the magnitude. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011q)). The solution $g(\mathbf{x})$, the truncated real image, is obtained. The intensity of the image should agree with the value measured in (A).

Substitute the $G(\bar{k}\mathbf{u})$ determined into (17). The mutual intensity obtained from (2) should agree with the experimentally measured magnitude in the far field of the image.

The traditional approach is to make use of the property of a lens and the intensity of the image $g(\mathbf{x})$. The Sparrow criterion of resolution of the image (or alike) and the corresponding angular resolution are applied. Good quality of the image is needed. The aberrations of the lens and other defects of the system and their effects can be treated as refinement. Taking account of these, an approximate intensity of the image predicted by geometrical optics, $\left|f(\mathbf{x})\right|^2$ (approx.) is obtained. From $\left|f(\mathbf{x})\right|^2$ (approx.), $\left|f(\mathbf{x}_o)\right|^2$ (approx.) and, $\left|f(\mathbf{x}_o)\right|$ (approx.) are obtained.

A better wavy is to use $G(\bar{k}\mathbf{u})$ directly. The image $g(\mathbf{x})$ is not used. This is case (C) of Yuen (2008).

Let $\mathbf{K} = \bar{k}\mathbf{u}$

From (14), $F(\mathbf{K}) = G(\mathbf{K})/H(\mathbf{K})$ (19)

$f(\mathbf{x})$ is determined from $F(\mathbf{K})$. Hence $f(\mathbf{x}_o)$ is obtained.

The mutual intensity of the object

$$J_o(\mathbf{x}_{o1},\mathbf{x}_{o2}) = f(\mathbf{x}_{o1})f^*(\mathbf{x}_{o2}) \tag{20}$$

(see (12)).

The empirical amplitude transfer function $H(\mathbf{K})$ determined in Sect. 2.3 of Yuen (2008) will be used. It takes into account aberrations of the lens and other defects of the system. The lens and the image $g(\mathbf{x})$ may not have good quality, while the structure of the object $f(\mathbf{x}_o)$ obtained via $f(\mathbf{x})$ determined from (19) may still have good quality. This is particularly important in the case of using an imperfect X-ray microscope to determine the structure of a single molecule, single-molecule imaging. Since the image $g(\mathbf{x})$ is not used explicitly, its quality is not our main concern. We have by-passed the two-point Sparrow criterion of resolution (or alike) of the image, and the corresponding angular resolution (e.g., Reynolds et al. (1989), Ch. 13, 19). Even though the image $g(\mathbf{x})$ cannot be resolved into two peaks (poor angular resolution according to the Sparrow criterion of resolution of the image), but because of the other parts of $g(\mathbf{x})$, the solution of the object $f(\mathbf{x}_o)$ may be resolved into two points. Two-point resolution of the image just deals with the resolution of the peaks. By using $G(\mathbf{K})$, in effect we use the whole image $g(\mathbf{x})$ for determining the resolution of the structure obtained. What is important is the resolution of the structure obtained, not the resolution of the image $g(\mathbf{x})$, for example, choosing two points or using variance (Barrett & Myers (2004), pp. 299- 301). This resolution is the 'natural resolution' as it is natural to deal with the resolution of the structure obtained, not the image $g(\mathbf{x})$ (Reynolds et al. (1989), Sect. 19.3, 19.4). Superresolution by sharpening the central lobe of the real image, apodization of the side-lobes, pushing the side-lobes far away from the focal region, or localization of a wave is not needed (Sheppard (2007)). The natural resolution is equal to or higher than the resolution provided by the Sparrow criterion. Conceptually, this treatment is more complete, and is the beauty of Fourier optics.

Microscope and telescope employing phase-retrieval for imaging may be called phase-retrieval microscope and phase-retrieval telescope. As the structure of the object is determined by measuring the radiant intensity of its real image, the working principle is the same in phase-retrieval microscope and phase-retrieval telescope.

In applying the phase-retrieval method to a complex object, waves from the image in all directions have to be measured. For microscope, the object can be rotated. For an astronomical object, a lens telescope and a mirror telescope can be used. The data from these two telescopes can be combined.

At present, we do not have value of the natural resolution for phase-retrieval microscope. To get an idea of the natural resolution, we can compare with the resolution of the image $g(\mathbf{x})$ which is equal to $\overline{\lambda}/2$ (see (8)). The resolution distance on the object $=\overline{\lambda}/(2|M|)=\overline{\lambda}|x_{3o}|/(2x_3)$. The natural resolution may be comparable to this resolution distance.

If we use the resolution of the image for the phase-retrieval telescope, the angular resolution $=\overline{\lambda}/(2f)$, where f is the focal length of the lens. Unlike the conventional telescope, this depends on f, but not on D. If large value of f is used, the angular resolution is very small.

The resolution distance on the object $=\overline{\lambda}/(2|M|)=\overline{\lambda}|x_{3o}|/(2f)$

According to the resolution needed, the wavelengths of the waves employed may or may not be in the visible region. As detector is used to measure the radiant intensity, all wavelengths are treated alike. The radiant intensities need not be measured simultaneously. The resolution of the real image $=\overline{\lambda}/2$. For very small object or $\overline{\lambda}$, it may be difficult to place a detector at the image to measure the image directly as in conventional microscope. But the radiant intensity of the real image can always be measured (just like measuring intensity of diffracted wave from a molecule). The object can be real or complex, for example, a phase object (compare with the phase-contrast microscope).

As the image is not dealt with, we have avoided the problems for object with sharp edges: ringing and sharp-edge location from intensities of coherent image. Scanning confocal microscopy may not be needed as, for phase-retrieval microscope, the empirical amplitude transfer function includes the effect of the rays from other points of the object. Superficially we may think that we need not deal with the depth of image, depth of focus and defocus in electron microscopy. This needs to be investigated in practice.

For incident wave upon the object which is not along the axis of the lens, the phase for oblique incidence is included as a factor of the object.

(C) Inverse problem using the mutual intensity in the far field of the image

From (2) and (17), apart from the factor $(2\pi)^6 \{\exp[i\bar{k}(\varsigma_1 - \varsigma_2)]\}/(\varsigma_1\varsigma_2)$, the mutual intensity in the far field of the image,

$$J(\varsigma_1,\varsigma_2) \equiv \Gamma(\varsigma_1,\varsigma_2,0) = (2\pi)^6 \{\exp[i\bar{k}(\varsigma_1 - \varsigma_2)]\} G(\bar{k}\mathbf{u}_1) G^*(\bar{k}\mathbf{u}_2)/(\varsigma_1\varsigma_2) \tag{21}$$

is the six-dimensional (or four-dimensional for planar object) spatial Fourier transform of $g(\mathbf{x}_1)g^*(-\mathbf{x}_2)$ of the image (see (9)).

$g(\mathbf{x}_1)g^*(-\mathbf{x}_2)$ is a six-dimensional complex body (or a four-dimensional complex body for a planar object) with each of its real and imaginary components satisfying the constraints in Sect. 2.1, (C). Follow Sect. 2.1, (C). Iterative algorithms with oversampling in Yuen (2011k) are applied to obtain the phase of $J(\varsigma_1,\varsigma_2)$ from the magnitude. Constraint (v) of sharp boundary is used because, in practice, only a truncated portion of the image can be dealt with. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011q)). The solution $g(\mathbf{x}_1)g^*(-\mathbf{x}_2)$ is obtained. Replace $-\mathbf{x}_2$ by $\mathbf{x}_2$. The mutual intensity of the image

$$J_i(\mathbf{x}_1,\mathbf{x}_2) = g(\mathbf{x}_1)g^*(\mathbf{x}_2) \tag{22}$$

is determined. Its magnitude should agree with the value measured in (A).

Substitute $g(\mathbf{x}_1)g^*(\mathbf{x}_2)$ determined into (18). The radiant intensity in the far field is obtained. The result should agree with the value measured in (A).

For $\mathbf{x}_1 = \mathbf{x}_2 = \mathbf{x}$, the intensity of the image,

$$I_i(\mathbf{x}) = |g(\mathbf{x})|^2 \tag{23}$$

$|g(\mathbf{x})|$ is obtained.

Use a generalized transfer function (Beran & Parrent (1964), p. 112),

$$L(\mathbf{K}_1,\mathbf{K}_2) = H(\mathbf{K}_1)H^*(-\mathbf{K}_2) \tag{24}$$

$$\tilde{J}_i(\mathbf{K}_1,\mathbf{K}_2) = L(\mathbf{K}_1,\mathbf{K}_2)\tilde{J}_o(\mathbf{K}_1,\mathbf{K}_2) \tag{25}$$

where $\tilde{J}_i(\mathbf{K}_1,\mathbf{K}_2)$ is the spatial Fourier transform of $J_i(\mathbf{x}_1,\mathbf{x}_2)$ obtained in

(22),

$\widetilde{J}_o(\mathbf{K}_1,\mathbf{K}_2)$ is the spatial Fourier transform of $J_o(\mathbf{x}_1,\mathbf{x}_2)$.

Mutual intensity of the object $J_o(\mathbf{x}_{o1},\mathbf{x}_{o2})$ is obtained.
The intensity of the object $I_o(\mathbf{x}_o)=J_o(\mathbf{x}_o,\mathbf{x}_o)$. (26)

This should equal to the intensity of object $=\left|f(\mathbf{x}_o)\right|^2$ in (3). Natural resolution of the object is dealt with. Empirical amplitude transfer function $H(\mathbf{K})$ is used. It takes into account aberrations of the lens and other defects of the system. We have by- passed the Sparrow criterion of resolution (or alike) of the image, and the corresponding angular resolution.

By employing a lens, the image of the object is a secondary source. Scattering of wave is not involved. Hence the first Born approximation is not used. Phase-retrieval microscope and phase-retrieval telescope are applicable whether the first Born approximation is applicable or not.

2.3. Transforming an object (a molecule, a cell, an astronomical object) to a two-dimensional crystal, lenslets microscope. Direct and inverse problems

(A) Direct problem

To determine the structure of a nonperiodic object $f(\mathbf{x}_o)$, for example, a fixed molecule, a biological cell or an astronomical object, the radiant intensity (see (18)) is weak. A two-dimensional array of a large number of lenslets in the x_1-0-x_2 plane can be used. This is called a lenslets microscope. The array of lenslets forms a linear system. Its impulse response is formed by adding replica of $h(\mathbf{x})\equiv h(x_1,x_2)$ for one lenslet successfully in x_1, x_2 to give a periodic function, the extended $h(\mathbf{x})$. The periods are chosen such that the Fourier coefficients of this extended $h(\mathbf{x})$ are nonzero (Yuen (2008), (D)).

The focal length of the lenslets, the object distance and the unit cell constants a,b of the extended $h(\mathbf{x})$ are chosen so that the image predicted by geometrical optics in one lenslet, $f(\mathbf{x})$, can be contained in one unit cell of the extended $h(\mathbf{x})$ (Yuen

(2008), (D), constraint (i)). The images predicted by geometric optics in all the lenslets is formed by adding replica of $f(\mathbf{x}) \equiv f(x_1, x_2, x_3)$ in one lenslet successfully in x_1, x_2 to give a periodic function, the extended $f(\mathbf{x})$, with same unit cell constants as the extended $h(\mathbf{x})$.

The image in all the lenslets is formed by adding replica of the coherent image $g(\mathbf{x}) \equiv g(x_1, x_2, x_3)$ in one lenslet successfully in x_1 and x_2 to give a periodic function, the extended $g(\mathbf{x})$, with the same unit cell constants as the extended $h(\mathbf{x})$.

For an incident wave of wave number $\bar{k}$ upon the object, the cross-spectral density function in the far field of an image is given by (17). The weak radiant intensity and cross-spectral densities for all the images are collected into strong diffraction rods of the image in all the lenslets. (In this book, we will use the term diffraction point, diffraction plane, or diffraction rod for diffraction intensity, radiant intensity, cross-spectral density function and mutual intensity). For N lenslets, the radiant intensities (see (18)) along these rods are N^2 that of one lenslet. Relative to the background, the radiant intensity and the magnitude of the mutual intensity $|J(\varsigma_1, \varsigma_2)|$ of these diffraction rods can be measured accurately. For a nonperiodic image, these in all directions have to be measured. For the array of images, only those for the diffraction rods need to be measured. For a three-dimensional object, use all orientations of the object. Depending on the chosen resolution, we use a maximum mean wave number $\bar{k}_{\max}$. For a planar or three-dimensional object, use separately all incident quasi-monochromatic waves of same intensity with mean wave number $\bar{k}$, $\bar{k} \le \bar{k}_{\max}$.

(B) Inverse problem using the radiant intensity of the diffraction rods of the image in all the lenslets

Use Yuen (2011h), (H). Iterative algorithms with oversampling in Yuen (2011g or

2011n) are applied to obtain the phase of $G(\bar{k}\mathbf{u})$ from $\left|G(\bar{k}\mathbf{u})\right|$ of the diffraction rods. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011s)). The solution, the extended $g(\mathbf{x})$ is determined. The intensity of this extended $g(\mathbf{x})$ = | extended $g(\mathbf{x})$ | ^{2}.

For each x_3, let $c_{glm}(x_3)$, $l,m = 0,\pm1,\pm2,...$, be the Fourier coefficients of the extended $g(\mathbf{x})$. These can be obtained from $G(\bar{k}\mathbf{u})$ of the diffraction rods.

The extended $g(\mathbf{x}) = \sum_{l,m=-\infty}^{\infty} c_{glm}(x_3)\exp[i(l\omega_{10}x_1 + m\omega_{20}x_2)]$ (27)

where $\omega_{10} = 2\pi/a$, $\omega_{20} = 2\pi/b$.

For each x_3, let $c_{flm}(x_3)$, $l,m = 0,\pm1,\pm2,...$, be the Fourier coefficients of the extended $f(\mathbf{x})$. Let c_{hlm}, $l,m = 0,\pm1,\pm2,...$, be the Fourier coefficients of the extended $h(\mathbf{x})$.

From (14) and Yuen (2008), (6), for periodic convolution,

$$c_{flm}(x_3) = \frac{c_{glm}(x_3)}{Uc_{hlm}} \qquad (28)$$

$$U = ab$$

The extended $f(\mathbf{x}) = \sum_{l,m=-\infty}^{\infty} c_{flm}(x_3)\exp[i(l\omega_{10}x_1 + m\omega_{20}x_2)]$ (29)

The object $f(\mathbf{x}_o)$ is determined.

Natural resolution of the object is dealt with. The image $g(\mathbf{x})$ is not used explicitly. The Sparrow criterion of resolution (or alike) of the image and the corresponding angular resolution are by-passed.

$h(\mathbf{x})$ contains the aberrations of the lenslets and other defects of the system. As a large number of lenslets are used, if the aberrations of the lenslets are random, not systematic, the effect of the aberrations of the lenslets upon the values of the diffraction rods may be negligible. This is very useful in the case of using an imperfect X-ray lenslets microscope to determine the structure of a single molecule.

We have used the advantage of measuring diffraction rods from a two-dimensional periodic structure to determine the structure of a nonperiodic object $f(\mathbf{x}_o)$.

For the advantages of lenslets microscope, see [1]-[3], [6]-[9], [12]-[19] of lenslets telescope in Sect. 2.4. In addition, as the diffraction rods need not be measured simultaneously, short exposures of a molecule to weak incident radiation can be used in order not to damage the molecule. During each short exposure, some of the diffraction rods are measured.

(C) Inverse problem using the mutual intensity of the diffraction rods of the image in all the lenslets

As in Sect. 2.2, (C), iterative algorithms with oversampling in Yuen (2011n) are applied to obtain the phase of the mutual intensity of the diffraction rods $J(\varsigma_1, \varsigma_2)$ of the image in all the lenslets from the magnitude. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011s)). The solution, $g_a(\mathbf{x}_1)g_a*(-\mathbf{x}_2)$ is obtained. $g_a(\mathbf{x})$ is the image in all the lenslets. $g_a(\mathbf{x})$ = the extended $g(\mathbf{x})$. Substitute $g_a(\mathbf{x}_1)g_a*(-\mathbf{x}_2)$ into (18). The value of the radiant intensity of the diffraction rods obtained should agree with the value measured in (A).

For $\mathbf{x}_1 = -\mathbf{x}_2 = \mathbf{x}$, the intensity of image in all the lenslets = $|g_a(\mathbf{x})|^2$ is obtained.

Approximate value of the intensity of the image in one lenslet can be obtained from the intensity of the image in all lenslets in one unit cell, in particular, for sufficiently large unit cell. The Sparrow criterion of resolution (or alike) of the approximate intensity of the image in one lenslet and the corresponding angular resolution are applied. Natural resolution of the object is not dealt with.

2.4. Application to astronomy. Lenslets telescope. Inverse problem

(A) Inverse problem using the radiant intensity of the diffraction rods of the image in all the lenslets

To determine the structure of an astronomical object with coherent illumination, phase-retrieval telescope with one lens may be employed. The advantages of phase-retrieval telescope are:

[1] The empirical amplitude transfer function $H(\mathbf{K})$ of a lens is used. It contains aberrations of the lens and other defects of the system. The lens and the image may not have good quality, while the structure of the object $f(\mathbf{x}_o)$ obtained may still have good quality. This is particularly useful for X-ray and gamma-ray lenses which are not perfect.

[2] The Sparrow criterion of resolution of the image and the corresponding angular resolution are by-passed. The natural resolution is equal to or higher than the resolution provided by the Sparrow criterion. The angular resolution depends on f, but not on D. If large value of f is used, the angular resolution is very small and very high resolution is achieved. Thin lens of small diameter may be used. Aberrations and effect of paraxial approximation is minimized.

[3] Works on the whole electromagnetic spectrum (radio, infrared, visible, ultraviolet, X-ray and gamma ray).

[4] Works day and night, depending on the wavelength used.

[5] Works through cloudy or dusty skies, rain or storms for radio or infrared waves.

[6] Gives three-dimensional surface or volume structure of the object including the size (shape and dimensions). Determination of volume structure is possible when radio wave or X-ray is used.

[7] Monochromatic wave is used; polychromatic effect is avoided.

[8] The resolution of the real image $=\overline{\lambda}/2$. For very small real image or $\overline{\lambda}$, it may be difficult to place a detector at the image to measure the image directly as in conventional telescope. But the radiant intensity of the real image can always be measured. An eyepiece is not needed.

[9] The radiant intensities need not be measured simultaneously.

[10] The data collected by different observatories can be combined to determine the structure of an astronomical object.

[11a] The effect of seeing (atmospheric turbulence) is minimized or even eliminated as

the real image is not recorded as a whole directly. Many telescopes can remain ground based; they need not be placed in space.

It is difficult to make very large lens, and very expensive. A lenslets telescope employing radiant intensity may be used. ‘Rather crude instrumentation can undertake reasonably accurate diffraction measurement’ (Rodenburg (2008), p. 94). The impulse response $h(\mathbf{x}) \equiv h(x_1, x_2)$ of the lenslets telescope is periodic and is chosen such that its Fourier coefficients are non-zero. The radiant intensity of the diffraction rods can be measured accurately. The phase of $G(\mathbf{K})$ is obtained by iterative algorithms with oversampling (Yuen (2011g or 2011n).

Besides the 11 advantages listed in above for phase-retrieval telescope, additional advantages for lenslets telescopes are:

[11b] Since the diffraction rods need not be measured simultaneously and the measurement time may not be too long if a large number of lenslets is used, the effect of seeing (atmospheric turbulence) is further minimized or even eliminated. During each time of small atmospheric turbulence, some of the diffraction rods are measured.

[12] Thin lenslets are used; less absorption, reduced aberrations and light-weighted. No deformation of the system due to the weight of the lenslets.

[13] Use lenslets of small diameter and long focal length. Effect of paraxial approximation is minimized. For example, the lenslets and the detector can be placed in two space-crafts. The manufacture of these lenslets is not difficult.

[14] Simple construction, no sophisticated moving components. It is static with easy operation. No sophisticated movement as that in adaptive optics is needed.

[15] The system is inexpensive and readily installed.

[16] If needed, mass production is possible which further reduces the cost.

[17] If the aberrations of the lenslets are random, not systematic, the effect of the aberrations of the lenslets upon the values of radiant intensity of the diffraction rods may be negligible.

[18] The radiant intensity of the real image formed by one lens is weak. In lenslets telescope, the scattering is coherent for all the unit cells. For N unit cells, the

radiant intensity of the diffraction rods is N^2 times that of one unit cell, mainly the image of the astronomical object formed by one lenslet. Relative to the background, the radiant intensity of the diffraction rods can be measured accurately in a shorter time (just like X-ray crystallography). Only radiant intensity of the diffraction rods needs to be measured. As mentioned in [6], high-resolution three-dimensional surface or volume structure of the object can be obtained.

[19] Structures including sizes (shapes and dimensions) of smaller, fainter and more distant astronomical objects can be determined.

[20] An infrared telescope employing a large lens emits infrared radiation. It has to be cooled to near absolute zero to minimize such radiation. Telescope employing an array of small lenslets is simple in construction and contains much less materials. Hence the infrared radiation from the telescope is much reduced.

(B) Inverse problem using the mutual intensity of the diffraction rods of the image in all the lenslets

The advantages of phase-retrieval telescope or lenslets telescope employing mutual intensity are similar to these 20 advantages, except [2]. Approximate value of the intensity of the image is obtained. The Sparrow criterion of resolution of the approximate intensity of the image (or alike) and the corresponding angular resolution are applied. Natural resolution of the object is not dealt with.

2.5. Crystal diffraction for quasi-monochromatic coherent wave. Direct and inverse problems

(A) Direct problem

Sect. 2.1 can be applied to diffraction of coherent wave by crystals, namely, Yuen (2011a, 2011h, 2011i), (B), (D). Diffraction intensity or magnitude of the mutual intensity of the diffraction points is measured.

(B) Inverse problem using the diffraction intensity of the diffraction points

Use the diffraction intensity of the diffraction points. For phase-retrieval method, see

Yuen (2011a, 2011h, 2011i), (B), (D). For iterative algorithms with oversampling, see Yuen (2011b or 2011j). Powerful methods in structure determination in X-ray crystallography, such as sharpening of Patterson function, direct methods, point-atom model, accelerated convergence of extension of Bragg intensities and structure factors, and sharpening in reciprocal space can also be applied (Yuen (2011o, 2011p)). $f(\mathbf{x}_o)$ of the crystal is determined.

(C) Inverse problem using the mutual intensity of the diffraction points

Use the magnitude of the mutual intensity of the diffraction points. For phase-retrieval method, see Yuen (2011h, 2011i), (B), (D). Iterative algorithms with oversampling in Yuen (2011j) are applied. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011o, 2011p)). The mutual intensity and $|f(\mathbf{x}_o)|$ of the crystal are determined.

2.6. Fibre diffraction for quasi-monochromatic coherent wave

Consider a fibre (freely rotating) periodic in x_3 with period c. Sect. 2.1 is applied. Because of the one-dimensional periodic structure, the diffraction intensity and mutual intensity are greatly enhanced to form strong diffraction planes which can be measured accurately. In the following about projections, we will apply Yuen (2011d), modified for projections which may be non-centrosymmetric or complex.

2.6.1. Projection of one period of a fibre (freely rotating) to a plane normal to the fibre axis

These are inverse problems. Diffraction intensity or magnitude of the mutual intensity of the diffraction plane for $\omega_3 = 0$ is used. For phase-retrieval method, see Yuen (2011d), Sect. 2.1. For iterative algorithms with oversampling, see Yuen (2011d), Sect. 2.2.

(A) Diffraction intensity of the diffraction plane for $\omega_3 = 0$ is used

Ab initio $f_p(x_{o1}, x_{o2})$ of the projection is determined.

(B) Magnitude of the mutual intensity of the diffraction plane for $\omega_3 = 0$ is used

Ab initio mutual intensity and $\left| f_p(x_{o1}, x_{o2}) \right|$ of the projection are determined

2.6.2. Projection to a plane containing the fibre axis.

Diffraction intensity or magnitude of the mutual intensity of the diffraction lines for $\omega_1 = 0$ is used. For phase-retrieval method, see Yuen (2011d), Sect. 2.3. For iterative algorithms with oversampling, see Yuen (2011d), Sect. 2.4.

(A) Diffraction intensity of the diffraction lines for $\omega_1 = 0$ is used

Ab initio $f_h(x_{o2}, x_{o3})$ of the projection is determined.

(B) Magnitude of the mutual intensity of the diffraction lines for $\omega_1 = 0$ is used

Ab initio mutual intensity and $\left| f_h(x_{o2}, x_{o3}) \right|$ of the projection are determined.

2.6.3. Projection to an axis normal to the fibre axis

Diffraction intensity or magnitude of the mutual intensity of the diffraction line for $\omega_2 = \omega_3 = 0$ is used. For phase-retrieval method, see Yuen (2011d), Sect. 2.5. For iterative algorithms with oversampling, see Yuen (2011d), Sect. 2.6.

(A) Diffraction intensity of the diffraction line for $\omega_2 = \omega_3 = 0$ is used

Ab initio $f_n(x_{o1})$ of the projection is determined.

(B) Magnitude of the mutual intensity of the diffraction line for $\omega_2 = \omega_3 = 0$ is used

Ab initio mutual intensity and $\left| f_n(x_{o1}) \right|$ of the projection are determined.

2.6.4. Projection to the fibre axis

Diffraction intensity or magnitude of the mutual intensity of the diffraction line for $\omega_1 = \omega_2 = 0$ is used. For phase-retrieval method, see Yuen (2011d), Sect. 2.7. For iterative algorithms with oversampling, see Yuen (2011d), Sect. 2.8.

(A) Diffraction intensity of the diffraction line for $\omega_1 = \omega_2 = 0$ is used

Ab initio $f_c(x_{o3})$ of the projection is determined.

(B) Magnitude of the mutual intensity of the diffraction line for $\omega_1 = \omega_2 = 0$ is used

Ab initio mutual intensity and $|f_c(x_{o3})|$ of the projection are determined.

Treatment for projections of a fixed fibre can also be obtained.

2.6.5. Diffraction of coherent wave by a fibre (freely rotating) or a fixed fibre

(A) Direct problem

Sect. 2.1 can be applied to diffraction of coherent wave by a fibre (freely rotating) or a fixed fibre, namely, Yuen (2011e, 2011m), (1), (3).

(B) Inverse problem using the diffraction intensity of the diffraction planes

For phase-retrieval method, see Yuen (2011e, 2011m), Sect. 2.1, 3.1. For iterative algorithms with oversampling, see Yuen (2011e), Sect. 2.2, 3.2, and Yuen (2011m), Sect. 2.2, 3.2 – Sect. 5. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011r)). *Ab initio* $f(\mathbf{x}_o)$ of the fibre is determined.

(C) Inverse problem using the magnitude of the mutual intensity of the diffraction planes

For phase-retrieval method, see Yuen (2011m), Sect. 2.1, 3.1. For iterative algorithms with oversampling, see Yuen (2011m), Sect. 2.2, 3.2 – Sect. 5. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011r)). *Ab initio* mutual intensity and $|f(\mathbf{x}_o)|$ are determined.

2.7. Membrane diffraction for quasi-monochromatic coherent wave.

Consider a planar membrane, periodic in x_1 and x_2, with periods a and b. Sect. 2.1 is applied. Because of the two-dimensional periodic structure, the diffraction intensity and mutual intensity are greatly enhanced to form strong diffraction rods which can be measured accurately. In the following about projections, we will apply Yuen (2011f), modified for projections which may be non-centrosymmetric or complex.

2.7.1. Projection of one period of the membrane upon an axis normal to the membrane.

These are inverse problems. Diffraction intensity or magnitude of the mutual intensity of the diffraction rod for $\omega_1 = \omega_2 = 0$ is used. For phase-retrieval method, see Yuen (2011f), Sect. 2.1. For iterative algorithms with oversampling, see Yuen (2011f), Sect. 2.2.

(A) Determination of *ab initio* one-dimensional centrosymmetric electron density profile of a membrane

Electron density profile of the membrane is the projection of the electron density of one period of the membrane upon an axis normal to the membrane. Diffraction intensity of the diffraction rod for $\omega_1 = \omega_2 = 0$ is used. *Ab initio* $f_n(x_{o3})$ of the projection is determined.

(B) Determination of *ab initio* one-dimensional centrosymmetric mutual intensity and $|f_n(x_{o3})|$ of the projection of the membrane

Magnitude of the mutual intensity of the diffraction rod for $\omega_1 = \omega_2 = 0$ is used. *Ab initio* mutual intensity and $|f_n(x_{o3})|$ of the projection are determined.

2.7.2. Projection to the plane of the membrane

Diffraction intensity or magnitude of the mutual intensity of the diffraction points for $\omega_3 = 0$ is used. For phase-retrieval method, see Yuen (2011f), Sect. 2.3. For iterative algorithms with oversampling, see Yuen (2011f), Sect. 2.4.

(A) Diffraction intensity of the diffraction points for $\omega_3 = 0$ is used

Ab initio $f_h(x_{o1}, x_{o2})$ of the projection is determined.

(B) Magnitude of the mutual intensity of the diffraction points for $\omega_3 = 0$ is used

Ab initio mutual intensity and $|f_h(x_{o1}, x_{o2})|$ of the projection are determined.

2.7.3. Diffraction of coherent wave by a planar periodic membrane

(A) Direct problem

Sect. 2.1 can be applied to diffraction of coherent wave by a planar periodic membrane, namely, Yuen (2011g, 2011n), (1).

(B) Inverse problem using the diffraction intensity of the diffraction rods

For phase-retrieval method, see Yuen (2011g, 2011n), Sect. 2.1. For iterative algorithms with oversampling, see Yuen (2011g), Sect. 2.2, and Yuen (2011n), Sect. 2.2 – Sect. 3. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011s)). *Ab initio* $f(\mathbf{x}_o)$ of the membrane is determined.

(C) Inverse problem using the magnitude of mutual intensity of the diffraction rods

For phase-retrieval method, see Yuen (2011n), Sect. 2.1. For iterative algorithms with oversampling, see Yuen (2011n), Sect. 2.2 – Sect. 3. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011s)). *Ab initio* mutual intensity and $|f(\mathbf{x}_o)|$ of the membrane is determined.

3. Discussions and conclusion

This article deals with the direct and inverse problems of imaging of object irradiated with quasi-monochromatic coherent light wave. Diffraction intensity, mutual intensity or radiant intensity is dealt with. In the inverse problems, phase-retrieval by means of iterative algorithms with oversampling is used.

Sect. 2.1 deals with lensless imaging. For the direct problem, the first Born approximation is used. The mean wavelength $\overline{\lambda}$ of the incident wave is chosen according to the resolution needed. A three-dimensional nonperiodic object can be rotated so that all diffraction intensity and the magnitude of all mutual intensity in the far field of the object, for the chosen resolution, can be measured. In the inverse problem using the diffraction intensity in the far field, $f(\mathbf{x}_o)$ is obtained. In the inverse problem using the mutual intensity in the far field, $|f(\mathbf{x}_o)|$ is obtained. The lensless-imaging method presented can be compared with the lensless imaging in Spence et al. (2002), Azaña (2003), Miao et al. (2004), Chapman et al. (2006), Rodenburg et al. (2007), Vartanyants et al. (2007) and Rodenburg (2008).

In Sect. 2.2, superresolution is obtained from the real image of a nonperiodic object in a lens. The first Born approximation is not used. For a three-dimensional object, use all orientations of the object. Depending on the chosen resolution, we use a maximum mean wave number $\bar{k}_{\max}$. For a planar or three-dimensional object, use separately all incident quasi-monochromatic waves of same intensity with mean wave number $\bar{k}$, $\bar{k} \le \bar{k}_{\max}$. The radiant intensity and the magnitude of the mutual intensity in the far field of the image are measured.

Inverse problem using the radiant intensity in the far field is treated. Without using the image, Fourier optics with the empirical amplitude transfer function is used to obtain $f(\mathbf{x}_o)$ directly. The mutual intensity of the object is obtained. The natural resolution of the structure of the object is dealt with. The image is not dealt with. The Sparrow criterion of resolution (or alike) of the image and the corresponding angular resolution are by-passed. In effect, the whole image is used in determining the resolution of the structure obtained. The empirical amplitude transfer function contains aberrations of the lens and other defects of the system. The lens and the image may not have good quality, while the structure of the object obtained may still have good quality. The natural resolution may be comparable to the resolution of the image ($=\bar{\lambda}/2$), and is equal to or higher than the resolution provided by the Sparrow criterion. The angular resolution depends on the focal length of the lens, and not on its diameter. If very long focal length is used, the angular resolution is very small and very high resolution is achieved. Thin lens of small diameter may be used.

In the inverse problem using the mutual intensity in the far field of the image, the mutual intensity and intensity of the object are obtained. Natural resolution of the object is dealt with. Empirical amplitude transfer function $H(\mathbf{K})$ is used.

Phase-retrieval microscope and phase-retrieval telescope is a radical change in concept and design from the conventional microscope and telescope. An eyepiece is not needed. The object can be real or complex. The radiant intensity or mutual intensity can always be measured even if the image is small. Three-dimensional surface or volume structure of the object including the size (shape and dimensions) is obtained. The empirical amplitude transfer function contains aberrations of the lens and other defects of the system. It remains to determine the value of the natural resolution for phase-retrieval microscope in coherent imaging.

Transforming a nonperiodic object to a two-dimensional crystal is very useful in structure determination. By means of a two-dimensional array of lenslets, the weak scattering from the object is collected into strong diffraction rods. Relative to the background, the diffraction rods can be measured accurately. For a biological cell or molecule, short exposure of radiation can be used to avoid damage to the cell or molecule. During each exposure, radiant intensity or magnitude of the mutual intensity of some diffraction rods can be measured. Not radiant intensity or magnitude of the mutual intensity in all directions, only the diffraction rods need to be measured. These are lenslets microscope and lenslets telescope.

In the inverse problem for lenslets microscope or lenslets telescope using the radiant intensity of the diffraction rods of the image in all the lenslets, the extended $g(\mathbf{x})$ is determined. The object $f(\mathbf{x}_o)$ is determined is determined by means of periodic convolution. Natural resolution of the object is dealt with. In the inverse problem using mutual intensity of the diffraction rods, approximate value of the intensity of the image in one lenslet is obtained, in particular, for sufficiently large unit cell. The Sparrow criterion of resolution of the approximate intensity of the image (or alike) is applied. Natural resolution of the object is not dealt with.

20 advantages are listed for lenslets telescope employing radiant intensity. These

include higher resolution, minimization or elimination of seeing, determination of structure of smaller, fainter and more distant astronomical objects, and inexpensive thin lenslets of long focal length. Advantages for lenslets telescope employing mutual intensity are similar. Most of these advantages are shared by lenslets microscope. For very small distant astronomical object, there is a size limit that we can use a lens so that we can 'see' the image (Dainty & Fienup (1987), p. 231). For very small object, phase-retrieval telescope has to be used. For very weak object, there is an intensity limit for application of phase-retrieval telescope because the diffracted wave is not much stronger than the background. For very weak object, lenslets telescope has to be used.

The generalized transfer function $H(\mathbf{K}_1)H^*(-\mathbf{K}_2)$ (see (24)) is the Fourier transform of $h(\mathbf{x}_1)\otimes h^*(\mathbf{x}_2)$. $\otimes$ stands for convolution. When $h(\mathbf{x})$ is periodic, $h(\mathbf{x}_1)\otimes h^*(\mathbf{x}_2)$ does not converge (Oppenheim et al. (1983), p. 133). Hence we cannot use the generalized transfer function in the inverse problem using the mutual intensity of the diffraction rods for lenslets microscope and lenslets telescope (Sect. 2.3, (C)). We have to deal with the Sparrow criterion of resolution of the approximate intensity of the image. It will be very useful if we can find a way to by-pass the Sparrow criterion of resolution of the image (or alike) so that natural resolution of the object can be dealt with.

For lenslets microscope and lenslets telescope, research can be done to determine the optimal dimensions and focal length of the lenslet for structure determination (see (15)). It is useful to develop the technology of lenslets microscope and lenslets telescope. For X-ray and gamma-ray microscopes and telescopes, much research is needed to develop arrays of small lenslets.

Lenslet array is employed in Zalevsky & Mendlovic (2004), pp. 118-123. If, instead of lenslets, an array of apertures is used, each aperture can be regarded as a pin-hole camera. Microlens arrays are employed in Bewersdorf et al. (1998) and Arai et al. (2006). The Hartmann-Shack wavefront sensor also uses an array of lenslets. The superresolution, enhancement and directionality of the strong diffraction rods occurring in the far field may be compared with those occurring in the mesofield and near-field for subwavelength apertures or nanoarray (Ebbesen et al. (1998), Egorov et al. (2004) and Stark et al. (2007)).

For coded masks in neutron radiography, it is shown that optimized apertures (like multiple hole masks) improve considerably the quality of reconstructed images compared to a single hole system whenever only a weak signal is obtained, which is not much bigger than the noise level of the detector system (Grünauer (2005)).

Solutions to three-dimensional inverse source problems have been obtained for scatterers when the first Born approximation is satisfied (NietoVesperinas (2006), pp. 95, 333). By dealing with the image in a lens, such limitation can be by-passed. Phase-retrieval microscope, phase-retrieval telescope, lenslets microscope and lenslets telescope are applicable whether the first Born approximation is applicable or not.

Diffraction of quasi-monochromatic coherent wave by a nonperiodic object, or crystal diffraction, fibre diffraction, and membrane diffraction are dealt with. Powerful methods in X-ray crystallography can also be applied.

For an astronomical object, in principle, both the magnitude and the phase of the equal-time complex degree of coherence can be measured (Wolf (2007), p. 35). In practice, the measurement of the phase is almost impossible because of seeing (Mandel & Wolf (1995), p. 379, Dainty & Fienup (1987), p. 232).

Phase retrieval is an important and essential component of Fourier optics, Fourier acoustics (Williams (1999)) and matter-wave imaging. Optical wave, acoustic wave and matter wave, including electromagnetic wave, X-ray, electron, ion and neutron waves, involve both the magnitude and phase of wave. Just knowing the magnitude and not the phase is a great hindrance to the progress in the studies of optics, acoustics and matter wave. It presents certain degree of difficulty and incompleteness. The importance of phase can be seen in Goodman (1985), Sect. 7.4.3, 7.4.4, and Patil & Rastogi (2007). In fact, the phase captures more intelligibility of the object than the magnitude (Hayes (1982), p.151 and Lim (1990), Sect. 1.4.1). With phase retrieval, many problems may become solvable; phase retrieval may be regarded as the key to these problems. Much work can be done, and Fourier optics and Fourier acoustics fully exhibit their strength and usefulness. There will be a blooming of new developments.

With phase retrieval, similar developments and progresses may be expected in electrical engineering (Goodman (2005), p. 224).

To our knowledge, this is the first article which presents the phase-retrieval microscope and phase-retrieval telescope, transforming an object (a molecule, a cell, an astronomical object) to a crystal, lenslets microscope and lenslets telescope. These are very useful in single molecule imaging and astronomy.

Zarubin (1993), (4.2) and (5.9) may be used in Sect. 2.2, 2.3 and 2.4. In Zarubin (1996a), p. 230, it is mentioned that these results are applicable if the spatial coherence width of the incident wave is smaller than the size of the details of interest of the object. This means that the incident radiation is essentially “effectively incoherent”. Hence results for coherent radiation and partially coherent radiation need to be established. However, in Peřina (1985), p. 110, it is mentioned that the two-point resolution of the image in a lens is higher with incoherent light than with coherent light. It is intermediate for partially coherent light. Before results for coherent radiation and partially coherent radiation are established, may be we can use these results for coherent and partially coherent wave.

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2

Imaging of object irradiated with quasi-monochromatic incoherent wave.

Direct and inverse problems, lensless imaging (Michelson stellar interferometer), phase-retrieval microscope, phase-retrieval telescope, lenslets microscope and lenslets telescope, crystal diffraction, fibre diffraction, membrane diffraction

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This article deals with the direct and inverse problems of imaging of object irradiated with quasi-monochromatic incoherent light wave. Mutual intensity and radiant intensity are dealt with. Except for the diffraction intensity, the first Born approximation is not used. For a three-dimensional nonperiodicobject, use all orientations of the object. For lensless imaging, iterative algorithms with oversampling are applied to obtain the phase of equal-time complex degree of coherence from the experimentally measured magnitude by means of the van Cittert-Zernike theorem for planar object or the generalized van Cittert-Zernike theorem for three-dimensional object. This is the Michelson stellar interferometer. Superresolution via a lens is presented. For a planar or three-dimensional object, use separately all incident quasi-monochromatic waves of same intensity with mean wave number $\bar{k}$, $\bar{k} \le k_{\max}$. The radiant intensity and the magnitude of the mutual intensity are measured. Iterative algorithms with oversampling are applied to obtain the phase of the mutual intensity in the far field of the image from the magnitude. Empirical optical transfer function is used for determination of the structure of the object. Natural resolution of the object is dealt with. The Rayleigh criterion of resolution (or alike) of the image and the corresponding angular resolution are by-passed. Lenslets microscope is useful for determining structure of a cell or molecule, and lenslets telescope for structure of an astronomical object. Diffraction of incoherent wave by a nonperiodic object, or crystal diffraction, fibre diffraction, and membrane diffraction are dealt with. Powerful methods in X-ray crystallography can also be applied.

Keywords: Phase retrieval; iterative algorithms with oversampling; first Born approximation; equal-time complex degree of coherence; van Cittert-Zernike theorem; generalized van Cittert-Zernike theorem; three-dimensional object; nonperiodic object; lensless imaging; mutual intensity; radiant intensity; generalized transfer function; natural resolution; empirical amplitude transfer function; empirical optical transfer function; Rayleigh criterion of resolution; astronomy; Michelson stellar interferometer; crystal diffraction; diffraction points; fibre diffraction; diffraction planes; fibre projection; membrane diffraction; diffraction rods; optical wave, acoustic wave; matter wave; electromagnetic wave; X-ray, electron wave; ion wave; neutron wave.

1. Introduction

This is sequel to Yuen (2011t). The references in Yuen (2011t) are included. This article deals with the direct and inverse problems of imaging of object irradiated with quasi-monochromatic incoherent light wave. Mutual intensity and radiant intensity are dealt with.

For measurement of the equal-time complex degree of coherence, see, for example, Ambrosini et al. (1998), Castañeda & Jaroszewicz (2000), Basano et al. (2002), Lacefield & Waag (2002), Swartzlander & Schmit (2004), Begbie et al. (2001), Onodera et al. (2005), Tran et al. (2005), Lehman et al. (2007), Mejía & González (2007), Tran et al. (2007) and Carrasquilla-Alvarex et al. (2008).

The important Michelson stellar interferometer for incoherent source is discussed in Beran & Parrent (1964), Sect. 5.3, Roddier & Roddier (1976), Tallon & Tallon-Bosc (1992), Dyck et al. (1995), Mandel & Wolf (1995), Sect. 7.2, and Wolf (2007), Sect. 3.3.1. For reconstruction of incoherent sources by means of Michelson stellar interferometer, see Rosen & Yariv (1996a, 1996b, 1996c).

For imaging via the van Cittert-Zernike theorem, see, for example, Mallart & Fink (1991, 1994), Måsøy et al. (2005), Robert & Fink (2008).

For harmonic imaging, see, for example, Geiman et al. (2000) and Shen & Li (2001).

Generalizations of the Van Cittert-Zernike theorem are presented in Beran & Parrent (1964), (3-35), Carter & Wolf (1981), (5.8), Goodman (1985), Sect. 5.6.4., Mallart & Fink (1991), (10), (11), Zarubin (1993), (4.2), (5.9), and Taylor et al. (1994), (17), (22).

Scattering and diffraction of incoherent wave by crystal is presented in Pennycook & Jesson (1990), Chukhovskii & Förster (1995), Jesson & Pennycook (1995), Borgardt (1996), Dudarev (1997), Sinha et al. (1998), Borgardt (1999), McGibbon et al. (1999), Vartanyants & Robinson (2001), Watanabe et al. (2001), Zuo (2002), Vartanyants & Robinson (2003), Smekal et al. (2004), Henry et al. (2009).

For matter wave, see, for examples, Buljan (2005).

For inversion source problem, see, for examples Devaney (1979), Zarubin (1993, 1996a, 1996b), Barakat & Sandler (1999), Borghi et al. (2000), De Micheli & Viano (2000) and Wendoloski (2000).

In this article, except for diffraction intensity (Sect. 2.1, (A)), the first Born approximation is not used. For a three-dimensional nonperiodic object, use all orientations of the object. In Sect. 2.1, lensless imaging is presented. A quasi-monochromatic incoherent scalar wave is used. The magnitude of all equal-time complex degree of coherence is measured. Iterative algorithms with oversampling are applied to obtain the phase of equal-time complex degree of coherence from the experimentally measured magnitude by means of the van Cittert-Zernike theorem for planar object or the generalized van Cittert-Zernike theorem for three-dimensional object. The object is determined. This is the Michelson stellar interferometer.

In Sect. 2.2, via the real image of the object in a lens, superresolution is achieved. In the inverse problem, for a planar or three-dimensional object, use separately all incident quasi-monochromatic waves of same intensity with mean wave number $\bar{k}$, $\bar{k} \leq \bar{k}_{\max}$. The radiant intensity and the magnitude of the mutual intensity in the far field of the image are measured. Iterative algorithms with oversampling are applied to obtain the phase of the mutual intensity from the magnitude. Empirical optical transfer function is used to obtain the structure of the object. This includes aberrations of the lens and other defects of the system. The image of the object is not used explicitly. Natural resolution is applied to the object obtained. The Rayleigh criterion of resolution (or alike) of the image and the corresponding angular resolution are by-passed. The natural resolution is higher than the resolution provided by the Rayleigh criterion.

In Sect. 2.3, two-dimensional array of lenslets is used to form lenslets microscope or lenslets telescope. Only the diffraction rods are measured. In the inverse problem using the mutual intensity of the diffraction rods, because the incoherent imaging system is linear in intensity, the intensity of the object $\rho(\mathbf{x}_o)$ is determined. Natural resolution of the object is dealt with. Lenslets microscope is useful for determining structure of a cell or molecule, and lenslets telescope for structure of an astronomical object.

The direct and inverse problems of crystal diffraction for incoherent wave are dealt with in Sect. 2.4. Diffraction points of the mutual intensity are used. $\rho(\mathbf{x}_o)$ of the crystal is determined.

Determination of *ab initio* $\rho_p(x_{o1}, x_{o2})$ of projection of one period of a fibre (freely rotating) to a plane normal to the fibre axis is presented in Sect. 2.5.1. Sect. 2.5.2 deals with the projection to a plane containing the fibre axis. Sect. 2.5.3 deals with the projection to an axis normal to the fibre axis**.** Sect. 2.5.4 deals with the projection to the fibre axis. The direct and inverse problems of diffraction of incoherent wave by a fibre (freely rotating) or a fixed fibre are dealt with in Sect. 2.5.5. *Ab initio* $\rho(\mathbf{x}_o)$ of the fibre is determined. Diffraction planes of the mutual intensity are used.

Determination of *ab initio* $\rho_n(x_{o3})$ of the centrosymmetric projection upon an axis normal to a membrane is presented in Sect. 2.6.1. Sect. 2.6.2 deals with the projection to the plane of the membrane. The direct and inverse problems of diffraction for incoherent wave by a planar periodic membrane are dealt with in Sect. 2.6.3. *Ab initio* $\rho(\mathbf{x}_o)$ of the membrane is determined. Diffraction rods of the mutual intensity are used.

For diffraction of incoherent wave by a nonperiodic object, or crystal diffraction, fibre diffraction, and membrane diffraction, powerful methods in structure determination in X-ray crystallography, such as sharpening of Patterson function, direct methods, point-atom model, accelerated convergence of extension of Bragg intensities and structure factors, and sharpening in reciprocal space can also be applied.

This article may be applied to acoustic wave and matter wave, including electromagnetic wave, X-ray, electron, ion and neutron waves. For optical wave, acoustic wave and matter wave, including electromagnetic wave, X-ray, electron, ion and neutron waves, use Zarubin (1993), (4.2) for three-dimensional object and (5.9) for planar object.

2.1. Lensless imaging. Direct and inverse problems

(A) Direct problem

Consider a quasi-monochromatic incoherent scalar wave of mean angular frequency $\overline{\omega}$ incident on a nonperiodic object $f(\mathbf{x}_o,t)$. $\overline{\omega}=c\overline{k}$. c is the velocity of light. $\overline{k}$ is the mean wave number. From Iizuka (2008), (10.22), and Goodman (2005), (6-14), the intensity of the object $=\left|f(\mathbf{x}_o)\right|^2$. Denote this by $\rho(\mathbf{x}_o)$.

Hence $\left|f(\mathbf{x}_o)\right|^2=\rho(\mathbf{x}_o)$

For the object $f(\mathbf{x}_o,t)$, the instantaneous diffracted wave is given by its spatial Fourier transform $F(\mathbf{K},t)$. The first Born approximation is used.

The diffraction intensity

$$=\left\langle F(\mathbf{K},t)F^*(\mathbf{K},t)\right\rangle$$

$$=\left\langle\int f(\mathbf{x}_o,t)\exp(-i\mathbf{K}\cdot\mathbf{x}_o)d\mathbf{x}_o\int f^*(\mathbf{x}_o',t)\exp(i\mathbf{K}\cdot\mathbf{x}_o')d\mathbf{x}_o'\right\rangle$$

$$=\iint\left\langle f(\mathbf{x}_o,t)f^*(\mathbf{x}_o',t\right\rangle\exp[-i\mathbf{K}\cdot(\mathbf{x}_o-\mathbf{x}_o')]d\mathbf{x}_o'd\mathbf{x}_o$$

$\langle\ \rangle$ denotes averaging in time. For incoherent illumination,

$$\left\langle f(\mathbf{x}_o,t)f^*(\mathbf{x}_o',t)\right\rangle=\left|f(\mathbf{x}_o)\right|^2\delta(\mathbf{x}_o-\mathbf{x}_o')$$

(Iizuka (2008), (10.21)).

The diffraction intensity $=\int\left|f(\mathbf{x}_o)\right|^2d\mathbf{x}_o=\int\rho(\mathbf{x}_o)d\mathbf{x}_o=$ a constant (1) which is independent of $\mathbf{K}$. This agrees with Carter & Wolf (1981), (5.7b).

Consider a quasi-monochromatic incoherent scalar wave incident on a finite-size, non-negative, planar or three-dimensional object $\rho(\mathbf{x}_o)$. $\rho(\mathbf{x}_o)$ satisfies the constraints in Yuen (2011a or 2011h).

In the far field of the object, the scattered wave is partially coherent.

The intensity of the object $I_o(\mathbf{x}_o) = \rho(\mathbf{x}_o)$ (2)

From Beran & Parrent (1964), (7-36) and (7-49), for quasi-monochromatic incoherent object illumination, the mutual coherence function of the object $\Gamma_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}, \tau) = \rho(\mathbf{x}_{o2}) \exp(-i\overline{\omega}\tau) \delta(\mathbf{x}_{o2} - \mathbf{x}_{o1})$ (3)

The mutual intensity of the object

$$J_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}) \equiv \Gamma_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}, 0) = \rho(\mathbf{x}_{o2}) \delta(\mathbf{x}_{o2} - \mathbf{x}_{o1}) \quad (4)$$

$$\rho(\mathbf{x}_o) = J_o(\mathbf{x}_o, \mathbf{x}_o). \quad (5)$$

Consider the mutual intensity. Let ς_1 and ς_2 be two points in the far field. Denote the equal-time complex degree of coherence by $j(\varsigma_1, \varsigma_2)$. By the van Cittert-Zernike theorem for planar object (Born & Wolf (1989), p. 510, (26) for optics, Mallart & Fink (1991), (11) for acoustics and Zarubin (1993), (5.9), Taylor et al. (1994), (22) for matter wave), or the generalized van Cittert-Zernike theorem for three-dimensional object (Carter & Wolf (1981), (5.8) for optics, Mallart & Fink (1991), (10) for acoustics, and Zarubin (1993), (4.2), Taylor et al. (1994), (17) for matter wave), when the linear dimensions of the source and the distance between the observation points in the far field are small compared to the distances of these points from the source, the equal-time complex degree of coherence in the far field $j(\varsigma_1, \varsigma_2)$ is proportional to the Fourier transform $\mathrm{P}(\mathbf{K})$ of $\rho(\mathbf{x}_o)$. The first Born approximation is not used.

This case is the Michelson stellar interferometer for incoherent source (Beran & Parrent (1964), Sect. 5.3, Roddier & Roddier (1976), Tallon & Tallon-Bosc (1992), Dyck et al. (1995), Mandel & Wolf (1995), Sect. 7.2, Wolf (2007), Sect. 3.3.1).

The mean wavelength $\overline{\lambda}$ of the incident wave is chosen according to the resolution needed. A three-dimensional object can be rotated so that the magnitude of all mutual intensity in the far field $|J(\varsigma_1, \varsigma_2)|$ for the chosen resolution can be measured.

(B) Inverse problem using the mutual intensity in the far field of the object

Iterative algorithms with oversampling in Yuen (2011c or 2011k) are applied to obtain the phase of $j(\varsigma_1, \varsigma_2)$ from the magnitude. Powerful methods in structure determination in X-ray crystallography, such as sharpening of Patterson function, direct methods, point-atom model, accelerated convergence of extension of Bragg intensities and structure factors, and sharpening in reciprocal space can also be applied (Yuen (2011q)). The solution $\rho(\mathbf{x}_o)$ is obtained.

The resolution of the object $= \bar{\lambda}/2$ (6)

(Zarubin (1993), p. 502, Totzeck & Krumbügel (1994), p. 189, Goodman (2005), (3-74)).

Mutual intensity of the object is given by (4).

For reconstruction of incoherent sources by means of Michelson stellar interferometer, see Rosen & Yariv (1996a, 1996b, 1996c).

Remark: Sect. 2.1 can also be applied to imaging of object emitting quasi-monochromatic incoherent wave.

2.2. Image of object in a lens. Phase-retrieval microscope, phase-retrieval telescope. Direct and inverse problems

(A) Direct problem

This is a continuation of Sect. 2.1, Lensless imaging, (A) Direct problem.

Consider a quasi-monochromatic incoherent scalar wave of mean wave number $\bar{k}$ incident on a finite-size, non-negative, planar or three-dimensional object $\rho(\mathbf{x}_o)$ satisfying the constraints in Yuen (2011a or 2011h). As in Yuen (2011t), a lens is used to achieve superresolution. The incident wave upon the object is along the axis of the lens.

Let $h(\mathbf{x}) \equiv h(x_1, x_2)$ be the impulse response of the lens. $h(x_1, x_2)$ is proportional to the Fourier transform of the pupil function which is a two-dimensional function (Goodman (2005), (5-40)). The amplitude transfer function $H(\mathbf{K})$ is the Fourier transform of $h(\mathbf{x})$. Use a generalized transfer function (Beran & Parrent (1964), p. 112),

$$L(\mathbf{K}_1,\mathbf{K}_2) = H(\mathbf{K}_1)H^*(-\mathbf{K}_2) \tag{7}$$

$$\widetilde{J}_i(\mathbf{K}_1,\mathbf{K}_2) = L(\mathbf{K}_1,\mathbf{K}_2)\widetilde{J}_o(\mathbf{K}_1,\mathbf{K}_2) \tag{8}$$

where $\widetilde{J}_i(\mathbf{K}_1,\mathbf{K}_2)$ is the spatial Fourier transform of the mutual intensity $J_i(\mathbf{x}_1,\mathbf{x}_2)$ of the image

$\widetilde{J}_o(\mathbf{K}_1,\mathbf{K}_2)$ is the spatial Fourier transform of the mutual intensity $J_o(\mathbf{x}_1,\mathbf{x}_2)$ of the object.

The mutual intensity $J_i(\mathbf{x}_1,\mathbf{x}_2)$ of the image is the inverse spatial Fourier transform of $\widetilde{J}_i(\mathbf{K}_1,\mathbf{K}_2)$. (9)

The mutual coherence function of the image,

$$\Gamma_i(\mathbf{x}_1,\mathbf{x}_2,\tau) = J_i(\mathbf{x}_1,\mathbf{x}_2)\exp(-i\overline{\omega}\tau)$$

$$\overline{\omega} = c\overline{k}$$

The intensity of the image,

$$\psi(\mathbf{x}) \equiv \psi(x_1,x_2,x_3) = J_i(\mathbf{x},\mathbf{x}) = \rho(\mathbf{x})\otimes|h(\mathbf{x})|^2 = \rho(x_1,x_2,x_3)\otimes|h(x_1,x_2)|^2 \tag{10}$$

(Beran & Parrent (1964), p. 112, Goodman (2005), (6-15), Goodman (1985), (7.2-44)). The intensity and the magnitude of the mutual intensity of the image are measured.

In practice, only a truncated portion of the real image can be dealt with.

Let $\Psi(\mathbf{K}) \equiv \Psi(K_1,K_2,K_3)$, $\mathrm{P}(\mathbf{K}) \equiv \mathrm{P}(K_1,K_2,K_3)$ and $\Theta(\mathbf{K}) \equiv \Theta(K_1,K_2)$ be the Fourier transforms of $\psi(\mathbf{x})$, $\rho(\mathbf{x})$ and $|h(\mathbf{x})|^2$ respectively, normalized by their zero-frequency values (Goodman (2005), Sect. 6.3.1). $\Theta(\mathbf{K})$ is the optical transfer function.

$$\Psi(\mathbf{K}) \equiv \Psi(K_1, K_2, K_3) = \mathrm{P}(\mathbf{K})\Theta(\mathbf{K}) = \mathrm{P}(K_1, K_2, K_3)\Theta(K_1, K_2) \tag{11}$$

The image is partially coherent (Beran & Parrent (1964), p. 108). The degree of coherence varies over the image. The image acts as a secondary source of partially coherent wave. The first Born approximation is not used. From Carter & Wolf (1981), (3.5), Mandel & Wolf (1995), (5.2-25) for three-dimensional object, or Wolf (2007), p. 84, (16) for planar object, the cross-spectral density function in the far field of the image,

$$W(\varsigma_1\mathbf{u}_1, \varsigma_2\mathbf{u}_2, \overline{\omega}) = (2\pi)^6 \widetilde{W}_i(\bar{k}\mathbf{u}_1, -\bar{k}\mathbf{u}_2, \overline{\omega})\{\exp[i\bar{k}(\varsigma_1 - \varsigma_2)]\}/(\varsigma_1\varsigma_2) \tag{12}$$

$\mathbf{u}_1$, $\mathbf{u}_2$ are unit vectors.

From Carter & Wolf (1981), (3.9), Mandel & Wolf (1995), (5.2-15) or (5.2-19), (5.2-22) for three-dimensional object, or Wolf (2007), p.86, (20) for planar object, the radiant intensity in the far field of the image,

$$\Lambda(\mathbf{u}, \overline{\omega}) = (2\pi)^6 \widetilde{W}_i(\bar{k}\mathbf{u}, -\bar{k}\mathbf{u}, \overline{\omega}) \tag{13}$$

$\mathbf{u}$ is a unit vector

$\widetilde{W}_i$ is the six-dimensional (or four-dimensional for planar object) spatial Fourier transform of the cross-spectral density function of the image regarded as a source.

$J_i(\mathbf{x}_1, \mathbf{x}_2)$ denotes the mutual intensity of the image. In (12), apart from the factor $(2\pi)^6\{\exp[i\bar{k}(\varsigma_1 - \varsigma_2)]\}/(\varsigma_1\varsigma_2)$, the mutual intensity $J(\varsigma_1, \varsigma_2) \equiv \Gamma(\varsigma_1, \varsigma_2, 0)$ in the far field is the six-dimensional (or four-dimensional for planar object) spatial Fourier transform of $J_i(\mathbf{x}_1, -\mathbf{x}_2)$ with the image regarded as a source (from Yuen (2011t), (2)).

For a three-dimensional object, use all orientations of the object. Depending on the chosen resolution, we use a maximum mean wave number $\bar{k}_{\max}$. For a planar or three-dimensional object, use separately all incident quasi-monochromatic waves of same intensity with mean wave number $\bar{k}$, $\bar{k} \leq \bar{k}_{\max}$. The radiant intensity and the magnitude of the mutual intensity in the far field $|J(\varsigma_1, \varsigma_2)|$ are measured.

(B) Inverse problem using the mutual intensity in the far field of the image

$J_i(\mathbf{x}_1, -\mathbf{x}_2)$ is a six-dimensional complex body (or a four-dimensional complex body for a planar object) with each of its real and imaginary components satisfying the constraints (Yuen (2011i), (C)):

(i) finite size

(ii) non-negative

(iii) bounded

(iv) sectionally continuous

(v) sharp boundary. The component function $\neq 0$ almost everywhere at the boundary surface of the six-dimensional body (or four-dimensional body for a planar object)

The real and imaginary components satisfy the constraint

(vi) the size (shape and dimensions), and the regions of zeros (not isolated points) of the real part and the imaginary part are the same.

Constraint (v) is used because, in practice, only a truncated portion of the image can be dealt with.

For an incident wave of mean wave number $\bar{k}$ upon the object, in the far field of the image, the cross-spectral density function is given by (12) for all orientations of $\mathbf{u}_1$ and $\mathbf{u}_2$. Iterative algorithms with oversampling in Yuen (2011k) are applied to obtain the phase of $J(\varsigma_1, \varsigma_2)$ from the magnitude. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011q)). The solution $J_i(\mathbf{x}_1, -\mathbf{x}_2)$ is obtained.

Substitute $J_i(\mathbf{x}_1, -\mathbf{x}_2)$ determined into (13). The radiant intensity obtained should agree with the experimentally measured values.

Replace $-\mathbf{x}_2$ by $\mathbf{x}_2$. Mutual intensity of the image $J_i(\mathbf{x}_1, \mathbf{x}_2)$ is determined. Its magnitude should agree with the value measured in (A).

For $\mathbf{x}_1 = \mathbf{x}_2 = \mathbf{x}$, the intensity of the image

$$\psi(\mathbf{x}) = J_i(\mathbf{x}, \mathbf{x}) \tag{14}$$

is determined. This intensity should agree with the value measured in (A). $\Psi(\mathbf{K})$ which is the Fourier transform of $\psi(\mathbf{x})$ can be obtained.

The traditional approach is to make use of the property of a lens and the intensity of the image $\psi(\mathbf{x})$ obtained in (14). Good quality of the image is needed. The aberrations of the lens and other defects of the system can be treated as refinement. Taking account of these, an approximate image predicted by geometrical optics, $\rho(\mathbf{x})$ (approx.) of the object is obtained. From $\rho(\mathbf{x})$ (approx.), an approximate solution of the object, $\rho(\mathbf{x}_o)$ (approx.) is obtained. The Rayleigh criterion of resolution of the image (or alike, for example, Sparrow criterion, Reynolds et al. (1989), Ch. 6, 13, 19) and the corresponding angular resolution are applied.

The Rayleigh criterion of (angle) resolution $= 1.22\overline{\lambda} / D$ (15)

$\overline{\lambda}$ is the mean wavelength, and D the diameter of the lens.

For a conventional microscope, from Hecht (1989), (10.58), the radius of the first dark ring of the Airy pattern of the image,

$$q = 1.22 x_3 \overline{\lambda} / D \tag{16}$$

x_3 is much large than D. Hence q is large.

The resolution distance on the object $= q / |M| = 1.22 |x_{3o}| \overline{\lambda} / D$ (17)

A better way is to use $\Psi(\mathbf{K})$, the Fourier transform of $\psi(\mathbf{x})$ obtained in (14), directly. From (11),

$$\mathrm{P}(\mathbf{K}) = \Psi(\mathbf{K}) / \Theta(\mathbf{K}) \tag{18}$$

$\rho(\mathbf{x})$ is determined from $\mathrm{P}(\mathbf{K})$. The object $\rho(\mathbf{x}_o)$ is obtained.

Empirical amplitude transfer function $H(\mathbf{K})$ and empirical optical transfer function $\Theta(\mathbf{K})$ determined in Sect. 2.3 of Yuen (2008) will be used. They take into account aberrations of the lens and other defects of the system. Since the intensity of the image is not used explicitly, we have by-passed the two-point Rayleigh criterion of resolution (or alike) of the image, and the corresponding angular resolution. Natural resolution of the object $\rho(\mathbf{x}_o)$ is dealt with. The natural resolution is higher than the resolution provided by the Rayleigh criterion.

At present, we do not have values of the natural resolution for phase-retrieval microscope. To get an idea of the resolution of phase-retrieval microscope, we can compare the resolution of the intensity of the image $\psi(\mathbf{x})$ in the phase-retrieval microscope with the resolution of the conventional microscope. The resolution of the image $=\bar{\lambda}/2$ (see (6)). It is higher than the 'diffraction limited' resolution q of the conventional microscope (limited according to the Rayleigh criterion of resolution of the image).

The resolution distance on the object $=\bar{\lambda}/(2|M|)=\bar{\lambda}|x_{3o}|/(2x_3)$

which is much smaller than that of the conventional microscope (see (17)).

For conventional telescope, from Hecht (1989), (10.58),

$$q = 1.22 f\bar{\lambda}/D$$

The resolution distance on the object $= q/|M| = 1.22|x_{3o}|\bar{\lambda}/D$ (17)

	f (m)	D (m)	q	Rayleigh criterion of (angle) resolution (m^{-1})
The Hubble Space Telescope*,	56.7	2.4	$29\bar{\lambda}$	$0.51\bar{\lambda}$
The proposed James Webb Space Telescope*,	131.4	6.5	$25\bar{\lambda}$	$0.19\bar{\lambda}$
InFOC μ S hard X-ray telescope	8	0.4	$24\bar{\lambda}$	$3.1\bar{\lambda}$

(Tueller et al. (2005)),

MAX gamma-ray telescope (Barrière et al. (2006)),	86	2.24	$47\bar{\lambda}$	$0.54\bar{\lambda}$

* Values of f and D are from http://hubble.nasa.gov and www.jwst.nasa.gov.

For phase-retrieval telescope, resolution of intensity of the image $\psi(\mathbf{x})$, $=\bar{\lambda}/2$. It is higher than the 'diffraction limited' resolution q of these conventional telescopes.

The angular resolution $=\bar{\lambda}/(2f)$. Unlike the conventional telescope, this depends on f, but not on D. If large value of f is used, the angular resolution is very small.

The resolution distance on the object $=\bar{\lambda}/(2|M|)=\bar{\lambda}|x_{3o}|/(2f)$

For both the phase-retrieval microscope and phase-retrieval telescope, the natural resolution may be comparable to the resolution of intensity of the image $\psi(\mathbf{x})$.

The 11 advantages of phase-retrieval microscope and phase-retrieval telescope employing mutual intensity in Yuen (2011t), Sect. 2.4 are included.

Use the mutual intensity of the image $J_i(\mathbf{x}_1,\mathbf{x}_2)$ determined in above, and the generalized transfer function in (7) and (8). The mutual intensity $J_o(\mathbf{x}_{o1},\mathbf{x}_{o2})$ of the object is determined. $J_o(\mathbf{x}_{o1},\mathbf{x}_{o2})$ should satisfy (4). $\rho(\mathbf{x}_o)$ and $J_o(\mathbf{x}_{o1},\mathbf{x}_{o2})$ obtained should satisfy (5). The intensity of the object $I_o(\mathbf{x}_o)=J_o(\mathbf{x}_o,\mathbf{x}_o)$. This should equal to the intensity of object $I_o(\mathbf{x}_o)=\rho(\mathbf{x}_o)$ in (2). Natural resolution of the object is dealt with.

2.3. Lenslets microscope and lenslets telescope. Direct and inverse problems

(A) Direct problem

Two-dimensional array of lenslets can be used to form lenslets microscope or lenslets telescope (Yuen (2011t), Sect. 2.3 and 2.4). The array of lenslets forms a linear system. Its impulse response is formed by adding replica of $h(\mathbf{x})\equiv h(x_1,x_2)$ for one lenslet successfully in x_1, x_2 to give a periodic function, the extended $h(\mathbf{x})$. The periods are chosen such that the Fourier coefficients of this extended $h(\mathbf{x})$ are nonzero (Yuen

(2008), (D)).

The images in the lenselets are identical. The focal length of the lenslets, the object distance and the unit cell constants a,b of the extended $h(\mathbf{x})$ are chosen so that the intensity of the image predicted by geometrical optics in one lenslet, $\rho(\mathbf{x})$, can be contained in one unit cell of the extended $h(\mathbf{x})$ (Yuen (2008), (D), constraint (i)). The images predicted by geometric optics in all the lenslets is formed by adding replica of $\rho(\mathbf{x}) \equiv \rho(x_1, x_2, x_3)$ in one lenslet successfully in x_1, x_2 to give a periodic function, the extended $\rho(\mathbf{x})$, with same unit cell constants as the extended $h(\mathbf{x})$.

The intensity of the image in all the lenslets is formed by adding replica of the intensity $\psi(\mathbf{x}) \equiv \psi(x_1, x_2, x_3)$ of the image in one lenslet successfully in x_1 and x_2 to give a periodic function, the extended $\psi(\mathbf{x})$, with the same unit cell constants as the extended $h(\mathbf{x})$.

In the far field, the radiant intensities and mutual intensities of the images in the lenslets are identical. For an incident wave of wave number $\bar{k}$ upon the object, the weak radiant intensities and mutual intensities of the two-dimensional array of images of the object are collected to form strong diffraction rods; it is enhanced by the array of images. The radiant intensity and the magnitude of the mutual intensity $|J(\varsigma_1, \varsigma_2)|$ of the diffraction rods of the image in all the lenslets are measured. For a three-dimensional object, use all orientations of the object. Depending on the chosen resolution, we use a maximum mean wave number $\bar{k}_{\max}$. For a planar or three-dimensional object, use separately all incident quasi-monochromatic waves of same intensity with mean wave number $\bar{k}$, $\bar{k} \le \bar{k}_{\max}$.

(B) Inverse problem using the mutual intensity of the diffraction rods of the image in all the lenslets

Follow Sect. 2.2. Use Yuen (2011i), (H). Iterative algorithms with oversampling in Yuen (2011n) are applied to obtain the phase of the mutual intensity $J(\varsigma_1, \varsigma_2)$ of the diffraction rods from the magnitude. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011s)). The solution, $J_{il}(\mathbf{x}_1,-\mathbf{x}_2)$, is obtained. $J_{il}(\mathbf{x}_1,\mathbf{x}_2)$ is the mutual intensity of the image in all the lenslets. Substitute $J_{il}(\mathbf{x}_1,-\mathbf{x}_2)$ into (13). The value of the radiant intensity of the diffraction rods obtained should agree with the value measured in (A).

Replace $-\mathbf{x}_2$ by $\mathbf{x}_2$. The mutual intensity of the image in all the lenslets is obtained.

For $\mathbf{x}_1 = \mathbf{x}_2 = \mathbf{x}$, the intensity of the image in all lenslets is obtained. It is equal to the extended $\psi(\mathbf{x})$.

For each x_3, let $c_{\psi lm}(x_3)$, $l,m = 0,\pm1,\pm2,...$, be the Fourier coefficients of the extended $\psi(\mathbf{x})$.

The extended $\psi(\mathbf{x}) = \sum_{l,m=-\infty}^{\infty} c_{\psi lm}(x_3)\exp[i(l\omega_{10}x_1 + m\omega_{20}x_2)]$ (19)

where $\omega_{10} = 2\pi/a$, $\omega_{20} = 2\pi/b$.

For each x_3,

let $c_{\rho lm}(x_3)$, $l,m = 0,\pm1,\pm2,...$, be the Fourier coefficients of the extended $\rho(\mathbf{x})$,

let c_{h2lm}, $l,m = 0,\pm1,\pm2,...$, be the Fourier coefficients of the | extended $h(\mathbf{x})$ | ^{2}.

From (10) and Yuen (2008), (6) for periodic convolution,

$$c_{\rho lm}(x_3) = \frac{c_{\psi lm}(x_3)}{Uc_{h2lm}} \tag{20}$$

$U = ab$

The extended $\rho(\mathbf{x}) = \sum_{l,m=-\infty}^{\infty} c_{\rho lm}(x_3)\exp[i(l\omega_{10}x_1 + m\omega_{20}x_2)]$ (21)

The object $\rho(\mathbf{x}_o)$ is determined.

Natural resolution of the object is dealt with. The extended intensity of the image is not used explicitly. The Rayleigh criterion of resolution (or alike) of the image and the corresponding angular resolution are by-passed.

Empirical amplitude transfer function $H(\mathbf{K})$ and empirical optical transfer function $\Theta(\mathbf{K})$ are used. $h(\mathbf{x})$ contains the aberrations of the lenslets and other defects of the system. As a large number of lenslets are used, if the aberrations of the lenslets are random, not systematic, the effect of the aberrations of the lenslets upon the values of the diffraction rods may be negligible. This is very useful in the case of using an imperfect X-ray lenslets microscope to determine the structure of a single molecule

Lenslets microscope is useful for determining structure of a cell or molecule, and lenslets telescope for structure of an astronomical object. The 20 advantages of lenslets microscope or lenslets telescope employing mutual intensity in Yuen (2011t), Sect. 2.3 and 2.4 are included. Only the diffraction rods are measured.

2.4. Crystal diffraction for quasi-monochromatic incoherent wave. Direct and inverse problems

(A) Direct problem

Sect. 2.1 can be applied to diffraction of incoherent wave by crystals, namely, Yuen (2011a, 2011h, 2011i), (B), (D). Magnitude of the mutual intensity of the diffraction points is measured.

(B) Inverse problem using the mutual intensity of the diffraction points

Use the magnitude of the mutual intensity of the diffraction points. For phase-retrieval method, see Yuen (2011a, 2011h, 2011i), (B), (D). For iterative algorithms with oversampling, see Yuen (2011b or 2011j). Powerful methods in structure determination in X-ray crystallography, such as sharpening of Patterson function, direct methods, point-atom model, accelerated convergence of extension of Bragg intensities and structure factors, and sharpening in reciprocal space can also be applied (Yuen (2011o, 2011p)). $\rho(\mathbf{x}_o)$ of the crystal is determined.

2.5. Fibre diffraction for quasi-monochromatic incoherent wave

Consider a fibre (freely rotating) periodic in x_3 with period c. Sect. 2.1 is applied. Because of the one-dimensional periodic structure, the mutual intensity is greatly enhanced to form strong diffraction planes which can be measured accurately. In the following about projections, we will apply Yuen (2011d), modified for projections which may be non-centrosymmetric or complex.

2.5.1. Projection of one period of a fibre (freely rotating) to a plane normal to the fibre axis

This is an inverse problem. Magnitude of mutual intensity of the diffraction plane for $\omega_3 = 0$ is used. For phase-retrieval method, see Yuen (2011d), Sect. 2.1. For iterative algorithms with oversampling, see Yuen (2011d), Sect. 2.2. *Ab initio* $\rho_p(x_{o1}, x_{o2})$ of the projection is determined.

2.5.2. Projection to a plane containing the fibre axis.

Magnitude of the mutual intensity of the diffraction lines for $\omega_1 = 0$ is used. For phase-retrieval method, see Yuen (2011d), Sect. 2.3. For iterative algorithms with oversampling, see Yuen (2011d), Sect. 2.4. *Ab initio* $\rho_h(x_{o2}, x_{o3})$ of the projection is determined.

2.5.3. Projection to an axis normal to the fibre axis

Magnitude of the mutual intensity of the diffraction line for $\omega_2 = \omega_3 = 0$ is used. For phase-retrieval method, see Yuen (2011d), Sect. 2.5. For iterative algorithms with oversampling, see Yuen (2011d), Sect. 2.6. *Ab initio* $\rho_n(x_{o1})$ of the projection is determined.

2.5.4. Projection to the fibre axis

Magnitude of the mutual intensity of the diffraction line for $\omega_1 = \omega_2 = 0$ is used. For phase-retrieval method, see Yuen (2011d), Sect. 2.7. For iterative algorithms with oversampling, see Yuen (2011d), Sect. 2.8. *Ab initio* $\rho_c(x_{o3})$ of the projection is determined.

Treatment for projections of a fixed fibre can also be obtained.

2.5.5. Diffraction of incoherent wave by a fibre (freely rotating) or a fixed fibre

(A) Direct problem

Sect. 2.1 can be applied to diffraction of incoherent wave by a fibre (freely rotating) or a fixed fibre, namely, Yuen (2011e, 2011m), (1), (3).

(B) Inverse problem using the mutual intensity of the diffraction planes

For phase-retrieval method, see Yuen (2011e, 2011m), Sect. 2.1, 3.1. For iterative algorithms with oversampling, see Yuen (2011e), Sect. 2.2, 3.2, and Yuen (2011m), Sect. 2.2, 3.2 – Sect. 5. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011r)). *Ab initio* $\rho(\mathbf{x}_o)$ of the fibre is determined.

2.6. Membrane diffraction for quasi-monochromatic incoherent wave.

Consider a planar membrane, periodic in x_1 and x_2, with periods a and b. Sect. 2.1 is applied. Because of the two-dimensional periodic structure, the mutual intensity is greatly enhanced to form strong diffraction rods which can be measured accurately. In the following about projections, we will apply Yuen (2011f), modified for projections which may be non-centrosymmetric or complex.

2.6.1. Projection of one period of the membrane upon an axis normal to the membrane.

This is an inverse problem. Mutual intensity of the diffraction rod for $\omega_1 = \omega_2 = 0$ is used. For phase-retrieval method, see Yuen (2011f), Sect. 2.1. For iterative algorithms with oversampling, see Yuen (2011f), Sect. 2.2. *Ab initio* $\rho_n(x_{o3})$ of the projection is determined.

2.6.2. Projection to the plane of the membrane

Magnitude of the mutual intensity of the diffraction points for $\omega_3 = 0$ is used. For phase-retrieval method, see Yuen (2011f), Sect. 2.3. For iterative algorithms with oversampling, see Yuen (2011f), Sect. 2.4. *Ab initio* $\rho_h(x_{o1}, x_{o2})$ of the projection is determined.

2.6.3. Diffraction of incoherent wave by a planar periodic membrane

(A) Direct problem

Sect. 2.1 can be applied to diffraction of incoherent wave by a planar periodic membrane, namely, Yuen (2011g, 2011n), (1).

(B) Inverse problem using the mutual intensity of the diffraction rods

For phase-retrieval method, see Yuen (2011g, 2011n), Sect. 2.1. For iterative algorithms with oversampling, see Yuen (2011g), Sect. 2.2, and Yuen (2011n), Sect. 2.2 – Sect. 3. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011s)). *Ab initio* $\rho(\mathbf{x}_o)$ of the membrane is determined.

3. Discussions and conclusion

This article deals with the direct and inverse problems of imaging of object irradiated with quasi-monochromatic incoherent light wave. Diffraction intensity, mutual intensity or radiant intensity is dealt with. Except for diffraction intensity (Sect. 2.1, (A)), the first Born approximation is not employed. Phase-retrieval method is applied to the inverse problem. For a three-dimensional nonperiodic object, use all orientations of the object.

Lensless imaging is dealt with. In the far field of the object, $|j(\varsigma_1,\varsigma_2)|$ for $\mathbf{K}$, $|\mathbf{K}| \leq |\mathbf{K}|_{\max}$ are measured experimentally. $|\mathbf{K}|_{\max}$ corresponds to the $\overline{\lambda}$ chosen. The phase of the equal-time complex degree of coherence is obtained from the magnitude by iterative algorithms with oversampling. Structure of the object $\rho(\mathbf{x}_o)$ is obtained by means of the van Cittert-Zernike theorem for planar object or the generalized van Cittert-Zernike theorem for three-dimensional object. This is the Michelson stellar interferometer.

Via the image in a lens, superresolution is achieved. For the direct problem, the radiant intensity and the magnitude of the mutual intensity in the far field of the image are measured. For the inverse problem, for a planar or three-dimensional object, use separately all incident quasi-monochromatic waves of same intensity with mean wave number $\bar{k}$, $\bar{k} \le \bar{k}_{\max}$, and the magnitude of the mutual intensity in the far field of the image is measured. The intensity of the image $\psi(\mathbf{x})$ can be obtained. Use the optical transfer function. The object $\rho(\mathbf{x}_o)$ is obtained directly. The image of the object is not used explicitly. Empirical amplitude transfer function $H(\mathbf{K})$ and empirical optical transfer function $\Theta(\mathbf{K})$ are used. They take into account aberrations of the lens and other defects of the system. Since the intensity of the image is not used explicitly, we have by-passed the two-point Rayleigh criterion of resolution (or alike) of the image, and the corresponding angular resolution. Natural resolution of the object $\rho(\mathbf{x}_o)$ is dealt with. The natural resolution is higher than the 'diffraction limited' resolution of conventional microscope and telescope, limited according to the Rayleigh criterion of resolution which is regarded as a fundamental limit and 'a de-facto physical law'(Milanfar & Shakouri (2002).). The angular resolution depends on the focal length of the lens, and not on its diameter. If very long focal length is used, the angular resolution is very small and very high resolution is achieved. Thin lens of small diameter may be used. Hereafter may be we can use the term 'diffraction limited' for natural resolution, as it should be if the phase of the diffracted wave is retrieved. It remains to determine the value of the natural resolution for phase- retrieval microscope and phase-retrieval telescope in incoherent imaging. The natural resolutions of $\rho(\mathbf{x}_o)$ can be compared with the resolution in Beran & Parrent (1964), Ch. 8, Asakura (1974), Asakura & Mishina (1974), Nayyar & Verma (1978), Lipson & Lipson (1981), Sect. 9.5.2, Goodman (1985), Sect. 7.3.1, Born & Wolf (1989), Sect. 10.5.2, Reynolds et al. (1989), pp. 90-92, den Dekker & van den Bos (1997) and Sayanna et al. (2003).

Use the generalized transfer function. Mutual intensity and intensity of the object are determined.

Lenslets microscope is useful for determining structure of a cell or molecule, and lenslets telescope for structure of an astronomical object. Only the diffraction rods of the radiant intensity and mutual intensity of the image in all the lenslets are measured. In the inverse problem using the mutual intensity of the diffraction rods, because the incoherent imaging system is linear in intensity ((10) and Goodman (2005), p. 135), the intensity of the object $\rho(\mathbf{x}_o)$ is determined. Natural resolution of the object is dealt with. The intensity of the image is not used explicitly. The Rayleigh criterion of resolution (or alike) of the image and the corresponding angular resolution are by-passed. This is different from Yuen (2011t), Sect. 2.3, (C), where only approximate value of the intensity of the image is obtained. The Sparrow criterion of resolution of the approximate intensity of the image (or alike) is applied and natural resolution of the object is not dealt with.

The 20 advantages of lenslets microscope or lenslets telescope employing mutual intensity in Yuen (2011t), Sect. 2.3 and 2.4 are included.

Diffraction of incoherent wave by a nonperiodic object, or crystal diffraction, fibre diffraction, and membrane diffraction are dealt with. Powerful methods in X-ray crystallography can also be applied.

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3

Imaging of object irradiated with quasi-monochromatic partially coherent wave.

Direct and inverse problems, lensless imaging, phase-retrieval microscope, phase-retrieval telescope, lenslets microscope and lenslets telescope, crystal diffraction, fibre diffraction, membrane diffraction

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This article deals with the direct and inverse problems of imaging of object irradiated with quasi-monochromatic partially coherent light wave. Mutual intensity and radiant intensity are dealt with. Iterative algorithms with oversampling are applied to obtain the phase of mutual intensity from the experimentally measured magnitude. Lensless imaging of a nonperiodic object is presented. The first Born approximation is employed. Superresolution via a lens is presented. The first Born approximation is not used. The radiant intensity and the magnitude of the mutual intensity in the far field of the image are measured. Mutual intensity of the image is determined. Empirical amplitude transfer function and generalized transfer function are employed for determination of the object. Natural resolution of the object is dealt with. The Sparrow or Rayleigh criterion of resolution (or alike) of the image and the corresponding angular resolution are by-passed. Lenslets microscope is useful for determining structure of a cell or molecule, and lenslets telescope for structure of an astronomical object. Diffraction of partially coherent wave by a nonperiodic object, or crystal diffraction, fibre diffraction, and membrane diffraction are dealt with. Powerful methods in X-ray crystallography can also be applied.

Keywords: Phase retrieval; iterative algorithms with oversampling; mutual intensity; radiant intensity; nonperiodic object; natural resolution; generalized transfer function; empirical amplitude transfer function; Sparrow criterion of resolution; Rayleigh criterion of resolution; first

Born approximation; crystal diffraction; diffraction points; fibre diffraction; diffraction planes; fibre projection; membrane diffraction; diffraction rods; optical wave, acoustic wave; matter wave; electromagnetic wave; X-ray, electron wave; ion wave; neutron wave; astronomy.

1. Introduction

This article is sequel to Yuen (2011t, 2011u). The references in these articles are included. This article deals with the direct and inverse problems of imaging of object irradiated with quasi-monochromatic partially coherent light wave. Diffraction intensity, mutual intensity or radiant intensity is dealt with. It is mentioned in Carter & Wolf (1985) that there have been hardly any publications for the inverse problem. In this article, phase-retrieval method is applied to deal with this inverse problem.

For imaging with partially coherent light, see, for example, Beran & Parrent (1964), Ch. 7, 8, Marathay (1982), Ch. 6, Goodman (1985), Ch. 7, Reynolds et al. (1989), Ch. 11, 13, 16-18, Castañeda & Medina (1997), Castañeda, R. (1999), Oldenbourg & Török (2000), Török (2000), García-Sucerquia & Castañeda (2002), Teng et al. (2003), Castañeda (2004), García-Rodríguez et al. (2004), Sanchez-Brea et al, (2004), Cai & Zhu (2005), Nesterets et al. (2005), Qu et al. (2006), Withington et al. (2006a, 2006b), Crosby et al. (2007), Guigay et al. (2007), Qu et al. (2007), Williams et al. (2007), Chen et al. (2009), Flewett et al. (2009), Whitehead et al. (2009) and Quiney & Nugent (2011). It is discussed in Sect. 5.2 of the review, Thibault & Elser (2010).

Partially coherent imaging in high-resolution transmission electron microscopy and X-ray microscopy are presented in Chang et al. (2005) and von Hofsten et al. (2007) respectively.

For imaging via the generalized van Cittert-Zernike theorem, see, for example, Liu & Waag (1995).

For inversion source problem, see, for examples, Devaney (1979), Carter & Wolf (1985), LaHaie, I. J. (1985, 1986) and Habashy et al. (1997).

Lensless imaging is discussed in Carney & Wolf (2001), Azaña (2003), Cao et al. (2005), Scarcelli et al. (2006a, 2006b) and Cai & Wang (2007).

Reconstruction of an object from its image and similarity between object and image is discussed in Peřina (1985), Sect. 9.2.

For resolution, see, for example, Beran & Parrent (1964), Ch. 8, Asakura (1974), Asakura & Mishina (1974), Nayyar & Verma (1978), Lipson & Lipson (1981), Sect. 9.5.2, Goodman (1985), Sect. 7.3.1, Born & Wolf (1989), Sect. 10.5.2, Reynolds et al. (1989), pp. 90-92, den Dekker & van den Bos (1997), Thompson (2002) and Sayanna et al. (2003).

Scattering and diffraction of partially coherent wave by crystal is presented in Frankl (1980), Chukhovskii & Förster (1995), Dušek (1995), Jesson & Pennycook, (1995), Borgardt (1996), Felber et al. (1998), Sinha et al. (1998), Borgardt (1999), Vartanyants & Robinson (2001), Szöke (2001), Vartanyants & Robinson (2003a, 2003b), Leake et al. (2009).

For matter wave, see, for example, Nugent & Paganin (2000).

Lensless matter wave imaging is presented in Zarubin (1993, 1996a, 1996b).

In this article, Sect. 2.1 presents lensless imaging. The first Born approximation is used. A three-dimensional nonperiodic object is rotated so that the magnitude of all mutual intensity in the far field for the chosen resolution can be measured. By applying iterative algorithms with oversampling in Yuen (2011k) to the mutual intensity, $\left| f(\mathbf{x}_o) \right|$ is determined.

Imaging via a lens is presented in Sect. 2.2. The first Born approximation is not used. For a three-dimensional object, use all orientations of the object. For a planar or three-dimensional object, use separately all incident quasi-monochromatic waves of same intensity with mean wave number $\bar{k}$, $\bar{k} \le \bar{k}_{\max}$. The radiant intensity and the magnitude of the mutual intensity in the far field of the image are measured. Iterative algorithms with oversampling are applied to the mutual intensity. The mutual intensity of the image is determined. By means of a generalized transfer function, the mutual intensity and the intensity of the object are obtained. Empirical amplitude transfer function $H(\mathbf{K})$ is used. It takes into account aberrations of the lens and other defects of the system. Natural resolution of the object is dealt with. We have by-passed the

Sparrow or Rayleigh criterion of resolution (or alike) of the image, and the corresponding angular resolution.

For lenslets microscope and lenslets telescope, see Sect. 2.3. The mutual intensities at each diffraction rod add to each other; it is enhanced by the array of images. Only the radiant intensity and the magnitude of the mutual intensity of the diffraction rods of the image in all the lenslets are measured. From the magnitude of the mutual intensity of the diffraction rods, approximate value of the intensity of the image is obtained, in particular, for sufficiently large unit cell. The Sparrow criterion of resolution (or alike) of the approximate intensity of the image is applied. Natural resolution of the object is not dealt with.

By employing a lens or a number of lenslets in phase-retrieval microscope, phase-retrieval telescope, lenslets microscope or lenslets telescope, the image of the object is a secondary source. Limitation of the first Born approximation is by-passed.

Sect. 2.4 presents the direct and inverse problems of crystal diffraction for partially coherent wave. Diffraction points of the mutual intensity and intensity are measured. From the inverse problem using the mutual intensity, $\left|f(\mathbf{x}_o)\right|$ of the crystal is determined.

Determination of *ab initio* $\left|f_p(x_{o1}, x_{o2})\right|$ of projection of one period of a fibre to a plane normal to the fibre axis is presented in Sect. 2.5.1. Sect. 2.5.2 deals with the projection to a plane containing the fibre axis. Sect. 2.5.3 deals with the projection to an axis normal to the fibre axis**.** Sect. 2.5.4 deals with the projection to the fibre axis. The direct and inverse problems of diffraction of partially coherent wave by a fibre (freely rotating) or a fixed fibre are dealt with in Sect. 2.5.5. *Ab initio* $\left|f(\mathbf{x}_o)\right|$ of the fibre is determined. Diffraction planes of the mutual intensity are used.

Determination of *ab initio* $\left|f_n(x_{o3})\right|$ of the centrosymmetric projection upon an axis normal to a membrane is presented in Sect. 2.6.1. Sect. 2.6.2 deals with the projection to the plane of the membrane. The direct and inverse problems of diffraction for partially coherent wave by a planar periodic membrane are dealt with in Sect. 2.6.3. *Ab initio* $|f(\mathbf{x}_o)|$ of the membrane is determined. Diffraction rods of the mutual intensity are used.

For diffraction of partially coherent wave by a nonperiodic object, or crystal diffraction, fibre diffraction, and membrane diffraction, powerful methods in structure determination in X-ray crystallography, such as sharpening of Patterson function, direct methods, point-atom model, accelerated convergence of extension of Bragg intensities and structure factors, and sharpening in reciprocal space can also be applied.

This article may be applied to acoustic wave and matter wave, including electromagnetic wave, X-ray, electron, ion and neutron waves. For optical wave, acoustic wave and matter wave, including electromagnetic wave, X-ray, electron, ion and neutron waves, use Zarubin (1993), (4.2) for three-dimensional object and (5.9) for planar object.

2.1. Lensless imaging. Direct and inverse problems

(A) Direct problem

Consider a quasi-monochromatic partially coherent scalar wave of mean angular frequency $\overline{\varpi}$ incident on a finite-size, real or complex, planar or three-dimensional object $f(\mathbf{x}_o)$. $\overline{\varpi} = c\overline{k}$. c is the velocity of light. $\overline{k}$ is the mean wave number. $f(\mathbf{x}_o)$ satisfies the constraints in Yuen (2011a, 2011h or 2011i). Use $\boldsymbol{\varsigma}_1 = \varsigma_1 \mathbf{u}_1$, $\boldsymbol{\varsigma}_2 = \varsigma_2 \mathbf{u}_2$ where $\mathbf{u}_1$, $\mathbf{u}_2$ are unit vectors. $\overline{k}\mathbf{u}_0$ is the mean wavevector of the incident wave and $\overline{k}\mathbf{u}_1$, $\overline{k}\mathbf{u}_2$ are mean wavevectors of scattered waves. The first Born approximation is used. From Wolf (2007), Sect. 6.2, (6), and Sect. 6.1, (19), in the far field of the object, the mutual intensity

$\Gamma(\boldsymbol{\varsigma}_1, \boldsymbol{\varsigma}_2, 0)$

$$= \left(\frac{\exp[-i\bar{k}(\varsigma_1 - \varsigma_2)]}{\varsigma_1 \varsigma_2} \right) S^{(i)}(\bar{\omega}) \int_O \int_O \mu^{(i)}(\mathbf{x}_{o1}, \mathbf{x}_{o2}, \bar{\omega}) f^*(\mathbf{x}_{o1}) f(\mathbf{x}_{o2})$$

$$\exp(i\bar{k}\mathbf{u}_1 \cdot \mathbf{x}_{o1}) \exp(-i\bar{k}\mathbf{u}_2 \cdot \mathbf{x}_{o2}) d^3 x_{o1} d^3 x_{o2} \tag{1}$$

O denotes the object $f(\mathbf{x}_o)$.

$S^{(i)}(\bar{\omega})$ is the spectrum of the light incident on the object.

$\mu^{(i)}(\mathbf{x}_{o1}, \mathbf{x}_{o2}, \bar{\omega})$ is the spectral degree of coherence of the incident light (see Wolf (1983), (3.2), Friberg & Wolf (1995), (18) and Wolf & James (1996), (7.8)).

Obtain all $|\Gamma(\varsigma_1, \varsigma_2, 0)|$ in the far field, for all orientations of $\mathbf{u}_1$ and $\mathbf{u}_2$.

Apart from the factor $\left(\frac{\exp[-i\bar{k}(\varsigma_1 - \varsigma_2)]}{\varsigma_1 \varsigma_2} \right) S^{(i)}(\bar{\omega})$, the mutual intensity $J(\varsigma_1, \varsigma_2) \equiv \Gamma(\varsigma_1, \varsigma_2, 0)$ in the far field is the six-dimensional (or four-dimensional for planar object) spatial Fourier transform of $\mu^{(i)}(\mathbf{x}_{o1}, \mathbf{x}_{o2}, \bar{\omega}) f^*(-\mathbf{x}_{o1}) f(\mathbf{x}_{o2})$ with the image regarded as a source

The intensity in the far field $I(\varsigma) = J(\varsigma, \varsigma)$ (2)

(B) Inverse problem using the mutual intensity in the far field of the object

The mean wavelength $\bar{\lambda}$ of the incident wave is chosen according to the resolution needed. A three-dimensional object can be rotated so that the intensity and the magnitude of all mutual intensity in the far field $|J(\varsigma_1, \varsigma_2)|$ for the chosen resolution can be measured. This statement is true because it is true when the partially coherent wave is treated as a combination of coherent and incoherent components (Yuen

(2011w), Sect. 2.1, (A)).

$\mu^{(i)}(\mathbf{x}_{o1},\mathbf{x}_{o2},\overline{\omega})f^*(-\mathbf{x}_{o1})f(\mathbf{x}_{o2})$ is a six-dimensional complex body (or a four-dimensional complex body for a planar object) with each of its real and imaginary components satisfying the constraints (same as Yuen (2011i), (C)):

(i) finite size

(ii) non-negative

(iii) bounded

(iv) sectionally continuous

(v) sharp boundary. The component function $\neq 0$ almost everywhere at the boundary surface of the six-dimensional body (or four-dimensional for planar object)

The real and imaginary components satisfy the constraint

(vi) the size (shape and dimensions), and the regions of zeros (not isolated points) of the real part and the imaginary part are the same.

Constraint (v) is used because, in practice, only a truncated portion of the image can be dealt with.

Iterative algorithms with oversampling in Yuen (2011k) are applied to obtain the phase of $J(\varsigma_1,\varsigma_2)$ from the magnitude. Powerful methods in structure determination in X-ray crystallography, such as sharpening of Patterson function, direct methods, point-atom model, accelerated convergence of extension of Bragg intensities and structure factors, and sharpening in reciprocal space can also be applied (Yuen (2011q)). The solution $\mu^{(i)}(\mathbf{x}_{o1},\mathbf{x}_{o2},\overline{\omega})f^*(-\mathbf{x}_{o1})f(\mathbf{x}_{o2})$ is obtained.

For known $\mu^{(i)}(\mathbf{x}_{o1},\mathbf{x}_{o2},\overline{\omega})$, $f^*(-\mathbf{x}_{o1})f(\mathbf{x}_{o2})$ is determined. For $-\mathbf{x}_{o1}=\mathbf{x}_{o2}=\mathbf{x}_o$, $|f(\mathbf{x}_o)|$ is determined.

By Zernike's propagation law for the mutual intensity (Wolf (2007), p. 55, (4)), the mutual intensity at the surface of the object

$$J_o(\mathbf{x}_{o1},\mathbf{x}_{o2})=\frac{1}{\bar{\lambda}^2}\int_{\mathrm{B}}\int_{\mathrm{B}}J(\varsigma_1,\varsigma_2)\frac{\exp[i\bar{k}(R_2-R_1)]}{R_1R_2}d^2\varsigma_1 d^2\varsigma_2 \tag{3}$$

B is a plane of observation. $\mathbf{x}_{o1}$ and $\mathbf{x}_{o2}$ are points of the surface of the object facing B.

The intensity at the surface of the object facing B, $= J_o(\mathbf{x}_o,\mathbf{x}_o)$ (4)

For all orientations of the plane of observation B, the intensity at all surfaces of the object are obtained.

The resolution of the object $= \bar{\lambda}/2$ (5)

(Zarubin (1993), p. 502, Totzeck & Krumbügel (1994), p. 189, Goodman (2005), (3-74)).

2.2. Image of object in a lens, phase-retrieval microscope, phase-retrieval telescope. Direct and inverse problems

(A) Direct problem

This is a continuation of Sect. 2.1, Lensless imaging, (A) Direct problem.

A lens is used to achieve superresolution. Consider a quasi-monochromatic partially coherent optical wave (or electromagnetic wave or X-ray) of mean angular frequency $\bar{\omega}$ incident on a finite-size, real or complex, planar or three-dimensional object along the axis of the lens. Let $J_o(\mathbf{x}_{o1},\mathbf{x}_{o2})$ be the mutual intensity of the object.

The mutual coherence function of the object,

$$\Gamma_o(\mathbf{x}_{o1},\mathbf{x}_{o2},\tau)=J_o(\mathbf{x}_{o1},\mathbf{x}_{o2})\exp(-i\bar{\omega}\tau)$$

The intensity of the object $I_o(\mathbf{x}_o)=J_o(\mathbf{x}_o,\mathbf{x}_o)$. (6)

Let $H(\mathbf{K})$ be the amplitude transfer function of the lens. Use the generalized transfer function (Beran & Parrent (1964), p. 112),

$$L(\mathbf{K}_1,\mathbf{K}_2)=H(\mathbf{K}_1)H^*(-\mathbf{K}_2) \tag{7}$$

$$\tilde{J}_i(\mathbf{K}_1,\mathbf{K}_2)=L(\mathbf{K}_1,\mathbf{K}_2)\tilde{J}_o(\mathbf{K}_1,\mathbf{K}_2) \tag{8}$$

where $\tilde{J}_i(\mathbf{K}_1,\mathbf{K}_2)$ is the spatial Fourier transform of $J_i(\mathbf{x}_1,\mathbf{x}_2)$,

$\widetilde{J}_o(\mathbf{K}_1, \mathbf{K}_2)$ is the spatial Fourier transform of $J_o(\mathbf{x}_1, \mathbf{x}_2)$.

The mutual intensity of the image $J_i(\mathbf{x}_1, \mathbf{x}_2)$ is obtained.

The mutual coherence function of the image,

$\Gamma_i(\mathbf{x}_1, \mathbf{x}_2, \tau) = J_i(\mathbf{x}_1, \mathbf{x}_2)\exp(-i\overline{\omega}\tau)$

The intensity of the image $I_i(\mathbf{x}) = J_i(\mathbf{x}, \mathbf{x})$ (9)

The intensity and the magnitude of the mutual intensity of the image are measured.

The image of the object in a lens acts as a secondary source of partially coherent wave. The first Born approximation is not used. From Carter & Wolf (1981), (3.5), Mandel & Wolf (1995), (5.2-25) for three-dimensional object, or Wolf (2007), p. 84, (16) for planar object, the cross-spectral density function in the far field of the image,

$$W(\varsigma_1\mathbf{u}_1, \varsigma_2\mathbf{u}_2, \overline{\omega}) = (2\pi)^6 \widetilde{W}_i(\overline{k}\mathbf{u}_1, -\overline{k}\mathbf{u}_2, \overline{\omega})\left\{\exp[i\overline{k}(\varsigma_1 - \varsigma_2)]\right\}/(\varsigma_1\varsigma_2) \tag{10}$$

$\mathbf{u}_1$, $\mathbf{u}_2$ are unit vectors.

From Carter & Wolf (1981), (3.9), Mandel & Wolf (1995), (5.2-15) or (5.2-19), (5.2-22) for three-dimensional object, or Wolf (2007), p.86, (20) for planar object, the radiant intensity in the far field of the image,

$$\Lambda(\mathbf{u}, \overline{\omega}) = (2\pi)^6 \widetilde{W}_i(\overline{k}\mathbf{u}, -\overline{k}\mathbf{u}, \overline{\omega}) \tag{11}$$

$\mathbf{u}$ is a unit vector

$\widetilde{W}_i$ is the six-dimensional (or four-dimensional for planar object) spatial Fourier transform of the cross-spectral density function of the image regarded as a source.

For a three-dimensional object, use all orientations of the object. Depending on the chosen resolution, we use a maximum mean wave number $\overline{k}_{\max}$. For a planar or three-dimensional object, use separately all incident quasi-monochromatic waves of same intensity with mean wave number $\overline{k}$, $\overline{k} \le \overline{k}_{\max}$. The radiant intensity and the magnitude of the mutual intensity $|J(\varsigma_1,\varsigma_2)|$ in the far field are measured.

(B) Inverse problem using the mutual intensity in the far field of the image

Follow Yuen (2011u), Sect. 2.2. In (10), apart from a factor $(2\pi)^6\{\exp[i\bar{k}(\varsigma_1-\varsigma_2)]\}/(\varsigma_1\varsigma_2)$, $J(\varsigma_1,\varsigma_2)$ in the far field of the image is the six-dimensional (or four-dimensional for planar object) spatial Fourier transform of $J_i(\mathbf{x}_1,-\mathbf{x}_2)$ with the image regarded as a source. Iterative algorithms with oversampling in Yuen (2011k) are applied to obtain the phase of $J(\varsigma_1,\varsigma_2)$ from the magnitude. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011q)). The solution $J_i(\mathbf{x}_1,-\mathbf{x}_2)$ is obtained.

Substitute $J_i(\mathbf{x}_1,-\mathbf{x}_2)$ determined into (11). The radiant intensity obtained should agree with the experimentally measured values.

Replace $-\mathbf{x}_2$ by $\mathbf{x}_2$. Mutual intensity of the image $J_i(\mathbf{x}_1,\mathbf{x}_2)$ is determined. Its magnitude should agree with the value measured in (A).

For $\mathbf{x}_1=\mathbf{x}_2=\mathbf{x}$, the intensity of the image $I_i(\mathbf{x})=J_i(\mathbf{x},\mathbf{x})$ (9)

is determined. This intensity should agree with the value measured in (A).

Use the mutual intensity of the image $J_i(\mathbf{x}_1,\mathbf{x}_2)$ and the generalized transfer function (7) and (8). The mutual intensity of the object $J_o(\mathbf{x}_{o1},\mathbf{x}_{o2})$ is determined. The intensity of the object $I_o(\mathbf{x}_o)=J_o(\mathbf{x}_o,\mathbf{x}_o)$. In (7), the empirical amplitude transfer function $H(\mathbf{K})$ determined in Yuen (2008), Sect. 2.3 will be used. It takes into account aberrations of the lens and other defects of the system. Natural resolution of the object is dealt with. We have by-passed the Sparrow or Rayleigh criterion of resolution (or alike) of the image, and the corresponding angular resolution.

2.3. Lenslets microscope and lenslets telescope. Direct and inverse problems

(A) Direct problem

Follow Sect. 2.2 and Yuen (2011t), Sect. 2.3. Two-dimensional array of lenslets can be used to form lenslets microscope or lenslets telescope. The array of lenslets forms a linear system. The focal length of the lenslets, the object distance and the unit cell constants a, b of the array of lenslets are chosen so that the image of a nonperiodic object predicted by geometrical optics in one lenslet can be contained in one unit cell (Yuen (2008), (D), constraint (i)).

In the far field, the radiant intensities and mutual intensities of the images in the lenslets are identical. For an incident wave of wave number $\bar{k}$ upon the object, the weak radiant intensities and mutual intensities of the two-dimensional array of images of the object are collected to form strong diffraction rods; it is enhanced by the array of images. The radiant intensity and the magnitude of the mutual intensity $|J(\varsigma_1, \varsigma_2)|$ of the diffraction rods of the image in all the lenslets are measured. For a three-dimensional object, use all orientations of the object. Depending on the chosen resolution, we use a maximum mean wave number $\bar{k}_{\max}$. For a planar or three-dimensional object, use separately all incident quasi-monochromatic waves of same intensity with mean wave number $\bar{k}$, $\bar{k} \le \bar{k}_{\max}$.

(B) Inverse problem using the mutual intensity of the diffraction rods of the image in all the lenslets

Follow Sect. 2.2, (B) and Yuen (2011t), Sect. 2.3, (C). Iterative algorithms with oversampling in Yuen (2011n) are applied to obtain the phase of the mutual intensity of the diffraction rods $J(\varsigma_1, \varsigma_2)$ from the magnitude. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011s)). The solution, $J_{il}(\mathbf{x}_1, -\mathbf{x}_2)$, is obtained. $J_{il}(\mathbf{x}_1, \mathbf{x}_2)$ is the mutual intensity of the image in all the lenslets. Substitute $J_{il}(\mathbf{x}_1, -\mathbf{x}_2)$ into (11). The value of the radiant intensity of the diffraction rods obtained should agree with the value measured in (A).

Replace $-\mathbf{x}_2$ by $\mathbf{x}_2$. The mutual intensity of the image in all the lenslets is obtained. For $\mathbf{x}_1 = \mathbf{x}_2 = \mathbf{x}$, the intensity of the image in all lenslets is obtained.

Approximate value of the intensity of the image in one lenslet can be obtained from the intensity of the image in all lenslets in one unit cell, in particular, for sufficiently large unit cell. The Sparrow criterion of resolution of the approximate intensity of the image in one lenslet (or alike) and the corresponding angular resolution are applied. Natural resolution of the object is not dealt with.

2.4. Crystal diffraction for quasi-monochromatic partially coherent wave. Direct and inverse problems

(A) Direct problem

Sect. 2.1 can be applied to diffraction of partially coherent wave by crystals, namely, Yuen (2011a, 2011h, 2011i), (B), (D). The intensity and the magnitude of the mutual intensity in the far field are measured.

(B) Inverse problem using the mutual intensity of the diffraction points

Use the magnitude of the mutual intensity of the diffraction points. For phase-retrieval method, see Yuen (2011h, 2011i), (B), (D). For iterative algorithms with oversampling, see Yuen (2011j). Powerful methods in structure determination in X-ray crystallography, such as sharpening of Patterson function, direct methods, point-atom model, accelerated convergence of extension of Bragg intensities and structure factors, and sharpening in reciprocal space can also be applied (Yuen (2011o, 2011p)). $|f(\mathbf{x}_o)|$ of the crystal is determined.

2.5. Fibre diffraction for quasi-monochromatic partially coherent wave.

Consider a fibre (freely rotating) periodic in x_3 with period c. Sect. 2.1 is applied. Because of the one-dimensional periodic structure, the mutual intensity is greatly enhanced to form strong diffraction planes which can be measured accurately. In the following about projections, we will apply Yuen (2011d), modified for projections which may be non-centrosymmetric or complex.

2.5.1. Projection of one period of a fibre (freely rotating) to a plane normal to the

fibre axis

This is an inverse problem. Mutual intensity of the diffraction plane for $\omega_3 = 0$ is used. For phase-retrieval method, see Yuen (2011d), Sect. 2.1. For iterative algorithms with oversampling, see Yuen (2011d), Sect. 2.2. *Ab initio* $\left|f_p(x_{o1}, x_{o2})\right|$ of the projection is determined.

2.5.2. Projection to a plane containing the fibre axis.

Magnitude of the mutual intensity of the diffraction lines for $\omega_1 = 0$ is used. For phase-retrieval method, see Yuen (2011d), Sect. 2.3. For iterative algorithms with oversampling, see Yuen (2011d), Sect. 2.4. *Ab initio* $\left|f_h(x_{o2}, x_{o3})\right|$ of the projection is determined.

2.5.3. Projection to an axis normal to the fibre axis

Magnitude of the mutual intensity of the diffraction line for $\omega_2 = \omega_3 = 0$ is used. For phase-retrieval method, see Yuen (2011d), Sect. 2.5. For iterative algorithms with oversampling, see Yuen (2011d), Sect. 2.6. *Ab initio* $\left|f_n(x_{o1})\right|$ of the projection is determined.

2.5.4. Projection to the fibre axis

Magnitude of the mutual intensity of the diffraction line for $\omega_1 = \omega_2 = 0$ is used. For phase-retrieval method, see Yuen (2011d), Sect. 2.7. For iterative algorithms with oversampling, see Yuen (2011d), Sect. 2.8. *Ab initio* $\left|f_c(x_{o3})\right|$ of the projection is determined.

Treatment for projections of a fixed fibre can also be obtained.

2.5.5. Diffraction of partially coherent wave by a fibre (freely rotating) or a fixed fibre

(A) Direct problem

Sect. 2.1 can be applied to diffraction of partially coherent wave by a fibre (freely rotating) or a fixed fibre, namely, Yuen (2011e, 2011m), (1), (3).

(B) Inverse problem using the mutual intensity of the diffraction planes

For phase-retrieval method, see Yuen (2011m), Sect. 2.1, 3.1. For iterative algorithms with oversampling, see Yuen (2011m), Sect. 2.2, 3.2 – Sect. 5. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011r)). *Ab initio* $\left| f(\mathbf{x}_o) \right|$ of the fibre is determined.

2.6. Membrane diffraction for quasi-monochromatic partially coherent wave.

Consider a planar membrane, periodic in x_1 and x_2, with periods a and b. Sect. 2.1 is applied. Because of the two-dimensional periodic structure, the mutual intensity is greatly enhanced to form strong diffraction rods which can be measured accurately. In the following about projections, we will apply Yuen (2011f), modified for projections which may be non-centrosymmetric or complex.

2.6.1. Determination of projection of one period of the membrane upon an axis normal to the membrane.

This is an inverse problem. Mutual intensity of the diffraction rod for $\omega_1 = \omega_2 = 0$ is used. For phase-retrieval method, see Yuen (2011f), Sect. 2.1. For iterative algorithms with oversampling, see Yuen (2011f), Sect. 2.2. *Ab initio* $\left| f_n(x_{o3}) \right|$ of the projection is determined.

2.6.2. Projection to the plane of the membrane

Magnitude of the mutual intensity of the diffraction points for $\omega_3 = 0$ is used. For phase-retrieval method, see Yuen (2011f), Sect. 2.3. For iterative algorithms with oversampling, see Yuen (2011f), Sect. 2.4. *Ab initio* $\left|f_h(x_{o1}, x_{o2})\right|$ of the projection is determined.

2.6.3. Diffraction of partially coherent wave by a planar periodic membrane

(A) Direct problem

Sect. 2.1 can be applied to diffraction of partially coherent wave by a planar periodic membrane, namely, Yuen (2011g, 2011n), (1).

(B) Inverse problem using the mutual intensity of the diffraction rods

For phase-retrieval method, see Yuen (2011n), Sect. 2.1. For iterative algorithms with oversampling, see Yuen (2011n), Sect. 2.2 – Sect. 3. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011s)). *Ab initio* $\left|f(\mathbf{x}_o)\right|$ of the membrane is determined.

3. Discussions and conclusion

This article deals with the direct and inverse problems of imaging of object irradiated with quasi-monochromatic partially coherent light wave. Diffraction intensity, mutual intensity or radiant intensity is dealt with.

Lensless imaging of a nonperiodic object is treated. The first Born approximation is used. In the direct problem, the intensity and the magnitude of the mutual intensity in the far field of the object are measured. In the inverse problem, a three-dimensional object is rotated so that the magnitude of all mutual intensity in the far field, for the chosen resolution, can be measured. This statement is true because it is true when the partially coherent wave is treated as a combination of coherent and incoherent components (Yuen (2011w)). It will be very interesting if this statement can be proved directly from (1). $\left|f(\mathbf{x}_o)\right|$ of the object is determined. The intensity at the surface of the object is obtained. The lensless-imaging method presented can be compared with the lensless imaging in Carney & Wolf (2001), Azaña (2003), Cao et al. (2005), Scarcelli et al. (2006a, 2006b) and Cai & Wang (2007).

Superresolution is obtained from the image of an object in a lens. The first Born approximation is not used. The radiant intensity and the magnitude of the mutual intensity in the far field of the image are measured. In the inverse problem, use separately all incident quasi-monochromatic waves of same intensity with mean wave number $\bar{k}$, $\bar{k} \le \bar{k}_{\max}$. Iterative algorithms with oversampling in Yuen (2011k) are applied to the magnitude of the mutual intensity in the far field. The mutual intensity of the image is determined. By means of a generalized transfer function, the mutual intensity and the intensity of the object are obtained. Empirical amplitude transfer function is used. It takes into account aberrations of the lens and other defects of the system. Natural resolution of the object is dealt with. We have by-passed the Sparrow or Rayleigh criterion of resolution (or alike) of the image, and the corresponding angular resolution. It remains to determine the value of the natural resolution for phase-retrieval microscope in partially coherent imaging.

For an object $f(\mathbf{x}_o)$ irradiated with quasi-monochromatic partially coherent light wave, mutual intensity and intensity of the object are obtained by forming an image in a lens and applying phase-retrieval method to the measured magnitude of the mutual intensity in the far field. It will be very useful if analytical expressions for the mutual intensity $J_o(\mathbf{x}_{o1}, \mathbf{x}_{o2})$ and intensity $I_o(\mathbf{x}_o)$ of the object are available when the spectrum and the spectral degree of coherence of the incident light, and $f(\mathbf{x}_o)$ are known.

For lenslets microscope or lenslets telescope, the mutual intensities and radiant intensities of the diffraction rods of the image in all the lenslets are measured. In the inverse problem, from the magnitude of the mutual intensity of the diffraction rods, approximate value of the intensity of the image in one lenslet is obtained, in particular, for sufficiently large unit cell. The Sparrow criterion of resolution of the approximate intensity of the image (or alike) and the corresponding angular resolution are applied. Natural resolution of the object is not dealt with. Lenslets microscope is useful for determining structure of a cell or molecule, and lenslets telescope for structure of an astronomical object. The 20 advantages of lenslets microscope or lenslets telescope employing mutual intensity in Yuen (2011t), Sect. 2.3 and 2.4 are included.

Diffraction of partially coherent wave by a nonperiodic object, or crystal diffraction, fibre diffraction, and membrane diffraction are dealt with. Powerful methods in X-ray crystallography can also be applied.

A partially coherent wave has a unique decomposition in its coherent and incoherent component (Yuen (2011v), Sect. 2). Yuen (2011w) deals with the direct and inverse problems in partially coherent imaging in terms of its coherent and incoherent components. In terms of $f(\mathbf{x}_o)$ and $\rho(\mathbf{x}_o)$, analytical expressions of the intensity $I_o(\mathbf{x}_o)$ and mutual intensity $J_o(\mathbf{x}_{o1}, \mathbf{x}_{o2})$ of the object are obtained (Yuen (2011w), (12), (13)).

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4

Partial coherence.

Unique decomposition of quasi-monochromatic partially coherent waves in coherent and incoherent components

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This article deals with a converse problem which is a fundamental problem of waves (scalar or vector wave, light, electromagnetic, acoustic and matter waves): Is a partially coherent wave always a combination of a coherent wave and an incoherent wave? By means of a fundamental property, we found that: A quasi-monochromatic partially coherent radiation from a self-illuminating quasi-monochromatic source of size within certain finite limits is a combination of a coherent wave and an incoherent wave. The decomposition is unique. A pure partially coherent wave does not exist. This applies to sources emitting light wave, electromagnetic wave, acoustic wave, and matter wave (particles). These include chemical-reaction source (including fluorescence, phosphorescence, luminescence) and nuclear-reaction source (including alpha-decay, beta-decay gamma-decay). We have a very rare result that a fundamental property has to be changed because of theory. The mutual coherence function of a quasi-monochromatic partially coherent wave is equal to the sum of the mutual coherence function of the coherent component and the mutual coherence function of the incoherent component. The superposition of the coherent component and the incoherent component or its evolution, is always incoherent.

Keywords: Irreducible partially coherent optical wave; irreducible partially coherent matter wave; partially polarized; fundamental properties; object of size within certain finite limits; region of size within certain finite limits; unique decompositions; rare result; vector spaces; decomposition factor; filter.

1. Introduction

The combination of a coherent wave and an incoherent wave is a partially coherent wave. This article deals with the converse problem: Is a partially coherent wave always a combination of a coherent wave and an incoherent wave?

If a partially coherent wave has coherent-wave constituent and incoherent-wave constituent, we may delete these two constituents, theoretically or experimentally by means of a filter, if available. If the answer to the converse problem is positive, after the coherent and incoherent constituents are deleted, nothing is left. Studies of partial coherence will be much simplified. If the answer to the converse problem is negative, after the coherent and incoherent constituents are deleted, there remains a pure partially coherent wave which contains no coherent or incoherent constituents. Such a pure partially coherent wave is very interesting. Its properties can then be investigated. Furthermore, the van Cittert-Zernike theorem shows that the spatial coherence of incoherent wave increases with propagation in free space (Reynolds et al. (1989), p. 88, Wolf (2007), Sect. 3.4, 3.5). We let a pure partially coherent optical wave propagate to infinity. If any coherent or incoherent constituents are generated on propagation, they will be deleted theoretically. What remains is an irreducible partially coherent optical wave. Such irreducible partially coherent optical waves are very interesting as they are fundamental properties of the universe, just like the velocity of light. The number of irreducible partially coherent optical waves and their properties may be investigated. Similarly, if we let pure partially coherent matter waves propagate to infinity and delete the coherent or incoherent constituents, we get irreducible partially coherent matter waves.

Whether the answer to the converse problem is positive or negative, the result is very interesting and important. This article deals with the converse problem. We are not aware of any publication about the converse problem. We hope that this article can stimulate interest in this converse problem which is a fundamental problem of waves (scalar or vector wave, light, electromagnetic, acoustic and matter waves): Is a partially coherent wave always a combination of a coherent wave and an incoherent wave?

Coherent light and incoherent light are ideal cases. In practice, all lights are partially coherent. It is under extensive research.

Coherence and polarization of light is discussed in, for examples, Beran & Parrent

(1964), Marathay (1982), Goodman (1985), Peřina (1985), Born & Wolf (1989), Ch. X, Mandel & Wolf (1995), Wolf & James (1996), Brosseau et al. (1998), Cai et al. (2002), Basano et al. (2003), Lin et al. (2003), Pan & Lü (2003), Apostol & Dogariu (2004), Chaikina et al. (2004), Peng & Lü (2004), Zhao et al. (2006), Cai & Chen (2007), Gan & Gbur (2007), Pan et al. (2007), Wolf (2007), Zhang et al. (2007) and van Dijk (2008).

For source of radiation, see, for examples, Foley et al. (1988), Nugent (1995), Blomstedt et al. (2005), Baleine et al. (2006), Blomstedt et al. (2007), Yannopapas & Vitanov (2007) and Saklatvala et al. (2008).

For decomposition of partially coherent waves, see, for examples, Martínez- Herrero (1979), Martínez-Herrero & Mejídas (1985), Kutay et al. (2001), Laabs et al. (2002), Borghi et al. (2003), Luo & Lü (2003), Withington et al. (2004), Lajunen et al. (2005), Martinsson et al. (2007) and Martínez-Herrero et al. (2009).

Coherent-to-incoherent wave conversion is presented in Perdiagão et al. (1987), Javanaud & Franklin (1988) and Parolari et al. (2000).

By Stokes decomposition, every partially polarized light is an incoherent superposition of a polarized component and an unpolarized component, and the decomposition is unique (Shurcliff & Ballard (1964), p. 79-80, Klein (1970), p. 514, Born & Wolf (1989), Sect. 10.8.2, 10.8.3, Collett (1993), Sect. 4.6, Brosseau (1998), p. 110, O'Neill (2003), p.151, Wolf (2007), p. 163)). It is shown in Wolf (2008) and Kuebel et al. (2009) that this decomposition is local.

Wolf (2008) and Kuebel et al. (2009) give the necessary and sufficient conditions for the decomposition to be global.

Yuen (2011d)) shows the decomposition in a region of size within certain finite limits. The decomposition is unique.

Similarity between object and image is discussed in Peřina (1985), Sect. 9.2.

There are many articles about coherence or polarization by Wolf and his collaborators, with many important results. The list of the articles is too long to be included here (see, for examples, references in Born & Wolf (1989), Mandel & Wolf (1995), Wolf & James (1996), Wolf (2007)).

Matter wave is discussed in Raymer et al. (1994), Naraschewski & Glauber (1999), Buljan et al. (2005), Miller et al. (2005), Buljan et al. (2006), Chen & Lin(2008), and references in Yuen (2011b).

In this article, we consider quasi-monochromatic scalar and vector waves (optical wave, electromagnetic wave, acoustic wave and matter wave). By means of Fundamental Property I, we have obtained the answer to the converse problem. Sect. 2 shows: A quasi-monochromatic partially coherent radiation from a self-illuminating quasi-monochromatic source of size within certain finite limits is a combination of a coherent wave and an incoherent wave. The decomposition is unique. This decomposition is an intrinsic property of wave. A pure partially coherent wave does not exist. This applies to sources emitting light wave, electromagnetic wave, acoustic wave, and matter wave (particles). These include chemical-reaction source (including fluorescence, phosphorescence, luminescence) and nuclear-reaction source (including alpha-decay, beta-decay gamma-decay).

We have a very rare case that a fundamental property has to be changed because of theory.

Sect. 3 shows that the mutual coherence function of a quasi-monochromatic partially coherent wave is equal to the sum of the mutual coherence function of the coherent component and the mutual coherence function of the incoherent component. The superposition of the coherent component and the incoherent component or their evolutions, is always incoherent. Their intensities, mutual coherence functions, mutual intensities, cross-spectral density functions and radiant intensities add.

2. Decomposition of partially coherent wave

Consider a quasi-monochromatic scalar or vector wave.

For coherent illumination of a finite-size object, the object exhibits $f(\mathbf{x}_o)$. $f(\mathbf{x}_o)$ is real or complex.

The intensity of the object $= \left| f(\mathbf{x}_o) \right|^2$ (1)

For incoherent object illumination, the object exhibits $\rho(\mathbf{x}_o)$. $\rho(\mathbf{x}_o)$ is non-negative.

The intensity of the object $= \rho(\mathbf{x}_o)$ (2)

If we combine a coherent object illumination and an incoherent illumination, the result is a partially coherent illumination.

Suppose that a partially coherent illumination is not a sum of a coherent object illumination and an incoherent illumination. If we break down a general object illumination into its constituents, a general object illumination is a sum of coherent, incoherent, and pure partially coherent constituents. There can be more than one pure partially coherent constituent.

Consider a pure partially coherent constituent. The object exhibits $\gamma(\mathbf{x}_o)$. If $\gamma(\mathbf{x}_o)$ can be determined from $f(\mathbf{x}_o)$ and $\rho(\mathbf{x}_o)$, this means that the pure partially coherent constituent can be determined from the coherent object illumination and incoherent illumination. This is the result that we are seeking. Hence tentatively we assume that $\gamma(\mathbf{x}_o)$ is not determined from $f(\mathbf{x}_o)$ and $\rho(\mathbf{x}_o)$. Considering all known and unknown objects, we do not know the relation between $\gamma(\mathbf{x}_o)$ and $f(\mathbf{x}_o)$, $\rho(\mathbf{x}_o)$. We can only treat $\gamma(\mathbf{x}_o)$ as independent from $f(\mathbf{x}_o)$, $\rho(\mathbf{x}_o)$.

Use a pure partially coherent constituent as object illumination. The object exhibits $\gamma(\mathbf{x}_o)$. We have shown that $\gamma(\mathbf{x}_o)$ is independent from $f(\mathbf{x}_o)$, $\rho(\mathbf{x}_o)$. Hence, out of all known and unknown objects, there exists an object with $\gamma(\mathbf{x}_o)$ and $f(\mathbf{x}_o)$, $\rho(\mathbf{x}_o)$ such that

$$\gamma(\mathbf{x}_o) = \underbrace{f(\mathbf{x}_o) \otimes \ldots \otimes f(\mathbf{x}_o)}_{m\text{-times}} \otimes \underbrace{\rho(\mathbf{x}_o) \otimes \ldots \otimes \rho(\mathbf{x}_o)}_{n\text{-times}} \qquad (3)$$

with $m>0$, $n>0$. $\otimes$ stands for convolution.

Fundamental Property I: For every object of size within certain finite limits, the supports exhibit by the object under coherent, incoherent, or any pure partially coherent illumination, or their combinations, are the same.

The support of the convolution of two functions is much larger than the supports of the two functions (Papoulis (1968), p. 33). Hence, in (3), the support of $\gamma(\mathbf{x}_o)$ is much larger than the supports of $f(\mathbf{x}_o)$ and $\rho(\mathbf{x}_o)$. By Fundamental Property I, a pure partially coherent illumination does not exist. An object illumination can only be a coherent illumination or incoherent illumination.

Hence, we have proved: In a region of size within certain finite limits, every quasi-monochromatic partially coherent object illumination is always a combination of a coherent object illumination and an incoherent object illumination. This decomposition is unique because a coherent illumination is different from an incoherent illumination. (4)

In the proof, we have used the size of the supports of objects. Hence, our result (4) only applies to finite-size regions.

As the scalar or vector nature of the wave is not included, the decomposition applies to scalar or vector wave.

Fundamental Property II: For every finite-size object under coherent, incoherent or partially coherent object illumination, a lens forms a high-quality image in the Gaussian image plane, which is close to the image predicted by geometrical optics. The magnification of the image predicted by geometrical optics is

$$M = -x_3 / |x_{3o}|$$

The origin of the co-ordinate system is located at the centre of the lens, with the object $f(\mathbf{x}_o) \equiv f(x_{1o}, x_{2o}, x_{3o})$ on the negative-x_3 axis. $|x_{3o}|$ is the object distance from the lens, and x_3 the image distance. For coherent, incoherent or partially coherent illumination, the image distance x_3 is the same and $|x_{3o}|$ and x_3 are related by the lens law

$$\frac{1}{|x_{3o}|} + \frac{1}{x_3} = \frac{1}{f} \qquad (5)$$

f is the focal length.

We have proved that in a region of size within certain finite limits, every quasi-monochromatic partially coherent object illumination is a combination of a coherent object illumination and an incoherent object illumination. Since the images for coherent and incoherent illuminations have Fundamental Property II, the image for every partially coherent illumination has Fundamental Property II.

The image for every partially coherent illumination has Fundamental Property II serves as a verification of (4).

Denote coherent wave and incoherent wave of unit quantities by W_c and W_i respectively. A partially coherent object illumination W is a combination of $c_{10}W_c$ and $c_{01}W_i$. c_{10} is real or complex. c_{01} is non-negative. The object exhibits $c_{10}f(\mathbf{x}_o)$ and $c_{01}\rho(\mathbf{x}_o)$.

The intensity of the object $I_o(\mathbf{x}_o) = \left|c_{10}f(\mathbf{x}_o)\right|^2 + c_{01}\rho(\mathbf{x}_o)$ (6)

$f(\mathbf{x}_o)$ and $\rho(\mathbf{x}_o)$ are different physical quantities. We cannot add their numerical values.

We treat W_c and W_i as quantities in a two-dimensional vector space A for waves, with W_c and W_i as a basis.

A partially coherent wave $W = c_{10}W_c + c_{01}W_i$ (7)

c_{10} is real or complex. c_{01} is non-negative.

The uniqueness of the decomposition in above is a property of a vector space.

We treat f and ρ as quantities in a two-dimensional vector space Ω for the object under partially coherent object illumination, with f and ρ as a basis. Under the partially coherent illumination (7), the object exhibits

$c_{10}f(\mathbf{x}_o)$ and $c_{01}\rho(\mathbf{x}_o)$ (8)

To determine c_{10} and c_{01}, use an object of known $f(\mathbf{x}_o)$ under coherent illumination of unit quantity, and known $\rho(\mathbf{x}_o)$ under incoherent illumination of unit quantity. Use a partially coherent radiation as the object illumination. From the intensity of the object (6), a relation of $|c_{10}|$ and c_{01} is found. Use the same partially coherent radiation and a large number of objects with known $f(\mathbf{x}_o)$ and $\rho(\mathbf{x}_o)$. There are many relations and two unknowns. The system is over-determined. If consistent values of $|c_{10}|$ and c_{01} are obtained, this also serves as an experimental verification for the decomposition of partially coherent radiations. A direct verification is to use filters which absorb coherent radiation or incoherent radiation.

For a partially coherent wave, analogous to the degree of polarization for partially polarized light (Wolf (2007), p. 163, (26)), we can define a decomposition factor α as the ratio of the intensity of the coherent wave to the total intensity of the partially coherent wave.

$$\alpha = \frac{|c_{10}|^2}{|c_{10}|^2 + c_{01}} \tag{9}$$

$\alpha = 1$ coherent

$\alpha = 0$ incoherent

$0 < \alpha < 1$ partial coherent

α may be compared with the degree of coherence $|\gamma(Q_1, Q_2, \tau)|$ in Wolf (2007), p. 33, (11).

An object illumination is the wave incident on an object. The wave is emitted from a self-illuminating source. (For light wave, see also Wolf (2007), p. 113, 'the process of scattering on a static linear medium and the process of radiation from a localized source distribution are mathematically equivalent to each other.') Hence, we have:

> A partially coherent radiation from a self-illuminating quasi-monochromatic source of size within certain finite limits is a linear combination of a coherent wave $a_{10}W_c$ and an incoherent wave $a_{01}W_i$. a_{10} is real or complex. a_{01} is non-negative. The decomposition is unique. This applies to sources emitting light wave, electromagnetic wave, acoustic wave, and matter wave (particles).

These include chemical-reaction source (including fluorescence, phosphorescence, luminescence) and nuclear-reaction source (including alpha-decay, beta-decay gamma-decay).

A general picture is:

(a) A partially coherent radiation from a self-illuminated quasi-monochromatic source of size within certain finite limits is a linear combination of a coherent wave $a_{10}W_c$ and an incoherent wave $a_{01}W_i$. a_{10} is real or complex. a_{01} is non-negative. The decomposition is unique.

$$W = a_{10}W_c + a_{01}W_i \tag{10}$$

(b) The coherent wave remains unchanged (Beran & Parrent (1964), p. 47, Theorem I).

By the van Cittert-Zernike theorem for planar object (Born & Wolf (1989), p. 510, (26) for optics, Mallart & Fink (1991), (11) for acoustics and Zarubin (1993), (5.9) for matter wave), or the generalized van Cittert-Zernike theorem for three-dimensional object (Carter & Wolf (1981), (5.8) for optics, Mallart & Fink (1991), (10) for acoustics, and Zarubin (1993), (4.2), Taylor et al. (1994), (17) for matter wave), the spatial coherence of incoherent wave increases with propagation in free space (see also Reynolds et al. (1989), p. 88, Wolf (2007), p. 54).

(c) It is incident on a object of size within certain finite limits. The object illumination $W = c_{10}W_c + c_{01}W_i$ (7)

c_{10} is real or complex. c_{01} is non-negative.

(d) The object is represented by two functions, $f(\mathbf{x}_o)$ and $\rho(\mathbf{x}_o)$. $f(\mathbf{x}_o)$ and $\rho(\mathbf{x}_o)$ have the same support. The object exhibits $c_{10}f(\mathbf{x}_o)$ and $c_{01}\rho(\mathbf{x}_o)$.

The intensity of the object

$$I_o(\mathbf{x}_o) = \left|c_{10}f(\mathbf{x}_o)\right|^2 + c_{01}\rho(\mathbf{x}_o) \tag{6}$$

(e) Under the incoherent object illumination $c_{01}W_i$, the intensity of the diffracted wave in the far field $=c_{01}\int \rho(\mathbf{x}_o)d\mathbf{x}_o$ (11)

(Yuen (2011c), (1)).

Under the partially coherent object illumination $W = c_{10}W_c + c_{01}W_i$ in (c), the intensity of the diffracted wave in the far field

$$=|c_{10}F(\mathbf{K})|^2 + c_{01}\int \rho(\mathbf{x}_o)d\mathbf{x}_o \qquad (12)$$

$F(\mathbf{K})$ is the Fourier transform of $f(\mathbf{x}_o)$.

Remarks:

For (a), it is interesting that the decomposition for partially coherent wave may be obtained from Fundamental Property I.

Of course, different parts of the source may emit different quasi-monochromatic partially coherent waves of same or different frequencies.

For incoherent sources, see Beran & Parrent (1964), Sect. 4.4.

(b). For radiation from a coherent source, see Beran & Parrent (1964), Sect. 5.1. The sun is a source of size within certain limits. As the incoherent radiation from the sun propagates, it becomes partially coherent (Reynolds et al. (1989), Sect. 11.2). This can be decomposed into a coherent component and an incoherent component. As this incoherent component propagates, it is decomposed, so on. Hence the spatial coherence of the incoherent constituent increases with propagation in free space.

Because of the decomposition, we can deal with the coherent component and the incoherent component separately.

(b) and (c). For radiation from incoherent sources, see Beran & Parrent (1964), Sect. 5.2. It remains to develop analytical expressions for evaluating c_{10} and c_{01}.

It is shown in Wolf (2007), Sect. 5.4 that sources of different states of spatial coherence may generate identical distributions of the radiant intensity. This can be applied to the decomposition of the radiation in (a) to (c).

Theory of coherence of light in the literature can be applied to (a), (b), .., (e) with many simplifications.

3. Addition of intensities, mutual coherence functions, mutual intensities, cross-spectral density functions and radiant intensities.

Let the field be given by $\exp(i2\pi\nu t)+E(t)$.

The first term stands for coherent wave (Beran & Parrent (1964), p. 47, Th. I), and the second term for incoherent wave.

The mutual coherence function (Hecht (1989), (12.14))

$$=\left\langle[\exp i2\pi\nu(t+\tau)+E_1(t+\tau)][\exp(-i2\pi\nu t)+E_2{}^*(t)]\right\rangle.$$

$$=\left\langle\exp i2\pi\nu(t+\tau)\exp(-i2\pi\nu t)\right\rangle+\left\langle\exp i2\pi\nu(t+\tau)E_2{}^*(t)\right\rangle$$

$$+\left\langle E_1(t+\tau)\exp(-i2\pi\nu t)\right\rangle+\left\langle E_1(t+\tau)E_2{}^*(t)\right\rangle$$

$$=\exp i2\pi\nu\tau+0+0+\left\langle E_1(t+\tau)E_2{}^*(t)\right\rangle$$

$$=\exp i2\pi\nu\tau+\left\langle E_1(t+\tau)E_2{}^*(t)\right\rangle$$

Hence

the mutual coherence function of a quasi-monochromatic partially coherent wave

= mutual coherence function of the coherent component

+ mutual coherence function of the incoherent component. (13)

Such summation holds for the special case, the mutual intensities.

The mutual coherence function of the incoherent component may be non-zero as correlation is generated when the wave propagates (for example, the van Cittert-Zernike theorem (Wolf (2007), Sect. 3.2, (3), (5)). But because this correlation is generated from an incoherent source, there is no correlation between the coherent component $\exp(i2\pi\nu t)$ and the incoherent component $E(t)$. The superposition of the coherent component $\exp(i2\pi\nu t)$ and the incoherent component $E(t)$ or their evolutions, is always incoherent. Their intensities, mutual coherence functions, mutual intensities, cross-spectral density functions and radiant intensities add.

4. Discussions and conclusion

By means of Fundamental Property I, we have obtained the answer to the converse problem stated at the beginning of this article:

> A quasi-monochromatic partially coherent radiation from a self-illuminating quasi-monochromatic source of size within certain finite limits is a combination of a coherent wave $a_{10}W_c$ and an incoherent wave $a_{01}W_i$. a_{10} is real or complex. a_{01} is non-negative. The decomposition is unique.
>
> This decomposition is an intrinsic property of wave. A pure partially coherent wave does not exist.
>
> This applies to sources emitting light wave, electromagnetic wave, acoustic wave, and matter wave (particles). These include chemical-reaction source (including fluorescence, phosphorescence, luminescence) and nuclear-reaction source (including alpha-decay, beta-decay gamma-decay).
>
> In a region of size within certain finite limits, every quasi-monochromatic partially coherent wave is a combination of a coherent component and an incoherent component, and the decomposition is unique.
>
> An object is represented by two functions. It exhibit $f(\mathbf{x}_o)$ under coherent illumination, and exhibits a non-negative function $\rho(\mathbf{x}_o)$ under incoherent illumination. Under a partially coherent illumination, the object of size within certain limits, exhibits $c_{10} f(\mathbf{x}_o)$ and $c_{01}\rho(\mathbf{x}_o)$ (8)
>
> c_{10} is real or complex. c_{01} is non-negative.
>
> The intensity of the object
>
> $$I_o(\mathbf{x}_o) = |c_{10} f(\mathbf{x}_o)|^2 + c_{01}\rho(\mathbf{x}_o) \tag{6}$$
>
> Fundamental Property II serves as a verification of the decomposition.
>
> $|c_{10}|$ and c_{01} can be determined by employing a large number of objects with known $f(\mathbf{x}_o)$ and $\rho(\mathbf{x}_o)$. This also serves as an experimental verification for the decomposition. A direct verification is to use filters which absorb coherent

radiation or incoherent radiation. The phase of c_{10} is not determined. Since the experimentally observed quantities, intensity, mutual coherence function, mutual intensity, diffraction intensity involve $\left|c_{10}\right|^2$, the phase of c_{10} is not used.

The partially coherent waves form a two-dimensional vector space A, with W_c and W_i as a basis. f and ρ form a basis for the two-dimensional vector space Ω for the object under partially coherent object illumination.

The mutual coherence function of a quasi-monochromatic partially coherent wave is equal to the sum of the mutual coherence function of the coherent component and the mutual coherence function of the incoherent component.

The superposition of the coherent component and the incoherent component or their evolutions, is always incoherent. Their intensities, mutual coherence functions, mutual intensities, cross-spectral density functions and radiant intensities add. Hence in dealing with partially coherent wave in an imaging system, the coherent component and the incoherent component can be treated separately. The results can then be combined to give the result for partially coherent wave. The problem of partially coherent wave in an imaging system reduces to the problems of coherent wave and incoherent wave in the imaging system.

In a region of size within certain finite limits, every partially polarized light is an incoherent superposition of a polarized light and an unpolarized light, and the decomposition is unique (Yuen (2011d), Sect. 2). As coherence and polarization ‘are manifestations of the same physical phenomenon, namely of correlations between fluctuations in light beams’ (Wolf (2007), p. 174), it is reasonable to expect decomposition of every partially coherent light as a sum of a coherent light and an incoherent light, and the decomposition is unique.

In Fundamental Property I, the ‘certain finite limits’ have to be determined experimentally. There is no definite and unique answer. It depends on the deviations that we can tolerate. Hence for the result (4), there is no definite and unique answer to the ‘certain finite limits’. As the wave propagates, there is change in degree of coherence (Wolf (2007), Sect. 3.4, 3.5)). The answer to ‘certain finite limits’ depends on the deviations that we can tolerate.

Note that, because of the statistical optical coherence theory, the phrase ‘of size within certain limits’ has to be added to the Fundamental Property I. This then lead to the same phrase in result (4). If this phrase is not used, we will have (4) being applicable to any region, no matter how large. As coherence and polarization ‘are manifestations of the same physical phenomenon, this will contradict the results of Wolf (2008) and Kuebel et al. (2009) for global decomposition of partially polarized wave. Again we have a very rare result that a fundamental property has to be changed because of theory (see Yuen (2011a), Sect. 2.2). This change to Fundamental Property I should be investigated and confirmed empirically. Conversely, if this change can be confirmed, it adds to the validity of the statistical theories of partial coherence, partial polarization and their unifications.

This article presents the decomposition of partially coherent wave in a region of size within certain limits. Wolf (2008) and Kuebel et al. (2009) give the necessary and sufficient conditions for global decomposition of partially polarized wave. It will be very interesting if we can obtain necessary and sufficient conditions for global decomposition of partially coherent wave.

It remains to determine the relation between the decomposition factor in (9) with the degree of coherence $|\gamma(Q_1, Q_2, \tau)|$ in Wolf (2007), p. 33, (11). For a partially coherent wave with complex degree of coherence $\gamma(Q, Q, \tau)$ or equal-time complex degree of coherence $\gamma(Q, Q, 0)$ (Wolf (2007), p. 36, (21a)), find the coefficients c_{10} and c_{01}, or vice versa.

There are non-denumerable numbers of partially coherent waves, while there are only one coherent wave and one incoherent wave. The decomposition will provide simplifications in partial coherence and give more insight.

This discovery is fundamental. It may be applied to theory of coherence in the literature, for example, the source models such as Schell-model sources and quasi-homogeneous sources in Wolf (2007), Sect. 5.3.

It is important and useful to develop filters for absorbing coherent light (reverse of laser action) and incoherent light. The decomposition of partially coherent waves can then be verified directly.

Under a partially coherent illumination, the object of size within certain finite limits, exhibits $c_{10} f(\mathbf{x}_o)$ and $c_{01}\rho(\mathbf{x}_o)$ (8)
c_{10} is real or complex. c_{01} is non-negative.
The intensity of the object

$$I_o(\mathbf{x}_o) = \left|c_{10} f(\mathbf{x}_o)\right|^2 + c_{01}\rho(\mathbf{x}_o) \qquad (6)$$

This can be and should be verified experimentally.

We do not have experimental verification of the following statement:

> For every object under coherent, incoherent or partially coherent object illumination, the intensity of the image predicted by geometrical optics is a constant multiplying the intensity of the object. Same constant is used for coherent, incoherent and all partially coherent object illuminations.

Because of diffraction effect, we may not be able to determine exactly the image predicted by geometrical optics. We just use a high-quality image in the Gaussian image plane, which is close to the image predicted by geometrical optics.

If this statement has been verified experimentally, it can be included as part of Fundamental Property II in Sect. 2. Since the images for coherent and incoherent illuminations satisfy this statement, the image for every partially coherent illumination satisfies this statement. The image for every partially coherent illumination satisfies this statement serves as a verification of (4).

Fundamental Property I may be included in the theory of partial coherence. Then using the theory of partial coherence with Fundamental Property I, may enable us to prove the unique decomposition of a partially coherent wave.

In Yuen (2011d), there is Fundamental Property I’: For every object of size within certain finite limits, the supports exhibit by the object under polarized, unpolarized, or any pure partially polarized illumination, or their incoherent superpositions, are the same. It is shown that: In a region of size within certain finite limits, every quasi-monochromatic partially polarized object illumination is always an incoherent superposition of a polarized object illumination and an unpolarized object illumination. This decomposition is unique. As the decomposition of partially polarized wave is well known, we may include Fundamental Property I' in the theory of partial polarization. Then using the theory of partial polarization with Fundamental Property I', may enable us to prove the unique decomposition of a partially coherent wave. If this is achieved, similar approach may be followed in using the theory of partial coherence with Fundamental Property I, to prove the unique decomposition of a partially coherent wave.

It will be very interesting if we can converse at will the coherent component to incoherent component (Perdiagão et al. (1987), Javanaud & Franklin (1988), Parolari et al. (2000)), or incoherent component to coherent component (Goodman (2005), Sect. 7.2).

From Iizuka (2008), (10.22), and Goodman (2005), (6-14), $|f(\mathbf{x}_o)|^2 = \rho(\mathbf{x}_o)$. This article is not affected.

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5

Partial polarization.

Unique decomposition of quasi-monochromatic partially polarized waves in polarized and unpolarized components, and unique decomposition of partially coherent and partially polarized waves in four components. Incoherent superposition and coherent superposition

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By means of a fundamental property, we have proved: In a region of size within certain finite limits, every quasi-monochromatic partially polarized wave is an incoherent superposition of a polarized component and an unpolarized component, and the decomposition is unique. A pure partially polarized wave does not exist. For coherent partially polarized vector wave, besides incoherent superposition, coherent superposition may also be used. In a region of size within certain finite limits, every quasi-monochromatic coherent partially polarized wave is a coherent superposition of a polarized component and an unpolarized component, and the decomposition is unique. These decompositions are intrinsic properties of waves. Sources emitting electromagnetic wave include chemical-reaction source (including fluorescence, phosphorescence, luminescence) and nuclear-reaction source (including gamma-decay). We have a very rare case that a fundamental property has to be changed because of theory.

Keywords: Partially polarized; coherent partially polarized; electromagnetic wave; fundamental properties; object of size within certain finite limits; region of size within certain finite limits; unique decompositions; rare result; vector spaces; tensor product.

1. Introduction

This article is sequel to Yuen (2011b).

The combination of a polarized wave and an unpolarized wave is a partially polarized wave. This article deals with the converse problem: Is a partially polarized wave always a combination of a polarized wave and an unpolarized wave?

In fact, an answer to this converse problem for incoherent superposition has been provided: Every partially polarized light is an incoherent superposition of a polarized component and an unpolarized component, and the decomposition is unique.

This is proved by means of the Stokes parameters (Shurcliff & Ballard (1964), p. 79-80, Born & Wolf (1989), Sect. 10.8.3, Collett (1993), Sect. 4.6), or polarization matrix (coherency matrix) (Klein (1970), p. 514, Born & Wolf (1989), Sect. 10.8.2, Brosseau (1998), p. 110, O'Neill (2003), p.151, Wolf (2007), p. 163). It is shown in Wolf (2008) and Kuebel et al. (2009) that this decomposition is local.

Wolf (2008) and Kuebel et al. (2009) give the necessary and sufficient conditions for the decomposition to be global. Beams that admit such decomposition are termed Stokes beams. See also Du, & Zhao (2008), LeMaster & Cain (2008), Lahiri & Wolf (2009), Tervo & Turunen, (2009), Visser et al. (2009), Wolf (2009) and Tervo et al. (2010).

A completely unpolarized beam may be spatially fully coherent (Wolf (2003), Ponomarenko & Wolf (2003)). State of coherence and state of polarization are independent. For coherent partially polarized vector wave, besides incoherent superposition, coherent superposition may also be used.

Following the treatment in Yuen (2011b), this article presents an answer to this converse problem for incoherent superposition and for coherent superposition in a region of size within certain finite limits. The decomposition is unique. It is proved by means of two fundamental properties. These decompositions of partially polarized wave are intrinsic properties of waves. We found that a pure partially polarized wave which contains no polarized or unpolarized waves, does not exist.

We have a very rare case that a fundamental property has to be changed because of theory.

Coherence and polarization of light is discussed in, for examples, Beran & Parrent (1964), Marathay (1982), Goodman (1985), Peřina (1985), Born & Wolf (1989), Ch. X, Mandel & Wolf (1995), Brosseau et al. (1998), Zhao et al. (2006) and Wolf (2007).

For source of radiation, see, for example, Gori et al. (2003). Schwartz & Dogariu (2006) presents an inverse source problem for partially polarized beams.

For decomposition of partially coherent waves, see, for example, Liu et al. (2007).

For partially coherent and partially polarized wave, see, for examples, Seshadri (2000), Setälä et al. (2003), Saastamoinen & Tervo (2004a, 2004b), Tervo et al. (2004), Pan et al. (2005), Setälä et al. (2005), Castaneda & Garcia-Sucerquia (2006), Gori et al. (2006), Liu et al. (2006), Pu et al. (2006), Réfrégier & Roueff (2006a), Martínez-Herrero & Mejídas (2007a, 2007b), Réfrégier (2007), Santarsiero (2007), Alonso & Wolf (2008).

Unpolarized-to-polarized light conversion is presented in Pitois et al. (2005).

Polarization of matter wave is presented in Kuang et al. (2002) and Yang (2005).

2. Decomposition of partially polarized wave. Incoherent superposition

Consider a quasi-monochromatic vector wave.

For a polarized object illumination of a finite-size object, the object exhibits $q(\mathbf{x}_o)$. $q(\mathbf{x}_o)$ is real or complex (see, for example, polarized-light microscopy (Robinson & Bradbury (1992)).

The intensity of the object $= \left|q(\mathbf{x}_o)\right|^2$. (1)

An object may consist of, for example, several optically anisotropic substances.

For unpolarized object illumination, the object exhibits $\sigma(\mathbf{x}_o)$. $\sigma(\mathbf{x}_o)$ is real or complex.

The intensity of the object $= \left|\sigma(\mathbf{x}_o)\right|^2$ (2)

If we combine (incoherent superposition, Shurcliff & Ballard (1964), p. 79-80) a polarized object illumination and an unpolarized object illumination, the result is a partially polarized illumination.

Suppose that a partially polarized illumination is not a sum of a polarized object illumination and an unpolarized illumination. If we break down a general object illumination into its constituents according to incoherent superposition, a general object illumination is an incoherent superposition of polarized, unpolarized, and pure partially polarized constituents. There can be more than one pure partially polarized constituent.

Consider a pure partially polarized constituent. The object exhibits $\kappa(\mathbf{x}_o)$. If $\kappa(\mathbf{x}_o)$ can be determined from $q(\mathbf{x}_o)$ and $\sigma(\mathbf{x}_o)$, this means that the pure partially polarized constituent can be determined from the polarized object illumination and unpolarized illumination. This is the result that we are seeking. Hence tentatively we assume that $\kappa(\mathbf{x}_o)$ is not determined from $q(\mathbf{x}_o)$ and $\sigma(\mathbf{x}_o)$. Considering all known and unknown objects, we do not know the relation between $\kappa(\mathbf{x}_o)$ and $q(\mathbf{x}_o)$, $\sigma(\mathbf{x}_o)$. We can only treat $\kappa(\mathbf{x}_o)$ as independent from $q(\mathbf{x}_o)$, $\sigma(\mathbf{x}_o)$.

Use a pure partially polarized constituent as object illumination. The object exhibits $\kappa(\mathbf{x}_o)$. We have shown that $\kappa(\mathbf{x}_o)$ is independent from $q(\mathbf{x}_o)$, $\sigma(\mathbf{x}_o)$. Hence, out of all known and unknown objects, there exists an object with $\kappa(\mathbf{x}_o)$ and $q(\mathbf{x}_o)$, $\sigma(\mathbf{x}_o)$ such that

$$\kappa(\mathbf{x}_o) = \underbrace{q(\mathbf{x}_o) \otimes \ldots \otimes q(\mathbf{x}_o)}_{m\text{-times}} \otimes \underbrace{\sigma(\mathbf{x}_o) \otimes \ldots \otimes \sigma(\mathbf{x}_o)}_{n\text{-times}} \tag{3}$$

with $m>0$, $n>0$. $\otimes$ stands for convolution.

Fundamental Property I': For every object of size within certain finite limits, the supports exhibit by the object under polarized, unpolarized, or any pure partially polarized illumination, or their incoherent superpositions, are the same.

The support of the convolution of two functions is much larger than the supports of the two functions (Papoulis (1968), p. 33). Hence, in (3), the support of $\kappa(\mathbf{x}_o)$ is much larger than the supports of $q(\mathbf{x}_o)$ and $\sigma(\mathbf{x}_o)$. By Fundamental Property I', a pure partially polarized illumination does not exist. An object illumination can only be a polarized illumination or unpolarized illumination.

Hence, we have proved: In a region of size within certain finite limits, every quasi-monochromatic partially polarized object illumination is always an incoherent superposition of a polarized object illumination and an unpolarized object illumination.

This decomposition is unique because a polarized illumination is different from an unpolarized illumination. (4)

In the proof, we have used the size of the supports of objects. Hence, our result (4) only applies to finite-size regions.

As stated in Sect. 1, this decomposition has been proved by means of the Stokes parameters or polarization matrix.

Fundamental Property II': For every finite-size object under polarized, unpolarized or partially polarized object illumination, a lens forms a high-quality image in the Gaussian image plane, which is close to the image predicted by geometrical optics. The magnification of the image predicted by geometrical optics is

$$M = -x_3 / |x_{3o}|$$

The origin of the co-ordinate system is located at the centre of the lens, with the object $f(\mathbf{x}_o) \equiv f(x_{1o}, x_{2o}, x_{3o})$ on the negative-x_3 axis. $|x_{3o}|$ is the object distance from the lens, and x_3 the image distance. For polarized, unpolarized or partially polarized illumination, the image distance x_3 is the same and $|x_{3o}|$ and x_3 are related by the lens law

$$\frac{1}{|x_{3o}|} + \frac{1}{x_3} = \frac{1}{f} \tag{5}$$

f is the focal length.

We have proved that in a region of size within certain finite limits, every quasi-monochromatic partially polarized object illumination is an incoherent superposition of a polarized object illumination and an unpolarized object illumination. Since the images for polarized and unpolarized illuminations have Fundamental Property II', the image for every partially polarized illumination has Fundamental Property II'.

The image for every partially polarized illumination has Fundamental Property II' serves as a verification of (4).

Denote polarized wave and unpolarized object wave of unit quantities by W_p and W_u respectively. A partially polarized object illumination W is an incoherent superposition of $c_{10}W_p$ and $c_{01}W_u$. c_{10} and c_{01} are real or complex. The object exhibits $c_{10}q(\mathbf{x}_o)$ and $c_{01}\sigma(\mathbf{x}_o)$.

The intensity of the object $I_o(\mathbf{x}_o) = |c_{10}q(\mathbf{x}_o)|^2 + |c_{01}\sigma(\mathbf{x}_o)|^2$ (6)

$q(\mathbf{x}_o)$ and $\sigma(\mathbf{x}_o)$ are different physical quantities. We cannot add their numerical values.

In a region of size within certain finite limits, the partially polarized electromagnetic waves form a four-dimensional vector space B_i (Stokes space in O'Neill (2003), p. 155)), with four linearly independent Stokes vector as a basis (Collett (1993), Sect. 4.3, O'Neill (2003), Sect. 9.4). The subscript i stands for incoherent superposition which is used as the addition + in the vector space B_i. For example, we may use W_{p1}, W_{p2}, W_{p3} and W_u as a basis of B_i. W_{p1}, W_{p2}, W_{p3} are three linearly independent polarized waves, and W_u is an unpolarized wave.

W_p and W_u are quantities in B_i. A partially polarized wave

$$W = c_{10}W_p + c_{01}W_u \tag{7}$$

With polarized and unpolarized waves W_{p1}, W_{p2}, W_{p3}, W_u as object illumination, the object exhibits $q_1(\mathbf{x}_o)$, $q_2(\mathbf{x}_o)$, $q_3(\mathbf{x}_o)$ and $\sigma(\mathbf{x}_o)$ respectively. q_1 , q_2 , q_3 and σ form a basis of the vector space Φ_i for the object under partially polarized object illumination. Incoherent superposition is used as the addition + in this vector space.

q and σ are elements of Φ_i .

The intensity of the object $I_o(\mathbf{x}_o) = \left|c_{10}q(\mathbf{x}_o)\right|^2 + \left|c_{01}\sigma(\mathbf{x}_o)\right|^2$ (6)

$\left|q(\mathbf{x}_o)\right|^2$ = sum of the intensity of its components = $\sum_{j=1}^{3}\left|q_j(\mathbf{x}_o)\right|^2$ (8)

An object illumination is the wave incident on an object. The wave is emitted from a self-illuminating source. Hence, we have:

> A partially polarized radiation from a self-illuminating quasi-monochromatic source of size within certain finite limits, is an incoherent superposition of a polarized wave $a_{10}W_p$ and an unpolarized wave $a_{01}W_u$. a_{10} and a_{01} are real or complex. The decomposition is unique.

Sources emitting electromagnetic wave include chemical-reaction source (including fluorescence, phosphorescence, luminescence) and nuclear-reaction source (including gamma-decay).

3. Decomposition of partially coherent and partially polarized electromagnetic wave. Incoherent superposition

Consider quasi-monochromatic partially coherent and partially polarized electromagnetic wave.

Coherent light does not imply polarized light.

Proof. From Yuen (2011b), Sect. 2, in a region of size within certain finite limits, every partially coherent light is a sum of a coherent component and an incoherent component. The decomposition is unique.

From Sect. 2, every partially polarized light is an incoherent superposition of a polarized component and an unpolarized component, and the decomposition is unique. Consider a partially coherent and partially polarized light. Suppose that coherent light implies polarized light. The coherent component is the polarized component. Then the unpolarized component is the incoherent component. But this contradicts that a completely unpolarized beam may be spatially fully coherent (Wolf (2003), Ponomarenko & Wolf (2003)). Q.E.D.

Hence state of coherence and state of polarization are independent. Visser et al. (2009) presents different coherence properties of unpolarized light beams.

Combing the unique decompositions of partially coherent wave and partially polarized wave, in a region of size within certain finite limits, every quasi-monochromatic partially coherent and partially polarized electromagnetic wave is a linear combination (incoherent superposition) of four waves:

a coherent polarized wave,
a coherent unpolarized wave,
an incoherent polarized wave,
an incoherent unpolarized wave.

All have unit quantities. The decomposition is unique.

It remains to determine experimentally or theoretically the four coefficients (real or complex). The experimental determination serves as verification for the decomposition. Natural light is incoherent (Hecht (1989), p. 516) and unpolarized (Wolf (2007), p. 160).

The partially coherent waves form a two-dimensional vector space A, with W_c and W_i as a basis (Yuen (2011b), Sect 2). From Sect. 2, the partially polarized electromagnetic waves form a four-dimensional vector space B_i, with four linearly independent Stokes vector as a basis. We may use W_{p1}, W_{p2}, W_{p3} and W_u as a basis of B_i . These two vector spaces form a tensor product which is an eight-dimensional vector space for partially coherent and partially polarized electromagnetic waves (Halmos (1974), Sect. 25). A basis of the tensor product is given by the tensor product of the elements of the bases of the two vector spaces, namely, $\{W_c, W_i\}$ and $\{W_{p1}, W_{p2}, W_{p3}, W_u\}$.

With partially coherent waves W_c and W_i as object illumination, the object exhibits $f(\mathbf{x}_o)$ and $\rho(\mathbf{x}_o)$. f and ρ form a basis of the vector space Ω for the object under partially coherent object illumination (Yuen (2011b), Sect. 2). From Sect. 2, with polarized and unpolarized waves W_{p1}, W_{p2}, W_{p3}, W_u as object illumination, the object exhibits $q_1(\mathbf{x}_o)$, $q_2(\mathbf{x}_o)$, $q_3(\mathbf{x}_o)$ and $\sigma(\mathbf{x}_o)$ respectively. q_1, q_2, q_3 and σ form a basis of the vector space Φ_i for the object under partially polarized object illumination. Incoherent superposition is used as the addition + in this vector space. The tensor product of Ω and Φ_i is an eight-dimensional vector space for the object under partially coherent and partially polarized object illumination. The tensor product of $\{f, \rho\}$ and $\{q_1, q_2, q_3, \sigma\}$ form a basis of this vector space for the object.

The intensity of the object is given by (6) and (8).

4. Decomposition of coherent partially polarized wave. Coherent superposition

If the partially polarized wave has incoherent component, incoherent superposition must be used. If the partially polarized wave is coherent, there will be two choices: incoherent superposition which is dealt with in Sect. 2, or coherent superposition which is dealt with in this section.

Treatment in Sect. 2 can be followed. A coherent partially polarized object illumination is considered. Incoherent superposition is replaced by coherent superposition.

Fundamental Property I' becomes: For every object of size within certain finite limit, the supports exhibit by the object under polarized, unpolarized, or any pure partially polarized illumination, or their coherent superpositions, are the same.

(6) and (8) are not used. c_{10} and c_{01} are replaced by d_{10} and d_{01}. The result is:

> A partially polarized radiation from a self-illuminating quasi-monochromatic source of size within certain finite limits, is a coherent superposition of a polarized component and an unpolarized component, and the decomposition is unique.
>
> In a region of size within certain finite limits, every quasi-monochromatic coherent partially polarized wave W is a coherent superposition of a polarized component and an unpolarized component.
>
> $$W = d_{10}W_p + d_{01}W_u \tag{9}$$
>
> The decomposition is unique.
>
> Under the partially polarized illumination (9), the object exhibits the coherent superposition of $d_{10}q(\mathbf{x}_o)$ and $d_{01}\sigma(\mathbf{x}_o)$.
>
> Every quasi-monochromatic coherent and partially polarized wave is a linear combination (coherent superposition) of two waves:
>
> a coherent polarized wave,
>
> a coherent unpolarized wave.
>
> All have unit quantities. The decomposition is unique.
>
> The partially polarized electromagnetic waves form a four-dimensional vector space B_c. The subscript c stands for coherent superposition which is used as the addition + in this vector space.
>
> The object under partially polarized illuminations forms a four-dimensional vector space Φ_c. Coherent superposition is used as the addition + in this vector space.

The coherent waves form a one-dimensional vector space A_1, with W_c as a basis. The partially polarized electromagnetic waves form a four-dimensional vector space B_c. The tensor product of A_1 and B_c is a four-dimensional vector space for coherent and partially polarized electromagnetic waves. A basis of the tensor product is given by the tensor product of the elements of the bases of the two vector spaces A_1 and B_c.

The object under coherent illumination forms a one-dimentional vector space Ω_1 with f as a basis. The object under partially polarized illuminations forms a four-dimensional vector space Φ_c. The tensor product of Ω_1 and Φ_c is a four-dimensional vector space for the object under coherent and partially polarized object illumination. A basis of the tensor product is given by the tensor product of the elements of the bases of the two vector spaces Ω_1 and Φ_c.

5. Discussions and conclusion

By means of Fundamental Property I', we have proved the decomposition of quasi-monochromatic partially polarized vector wave:

A partially polarized radiation from a self-illuminating quasi-monochromatic source of size within certain limits, is an incoherent superposition of a polarized wave $a_{10}W_p$ and an unpolarized wave $a_{01}W_u$. a_{10} and a_{01} are real or complex. The decomposition is unique.

In a region of size within certain finite limits, every quasi-monochromatic partially polarized wave W is an incoherent superposition of a polarized component and an unpolarized component, and the decomposition is unique.

$$W = c_{10}W_p + c_{01}W_u \tag{7}$$

A pure partially polarized wave does not exist.

The partially polarized electromagnetic waves form a four-dimensional vector space B_i. Incoherent superposition is used as the addition + in this vector space.

An object exhibits $q(\mathbf{x}_o)$ under polarized illumination, and exhibits $\sigma(\mathbf{x}_o)$ under unpolarized illumination. $q(\mathbf{x}_o)$ and $\sigma(\mathbf{x}_o)$ are real or complex.

The object under partially polarized illumination forms a four-dimensional vector space Φ_i. Incoherent superposition is used as the addition + in this vector space.

Under the partially polarized illumination (7), the object exhibits $c_{10}q(\mathbf{x}_o)$ and $c_{01}\sigma(\mathbf{x}_o)$.

In a region of size within certain finite limits, every quasi-monochromatic partially coherent and partially polarized electromagnetic wave is a linear combination (incoherent superposition) of four waves:

a coherent polarized wave,
a coherent unpolarized wave,
an incoherent polarized wave,
an incoherent unpolarized wave.

The decomposition is unique.

The partially coherent waves form a two-dimensional vector space A, with W_c and W_i as a basis. The partially polarized electromagnetic waves form a four-dimensional vector space B_i. These two vector spaces form a tensor product which is an eight-dimensional vector space for partially coherent and partially polarized electromagnetic waves.

The tensor product of the vector space Ω for the object under partially coherent object illumination and the vector space Φ_i for the object under partially polarized object illumination is an eight-dimensional vector space for the object under partially coherent and partially polarized object illumination.

For coherent partially polarized vector wave, besides incoherent superposition, coherent superposition may also be used. We proved that:

A partially polarized radiation from a self-illuminating quasi-monochromatic source of size within certain finite limits, is a coherent superposition of a polarized component and an unpolarized component, and the decomposition is unique.

In a region of size within certain finite limits, every quasi-monochromatic coherent partially polarized wave W is a coherent superposition of a polarized component and an unpolarized component.

$$W = d_{10}W_p + d_{01}W_u \tag{9}$$

The decomposition is unique.

Under the partially polarized illumination (9), the object exhibits the coherent superposition of $d_{10}q(\mathbf{x}_o)$ and $d_{01}\sigma(\mathbf{x}_o)$.

Every quasi-monochromatic coherent and partially polarized wave is a linear combination (coherent superposition) of two waves:

a coherent polarized wave,

a coherent unpolarized wave.

All have unit quantities. The decomposition is unique.

The coherent waves form a one-dimensional vector space A_1, with W_c as a basis. The partially polarized electromagnetic waves form a four-dimensional vector space B_c. The tensor product of A_1 and B_c is a four-dimensional vector space for coherent and partially polarized electromagnetic waves.

The object under coherent illumination forms a one-dimentional vector space Ω_1 with f as a basis. The object under partially polarized illuminations (9) forms a four-dimensional vector space Φ_c. The tensor product of Ω_1 and Φ_c is a four-dimensional vector space for the object under coherent and partially polarized object illumination.

The decompositions are intrinsic properties of waves.

Fundamental Property II' serves as a verification of the decompositions.

In Fundamental Property I', the 'certain finite limits' have to be determined experimentally. There is no definite and unique answer. It depends on the deviations that we can tolerate. Hence for the result (4), there is no definite and unique answer to the 'certain finite limits'. As the wave propagates, there is change in polarization (James (1994)). The answer to 'certain finite limits' depends on the deviations that we can tolerate. This also applies to coherent superposition of a polarized component and an unpolarized component.

This article presents the decomposition of partially polarized wave in a region of size within certain limits, while Wolf (2008) and Kuebel et al. (2009) give the necessary and sufficient conditions for global decomposition.

Note that, because of the statistical optical polarization theory, the phrase 'of size within certain limits' has to be added to the Fundamental Property I'. This then lead to the same phrase in result (4). If this phrase is not used, we will have (4) being applicable to any region, no matter how large. This will contradict the results of Wolf (2008) and Kuebel et al. (2009). We have a very rare result that a fundamental property has to be changed because of theory (see Yuen (2011a), Sect. 2.2, Yuen (2011b), Sect. 4). This change to Fundamental Property I' should be investigated and confirmed empirically. Conversely, if this change can be confirmed, it adds to the validity of the statistical theories of partial coherence, partial polarization and their unifications.

We do not have experimental verification of the following statement:

> For every object under polarized, unpolarized or partially polarized object illumination, the intensity of the image predicted by geometrical optics is a constant multiplying the intensity of the object. Same constant is used for polarized, unpolarized or all partially polarized object illuminations.

Because of diffraction effect, we may not be able to determine exactly the image predicted by geometrical optics. We just use a high-quality image in the Gaussian image plane, which is close to the image predicted by geometrical optics.

If this statement has been verified experimentally, it can be included as part of Fundamental Property II' in Sect. 2. Since the images for polarized and unpolarized illuminations satisfy this statement, the image for every partially polarized illumination satisfies this statement. The image for every partially polarized illumination satisfies this statement serves as a verification that every quasi-monochromatic partially polarized object illumination is a combination (incoherent superposition or coherent superposition) of a polarized object illumination and an unpolarized object illumination.

Apply the decomposition to all articles and books about partially coherent and partially polarized wave. This may result in simplifications. For examples:

Coherent-mode decomposition (for examples, Gori et al. (2003), Liu et al. (2007)).

Rotating correlation (Saastamoinen & Tervo (2004a)).

Linear relations of coherent wave (Réfrégier & Roueff (2006b)).

Separating partial polarization and partial coherence (Réfrégier & Goudail (2005)).

Separation technique of a mixing of two uncorrelated and perfectly polarized lights

(Roueff & Réfrégier (2008))

Rotational frequency shifts for electromagnetic fields of arbitrary states of polarization (Agarwal & Gbur (2006)).

A unified theory of polarization and coherence (Wolf (2007), Ch.9).

'In the past few years, diverse approaches to coherence for vectorial waves have been proposed, so that currently there is no unanimously accepted proposal' (Luis (2007b), p.2070, Karczewski (1963), Ponomarenko & Wolf (2003), Tervo et al. (2003), Wolf (2003), Setälä et al. (2004a, 2004b), Wolf (2004), Réfrégier & Goudail (2005), Luis (2006), Réfrégier & Morio (2006), Luis (2007a)). These decompositions may reduce the ambiguity.

It will be very interesting and useful if proofs of the unique decompositions for partially coherent wave, and partially coherent and partially polarized wave can be provided by means of theories of coherence and polarization, and their unification (like Wolf (2007), Sect. 8.2.3). Via these proofs, there may be new insights and discoveries. If such proof can be established, we can use the unique decomposition of coherence to prove the Fundamental Property II.

Fundamental Property I' may be included in the theory of partial polarization. Then using the theory of partial polarization with Fundamental Property I', may enable us to prove the unique decomposition of a partially polarized wave, which is well known, If this is achieved, similar approach may be followed in using the theory of partial coherence with Fundamental Property I, to prove the unique decomposition of a partially coherent wave.

It remains to investigate the coherent superposition of coherent partially polarized waves in theory of partially polarized wave, and its applications.

It will be very interesting if we can converse at will between the four components of a partially coherent and partially polarized wave (Pitois et al. (2005)).

This article applies to electromagnetic wave. Whether it can be applied to matter wave remains to be investigated (Kuang et al. (2002) and Yang (2005)).

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6

Imaging of object irradiated with a quasi-monochromatic partially coherent wave which is a combination of a coherent wave and an incoherent wave.

Direct and inverse problems, lensless imaging, phase-retrieval microscope, phase-retrieval telescope, lenslets microscope and lenslets telescope, crystal diffraction, fibre diffraction, membrane diffraction

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This article deals with the direct and inverse problems of imaging of object irradiated with quasi-monochromatic partially coherent light wave which is a combination of a quasi-monochromatic coherent wave and a quasi-monochromatic incoherent wave. Diffraction intensity, mutual intensity and radiant intensity are dealt with. Lensless imaging of a nonperiodic object with partially coherent illuminations is presented. Iterative algorithms with oversampling are applied to the diffraction intensity and mutual intensity to obtain the phase from the magnitudes. By dealing with the coherent and incoherent components, $c_{10} f(\mathbf{x}_o)$ and $c_{01}\rho(\mathbf{x}_o)$ are determined. For imaging in a lens with partially coherent object illumination, iterative algorithms with oversampling are applied to the mutual intensity in the far field of the image. By employing two partially coherent illuminations, the coherent and incoherent components of the mutual intensity are obtained. The results obtained in the previous articles about coherent imaging and incoherent imaging can then be adopted. $|f(\mathbf{x}_o)|$ and $\rho(\mathbf{x}_o)$ are determined. Natural resolution of the object is used. The Sparrow criterion of resolution (or alike) of the image is by-passed. Lenslets microscope and lenslets telescope are dealt with. Diffraction of partially coherent wave by a nonperiodic object, or crystal diffraction, fibre diffraction, and membrane diffraction are dealt with. Powerful methods in X-ray crystallography can also be applied.

Keywords: Phase retrieval; iterative algorithms with oversampling; diffraction intensity; mutual intensity; radiant intensity; two partially coherent illuminations; first Born approximation; natural resolution; Empirical amplitude transfer function; empirical optical transfer function; Sparrow criterion of resolution; crystal diffraction; diffraction points; fibre diffraction; diffraction planes; fibre projection; membrane diffraction; diffraction rods; optical wave, acoustic wave; matter wave; electromagnetic wave; X-ray, electron wave; ion wave; neutron wave; astronomy.

1. Introduction

This article is sequel to Yuen (2011t, 2011u, 2011v, 2011w). The references in these articles are included. The combination of a coherent wave and an incoherent wave is a partially coherent wave. This article deals with the direct and inverse problems of imaging of nonperiodic object irradiated with such a quasi-monochromatic partially coherent light wave. Diffraction intensity, mutual intensity or radiant intensity is dealt with. For the inverse problems, iterative algorithms with oversampling are employed. In fact, in a region of size within certain finite limits, a partially coherent wave has a unique decomposition in its coherent and incoherent component (Yuen (2011w), Sect. 2).

Yuen (2011v) deals with partially coherent imaging. In this article, we investigate partially coherent imaging in terms of the coherent and incoherent components (Yuen (2011w)). Results obtained in the previous articles about coherent imaging and incoherent imaging can be used. More results and insight are obtained

Martínez-Herrero (1980) presents object reconstruction in a spatially partially coherent imaging system. By using an expansion of the correlation function of partially coherent fields, Martínez-Herrero & Mejídas (1981) presents the relation between the coherent components at the object and image planes. Ostrovky et al. (2001) presents modal theory of partially coherent imaging.

Incoherent to coherent image converter is presented in Zerbino et al. (2007).

Sect. 2.1 presents lensless imaging of a nonperiodic object under partially coherent illumination. From the intensity and the visibility measured in the far field of the object, the source structure is to be determined. This is an inverse source problem. In the direct problem, the total diffraction intensity and the magnitude of the total mutual intensity in the far field are measured. A three-dimensional object is rotated so that the magnitudes of all diffraction intensity and mutual intensity can be measured. In the inverse problem, iterative algorithms with oversampling are applied to the diffracted intensity and mutual intensity in the far field to obtain the phase from the magnitudes. $c_{10}f(\mathbf{x}_o)$ and $c_{01}\rho(\mathbf{x}_o)$ are determined.

Via the real image of the object in a lens, superresolution is achieved.

Sect. 2.2 presents imaging of object in a lens with partially coherent illumination. In the direct problem, the total radiant intensity and the magnitude of the total mutual intensity in the far field of the image are measured. In the inverse problem, use separately all incident quasi-monochromatic waves of same intensity, c_{10} and c_{01}, with mean wave number $\bar{k}$, $\bar{k} \le \bar{k}_{\max}$. Iterative algorithms with oversampling are applied to obtain the phase from the magnitude of the mutual intensity. Using a numerical method, $|f(\mathbf{x})|$ and $\rho(\mathbf{x})$ can be determined.

A conceptually better, analytical method is to use one more quasi-monochromatic partially coherent wave (Sect. 2.3). The mutual intensity obtained can be divided into coherent and incoherent components. For the coherent component, the results of Yuen (2011t), Sect. 2.2, (B) can be used. $|f(\mathbf{x}_o)|$ is determined. For the incoherent component, the results of Yuen (2011u), Sect. 2.2, (B) can be used. $\rho(\mathbf{x}_o)$ is determined. The intensity of the object obtained from $|f(\mathbf{x}_o)|$ and $\rho(\mathbf{x}_o)$ should be the same as the intensity determined in Yuen (2011v). This gives more insight. Natural resolution of the object is dealt with. Empirical amplitude transfer function $H(\mathbf{K})$ and empirical optical transfer function $\Theta(\mathbf{K})$ are used. They take into account aberrations of the lens and other defects of the system. The Sparrow criterion of resolution (or alike) of the image is by-passed.

Sect. 2.4 presents lenslets microscope and lenslets telescope with partially coherent object illumination. For the inversion problem, mutual intensity of the diffraction rods of the image in all the lenslets is used. The total intensity of the image is given by the sum of the approximate intensity of the image for the coherent component and the intensity of the image for the incoherent component. The Sparrow criterion of resolution of the approximate total intensity of the image (or alike) is applied. Natural resolution of the object is not dealt with.

Sect. 2.5 presents the direct and inverse problems of crystal diffraction for partially coherent wave. Diffraction points of the diffraction intensity and mutual intensity are used. $f(\mathbf{x}_o)$ and $\rho(\mathbf{x}_o)$ of the crystal are determined.

Projection of one period of a fibre (freely rotating) to a plane normal to the fibre axis is presented in Sect. 2.6.1. *Ab initio* $f_p(x_{o1}, x_{o2})$ and $\rho_p(x_{o1}, x_{o2})$ of the projection are determined. Sect. 2.6.2 deals with the projection to a plane containing the fibre axis. Sect. 2.6.3 deals with the projection to an axis normal to the fibre axis**.** Sect. 2.6.4 deals with the projection to the fibre axis. The direct and inverse problems of diffraction of partially coherent wave by a fibre (freely rotating) or a fixed fibre are dealt with in Sect. 2.6.5. *Ab initio* $f(\mathbf{x}_o)$, or *ab initio* $\rho(\mathbf{x}_o)$ of the fibre are determined. Diffraction planes of the diffraction intensity or mutual intensity are used.

Centrosymmetric projection upon an axis normal to a membrane is presented in Sect. 2.7.1. *Ab initio* $f_n(x_{o3})$ and $\rho_n(x_{o3})$ of the projection are determined. Sect. 2.7.2 deals with the projection to the plane of the membrane. The direct and inverse problems of diffraction for partially coherent wave by a planar periodic membrane are dealt with in Sect. 2.7.3. *Ab initio* $f(\mathbf{x}_o)$ or *ab initio* $\rho(\mathbf{x}_o)$ of the membrane is determined. Diffraction rods of the diffraction intensity or mutual intensity are used.

For diffraction of partially coherent wave by a nonperiodic object, or crystal diffraction, fibre diffraction, and membrane diffraction, powerful methods in structure determination in X-ray crystallography, such as sharpening of Patterson function, direct methods, point-atom model, accelerated convergence of extension of Bragg intensities and structure factors, and sharpening in reciprocal space can also be applied.

This article illustrates that in dealing with partially coherent wave in an imaging system, the coherent component and the incoherent component can be treated separately. The results can then be combined to give the result for partially coherent wave. The problem of partially coherent wave in an imaging system reduces to the problems of coherent wave and incoherent wave in the imaging system. For the inverse problem in lensless imaging, the approaches of Yuen (2011v) and this article are complementary to each other.

This article may be applied to acoustic wave and matter wave, including electromagnetic wave, X-ray, electron, ion and neutron waves. For optical wave, acoustic wave and matter wave, including electromagnetic wave, X-ray, electron, ion and neutron waves, use Zarubin (1993), (4.2) for three-dimensional object and (5.9) for planar object.

2.1. Lensless imaging. Direct and inverse problems

(A) Direct problem

An object is represented by two functions. It exhibits a real or complex $f(\mathbf{x}_o)$ under coherent illumination of unit quantity, and exhibits a non-negative function $\rho(\mathbf{x}_o)$ under incoherent illumination of unit quantity. $f(\mathbf{x}_o)$ and $\rho(\mathbf{x}_o)$ have the same support.

Consider a quasi-monochromatic partially coherent scalar wave of mean angular frequency $\bar{\omega}$, which is a combination of a coherent wave and an incoherent wave. This partially coherent wave is incident on a finite-size, planar or three-dimensional object $f(\mathbf{x}_o)$ and $\rho(\mathbf{x}_o)$. $\bar{\omega} = c\bar{k}$. c is the velocity of light. $\bar{k}$ is the mean wave number. $f(\mathbf{x}_o)$ satisfies the constraints in Yuen (2011a, 2011h or 2011i). $\rho(\mathbf{x}_o)$ is non-negative and satisfies the constraints in Yuen (2011a or 2011h).

Consider a partially coherent object illumination

$$W = c_{10}W_c + c_{01}W_i. \tag{1}$$

c_{10} is real or complex. c_{01} is non-negative. Assume that $|c_{10}|$ and c_{01} have been determined (for example, Yuen (2011w), Sect. 2).

The object exhibits $c_{10} f(\mathbf{x}_o)$ and $c_{01}\rho(\mathbf{x}_o)$ (2)

(Yuen (2011w), (8)).

Yuen (2011w), Sect. 3 shows that
the mutual intensity of the quasi-monochromatic partially coherent wave
= mutual intensity of the coherent wave
+ mutual intensity of the incoherent wave. (3)

The superposition of the coherent illumination and the incoherent illumination or their evolutions in imaging, is always incoherent. Their intensities, mutual coherence functions, mutual intensities, cross-spectral densities and radiant intensities add.

For coherent illumination of unit quantity, the intensity of the object

$$I_{oco}(\mathbf{x}_o) = |f(\mathbf{x}_o)|^2 \tag{4}$$

The mutual intensity of the object $J_{oco}(\mathbf{x}_{o1}, \mathbf{x}_{o2}) = f(\mathbf{x}_{o1}) f^*(\mathbf{x}_{o2})$, (5)

(Yuen (2011t), (4))

From (2011t), (1), the mutual coherence function of the object,

$$\Gamma_{oco}(\mathbf{x}_{o1}, \mathbf{x}_{o2}, \tau) = J_{oco}(\mathbf{x}_{o1}, \mathbf{x}_{o2}) \exp(-i\overline{\omega}\tau)$$

The first Born approximation is used.

$\mathbf{K}$ = wavevector of scattered wave – wavevector of incident wave

In the far field of the object, the diffraction wave is given by $F(\mathbf{K})$.

From Yuen (2011t), (5), the intensity of the diffraction wave

$$I_{co}(\mathbf{K}) = |F(\mathbf{K})|^2 . \tag{6}$$

From Yuen (2011t), (6), the mutual intensity in the far field,

$$J_{co}(\varsigma_1, \varsigma_2) = (1/r^2) F^*(\mathbf{K}_1) F(\mathbf{K}_2) \tag{7}$$

The scattered wave is coherent (Wolf (2007), p. 117).

$J_{co}(\varsigma_1, \varsigma_2)$ is a six-dimensional Fourier transform of $(1/r^2) f^*(-\mathbf{x}_{o1}) f(\mathbf{x}_{o2})$

(Yuen (2011t), Sect. 2.1, (C)).

For incoherent illumination of unit quantity, the intensity of the object

$$I_{oin}(\mathbf{x}_o) = \rho(\mathbf{x}_o) \tag{8}$$

The mutual intensity of the object

$$J_{oin}(\mathbf{x}_{o1}, \mathbf{x}_{o2}) = \rho(\mathbf{x}_{o2})\delta(\mathbf{x}_{o2} - \mathbf{x}_{o1}) \tag{9}$$

(Yuen (2011u), (4)).

The mutual coherence function of the object

$$\Gamma_{oin}(\mathbf{x}_{o1}, \mathbf{x}_{o2}, \tau) = \rho(\mathbf{x}_{o2})\exp(-i\overline{\omega}\tau)\delta(\mathbf{x}_{o2} - \mathbf{x}_{o1}),$$

(Yuen (2011u), (3)).

From Yuen (2011u), (1)), the diffraction intensity in the far field

$$I_{in}(\mathbf{K}) = \int \rho(\mathbf{x}_o) d\mathbf{x}_o, \tag{10}$$

the first Born approximation being used.

By the van Cittert-Zernike theorem for planar object or the generalized van Cittert-Zernike theorem for three-dimensional object (Yuen (2011u), Sect. 2.1), the mutual intensity in the far field,

$$J_{in}(\varsigma_1, \varsigma_2) = (1/r^2)\mathrm{P}(\mathbf{K}) \tag{11}$$

$\mathrm{P}(\mathbf{K})$ is the three-dimensional Fourier transform of $\rho(\mathbf{x}_o)$. The first born approximation is not used

For the partially coherent object illumination, the total intensity of the object ((4), (8), Yuen (2011w), (6))

$$I_o(\mathbf{x}_o) = \left|c_{10} f(\mathbf{x}_o)\right|^2 + c_{01}\rho(\mathbf{x}_o) \tag{12}$$

The total mutual intensity of the object

$$J_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}) = \left|c_{10}\right|^2 J_{oco}(\mathbf{x}_{o1}, \mathbf{x}_{o2}) + c_{01} J_{oin}(\mathbf{x}_{o1}, \mathbf{x}_{o2}) \tag{13}$$

The total mutual coherence function of the object

$$\Gamma_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}, \tau) = \left|c_{10}\right|^2 \Gamma_{oco}(\mathbf{x}_{o1}, \mathbf{x}_{o2}, \tau) + c_{01}\Gamma_{oin}(\mathbf{x}_{o1}, \mathbf{x}_{o2}, \tau)$$

The total diffraction intensity of the wave in the far field of the object (from (6) and (10)),

$$I(\mathbf{K}) = \left|c_{10} F(\mathbf{K})\right|^2 + \int c_{01} \rho(\mathbf{x}_o) d\mathbf{x}_o \tag{14}$$

From (7) and (11), the total mutual intensity in the far field of the object,

$$J(\varsigma_1, \varsigma_2) = \left|c_{10}\right|^2 J_{co}(\varsigma_1, \varsigma_2) + c_{01} J_{in}(\varsigma_1, \varsigma_2) \tag{15}$$

The mean wavelength $\overline{\lambda}$ of the incident wave is chosen according to the resolution needed. A three-dimensional object can be rotated so that all diffraction intensity and the magnitude of all mutual intensity in the far field $\left|J(\varsigma_1, \varsigma_2)\right|$ for the chosen resolution can be measured

(B) Inverse problem using the total diffraction intensity in the far field of the object

In (14), the minimum value of $I(\mathbf{K})$,

$$I(\mathbf{K})_{\min} = \int c_{01} \rho(\mathbf{x}_o) d\mathbf{x}_o \tag{16}$$

From (14) and (16), $\left|c_{10} F(\mathbf{K})\right|$ is determined.

As in Yuen (2011t), Sect. 2.1, (B), iterative algorithms with oversampling in Yuen (2011c or 2011k) are applied to obtain the phase of $c_{10} F(\mathbf{K})$ from the magnitude. Powerful methods in structure determination in X-ray crystallography, such as sharpening of Patterson function, direct methods, point-atom model, accelerated convergence of extension of Bragg intensities and structure factors, and sharpening in reciprocal space can also be applied (Yuen (2011q)). The solution $c_{10} f(\mathbf{x}_o)$ is obtained.

The intensity of the object for the coherent component (from (4))

$$= \left|c_{10}\right|^2 I_{oco}(\mathbf{x}_o) = \left|c_{10} f(\mathbf{x}_o)\right|^2 \tag{17}$$

The mutual intensity of the object for the coherent component (from (5))

$$=|c_{10}|^2 J_{oco}(\mathbf{x}_{o1},\mathbf{x}_{o2}) = |c_{10}|^2 f(\mathbf{x}_{o1}) f^*(\mathbf{x}_{o2}) \tag{18}$$

The mutual intensity in the far field for the coherent component (from (7))

$$=|c_{10}|^2 J_{co}(\varsigma_1,\varsigma_2) = (1/r^2)|c_{10}|^2 F^*(\mathbf{K}_1)F(\mathbf{K}_2) \tag{19}$$

(C) Inverse problem using the total mutual intensity in the far field of the object

In (15), $J(\varsigma_1,\varsigma_2)$ is the sum of a six-dimensional Fourier transform and a three-dimensional Fourier transform. Hence we cannot, based on (15), use our phase-retrieval method to obtain the phase of $J(\varsigma_1,\varsigma_2)$ from the magnitude. However, according to Yuen (2011v), Sect. 2.1, (B), a phase-retrieval method can, in fact, be applied to obtain the phase of the mutual intensity, $J(\varsigma_1,\varsigma_2)$ in the far field of the object.

As in Yuen (2011u), Sect. 2, (B), iterative algorithms with oversampling in Yuen (2011k) is applied to obtain the phase of $J(\varsigma_1,\varsigma_2)$ from the magnitude. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011q)).

From (15), $c_{01}J_{in}(\varsigma_1,\varsigma_2) = J(\varsigma_1,\varsigma_2) - |c_{10}|^2 J_{co}(\varsigma_1,\varsigma_2)$

Since $|c_{10}|^2 J_{co}(\varsigma_1,\varsigma_2)$ is determined in (19), the mutual intensity due to the incoherent component $c_{01}J_{in}(\varsigma_1,\varsigma_2)$ is determined. Hence, from the van Cittert-Zernike theorem for planar object or the generalized van Cittert-Zernike theorem for three-dimensional object, $c_{01}\rho(\mathbf{x}_o)$ is determined. Substitute this value into (16), the result obtained should agree with the experimental value of $I(\mathbf{K})_{\min}$.

The total intensity of the object is given by (12).

The total mutual intensity of the object is given by (13).

The resolution of the object $=\overline{\lambda}/2$

(Zarubin (1993), p. 502, Totzeck & Krumbügel (1994), p. 189, Goodman (2005), (3-74)).

Using (12), the intensity at the surface of the object = $\int I_o(\mathbf{x}_o)dt$ (20)

The one-dimensional integral is over the thickness of the object

The intensity at the surface of the object obtained from (20) should be the same as the intensity at the surface of the object obtained in Yuen (2011v), (4).

From Iizuka (2008), (10.22), and Goodman (2005), (6-14),

$$|f(\mathbf{x}_o)|^2 = \rho(\mathbf{x}_o)\,. \tag{21}$$

$|f(\mathbf{x}_o)|$ and $\rho(\mathbf{x}_o)$ obtained should satisfy this relation.

In fact, in (B) and (C), $|c_{10}|$ and c_{01} need not be known.

2.2. Image of object in a lens, phase-retrieval microscope, phase-retrieval telescope. Direct and inverse problems

(A) Direct problem

This is a continuation of Sect. 2.1, Lensless imaging, (A) Direct problem.

Assume that $|c_{10}|$ and c_{01} have been determined. As in Yuen (2011t), Sect. 2.2, a lens is used to achieve superresolution. The first Born approximation is not used.

Consider quasi-monochromatic object illuminations along the axis of the lens.

For the coherent illumination of unit intensity, the image is $g(\mathbf{x})$ (Yuen (2011t), Sect. 2.2, (A), (13)).

$$g(\mathbf{x}) = f(\mathbf{x}) \otimes h(\mathbf{x}) \tag{22}$$

$h(\mathbf{x})$ is the impulse response.

The mutual intensity of the image $J_{ico}(\mathbf{x}_1, \mathbf{x}_2) = g(\mathbf{x}_1)g^*(\mathbf{x}_2)$, (23)

(Yuen (2011t), (12)).

The intensity of the image, $I_{ico}(\mathbf{x}) = J_{ico}(\mathbf{x}, \mathbf{x}) = |g(\mathbf{x})|^2$, (24)

(Yuen (2011t), (16)).

The image $g(\mathbf{x})$ of the coherent object acts as a secondary source. From Yuen (2011t), (17), the cross-spectral density function in the far field of the image,

$$W_{co}(\varsigma_1\mathbf{u}_1,\varsigma_2\mathbf{u}_2,\varpi) = (2\pi)^6 G(\bar{k}\mathbf{u}_1)G^*(\bar{k}\mathbf{u}_2)\{\exp[i\bar{k}(\varsigma_1-\varsigma_2)]\}/(\varsigma_1\varsigma_2) \tag{25}$$

The mutual intensity in the far field,

$$J_{co}(\varsigma_1,\varsigma_2) = (2\pi)^6 G(\bar{k}\mathbf{u}_1)G^*(\bar{k}\mathbf{u}_2)\{\exp[i\bar{k}(\varsigma_1-\varsigma_2)]\}/(\varsigma_1\varsigma_2) \tag{26}$$

(see Yuen (2011t), (2)).

From Yuen (2011t), (18), the radiant intensity in the far field

$$\Lambda_{co}(\mathbf{u},\varpi) = (2\pi)^6 \left|G(\bar{k}\mathbf{u})\right|^2, \tag{27}$$

For the incoherent illumination of unit intensity, the mutual intensity $J_{iin}(\mathbf{x}_1,\mathbf{x}_2)$ of the image is obtained in Yuen (2011u), (9).

The intensity of the image $\psi(\mathbf{x}) = J_{iin}(\mathbf{x},\mathbf{x}) = \rho(\mathbf{x}) \otimes \left|h(\mathbf{x})\right|^2$, (28)

(Yuen (2011u), (10)).

The image of the incoherent object acts as a secondary source. From Yuen (2011u), (12), the cross-spectral density function in the far field of the image,

$$W_{in}(\varsigma_1\mathbf{u}_1,\varsigma_2\mathbf{u}_2,\varpi) = (2\pi)^6 \widetilde{W}_i(\bar{k}\mathbf{u}_1,-\bar{k}\mathbf{u}_2,\varpi)\{\exp[i\bar{k}(\varsigma_1-\varsigma_2)]\}/(\varsigma_1\varsigma_2) \tag{29}$$

The mutual intensity in the far field,

$$J_{in}(\varsigma_1,\varsigma_2) = (2\pi)^6 \widetilde{W}_i(\bar{k}\mathbf{u}_1,-\bar{k}\mathbf{u}_2,\varpi)\{\exp[i\bar{k}(\varsigma_1-\varsigma_2)]\}/(\varsigma_1\varsigma_2) \tag{30}$$

From Yuen (2011u), (13), the radiant intensity in the far field,

$$\Lambda_{in}(\mathbf{u},\varpi) = (2\pi)^6 \widetilde{W}_i(\bar{k}\mathbf{u},-\bar{k}\mathbf{u},\varpi) \tag{31}$$

For the partially coherent object illumination, the total mutual intensity of the image is obtained from (23) and Yuen (2011u), (9),

$$J_i(\mathbf{x}_1,\mathbf{x}_2)=\left|c_{10}\right|^2 J_{ico}(\mathbf{x}_1,\mathbf{x}_2)+c_{01}J_{iin}(\mathbf{x}_1,\mathbf{x}_2) \tag{32}$$

The total intensity of the image is obtained from (24) and (28),

$$I_i(\mathbf{x})=\left|c_{10}\right|^2 I_{ico}(\mathbf{x})+c_{01}\psi(\mathbf{x}) \tag{33}$$

The total intensity and the magnitude of the total mutual intensity of the image are measured.

From (25) and (29), the total cross-spectral density function in the far field of the image

$$W(\varsigma_1\mathbf{u}_1,\varsigma_2\mathbf{u}_2,\varpi)=\left|c_{10}\right|^2 W_{co}(\varsigma_1\mathbf{u}_2,\varsigma_2\mathbf{u}_2,\varpi)+c_{01}W_{in}(\varsigma_1\mathbf{u}_1,\varsigma_2\mathbf{u}_2,\varpi)$$

From (26) and (30), the total mutual intensity in the far field

$$J(\varsigma_1,\varsigma_2)=\left|c_{10}\right|^2 J_{co}(\varsigma_1,\varsigma_2)+c_{01}J_{in}(\varsigma_1,\varsigma_2) \tag{34}$$

From (27) and (31), the total radiant intensity in the far field

$$\Lambda(\mathbf{u},\varpi)=\left|c_{10}\right|^2 \Lambda_{co}(\mathbf{u},\varpi)+c_{01}\Lambda_{in}(\mathbf{u},\varpi) \tag{35}$$

For a three-dimensional object, use all orientations of the object. Depending on the chosen resolution, we use a maximum mean wave number $\bar{k}_{\max}$. For a planar or three-dimensional object, use separately all incident quasi-monochromatic waves of same intensity with mean wave number $\bar{k}$, $\bar{k}\le\bar{k}_{\max}$. The radiant intensity and the magnitude of the mutual intensity in the far field are measured.

(B) Inverse problem using the total mutual intensity in the far field of the image

As in Yuen (2011t), (21), apart from the factor $(2\pi)^6\left\{\exp[i\bar{k}(\varsigma_1-\varsigma_2)]\right\}/(\varsigma_1\varsigma_2)$, the mutual intensity $J_{co}(\varsigma_1,\varsigma_2)$ in the far field (see (26)) is the six-dimensional (or four-dimensional for planar object) spatial Fourier transform of $g(\mathbf{x}_1)g^*(-\mathbf{x}_2)$.

As in Yuen (2011u), Sect. 2.2, apart from the factor

$(2\pi)^6\{\exp[i\bar{k}(\varsigma_1-\varsigma_2)]\}/(\varsigma_1\varsigma_2)$, the mutual intensity $J_{in}(\varsigma_1,\varsigma_2)$ in the far field (see (30)) is the six-dimensional (or four-dimensional for planar object) spatial Fourier transform of $J_{iin}(\mathbf{x}_1,-\mathbf{x}_2)$. Hence the total mutual intensity $J(\varsigma_1,\varsigma_2)$ in the far field of the image is the six-dimensional (or four-dimensional for planar object) spatial Fourier transform of

$$(2\pi)^6[|c_{10}|^2 g(\mathbf{x}_1)g^*(-\mathbf{x}_2)+c_{01}J_{iin}(\mathbf{x}_1,-\mathbf{x}_2)]\{\exp i\bar{k}(\varsigma_1-\varsigma_2)]\}(\varsigma_1\varsigma_2).$$

$$=(2\pi)^6 J_i(\mathbf{x}_1,-\mathbf{x}_2)\{\exp i\bar{k}(\varsigma_1-\varsigma_2)]\}(\varsigma_1\varsigma_2).$$

from (32).

As in Yuen (2011u), Sect. 2.2, (B), iterative algorithms with oversampling in Yuen (2011k) are applied to obtain the phase of $J(\varsigma_1,\varsigma_2)$ in the far field from the magnitude. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011q)). The solution

$(2\pi)^6 J_i(\mathbf{x}_1,-\mathbf{x}_2)\{\exp i\bar{k}(\varsigma_1-\varsigma_2)]\}(\varsigma_1\varsigma_2)$ is obtained.

From (32), $J_i(\mathbf{x}_1,-\mathbf{x}_2)=|c_{10}|^2 g(\mathbf{x}_1)g^*(-\mathbf{x}_2)+c_{01}J_{iin}(\mathbf{x}_1,-\mathbf{x}_2)$ (36)

Replace $-\mathbf{x}_2$ by $\mathbf{x}_2$. Mutual intensity of the image $J_i(\mathbf{x}_1,\mathbf{x}_2)$ is determined. Its magnitude should agree with the value measured in (A).

For $\mathbf{x}_1=\mathbf{x}_2=\mathbf{x}$, the intensity of the image

$$I_i(\mathbf{x})=|c_{10}|^2 g(\mathbf{x})g^*(\mathbf{x})+c_{01}J_{iin}(\mathbf{x},\mathbf{x})=|c_{10}|^2 I_{ico}(\mathbf{x})+c_{01}\psi(\mathbf{x}) \qquad (37)$$

is obtained. This intensity should agree with the value measured in (A).

We try to deal with the inverse problem in terms of the coherent and incoherent components. From (22) and (28), the intensity of the image in (37),

$$I_i(\mathbf{x}) = |c_{10}|^2 |f(\mathbf{x}) \otimes h(\mathbf{x})|^2 + c_{01}\rho(\mathbf{x}) \otimes |h(\mathbf{x})|^2 . \tag{38}$$

The impulse response $h(\mathbf{x})$ is close to a δ function (Goodman (2005), (5-24)).

Hence $I_i(\mathbf{x}) \approx |c_{10}|^2 |f(\mathbf{x}) \otimes \delta(\mathbf{x})|^2 + c_{01}\rho(\mathbf{x}) \otimes |\delta(\mathbf{x})|^2$

$\delta^2(\mathbf{x})$ is undefined (Papoulis (1962), p. 275, Folland (1992), p. 319). However, we are only using an approximate δ function, and $\delta^2(\mathbf{x})$ has shape close to that of $\delta(\mathbf{x})$. Therefore $I_i(\mathbf{x}) \approx |c_{10}|^2 |f(\mathbf{x})|^2 + c_{01}\rho(\mathbf{x})$

$$= (|c_{10}|^2 + c_{01})|f(\mathbf{x})|^2$$

from (21).

Hence an approximate $|f(\mathbf{x})|^2$ (and $\rho(\mathbf{x})$) is obtained. Use these as initial values and substitute into (38). By fine adjustments, subjected to the condition of (21), more accurate $|f(\mathbf{x})|^2$ and $\rho(\mathbf{x})$ may be obtained. In the adjustment, the phase of $f(\mathbf{x})$ is a variable.

Approximate intensity of the object is given by (12).

2.3. Inverse problem using the total mutual intensity in the far field of the image. Use one more quasi-monochromatic partially coherent wave

The method in Sect. 2.2, (B) is not very satisfactory as it involves numerical method and that the phase of $f(\mathbf{x})$ is a variable. Conceptually it will be better if we can obtain $J_{co}(\varsigma_1, \varsigma_2)$ and $J_{in}(\varsigma_1, \varsigma_2)$ from $J(\varsigma_1, \varsigma_2)$ (see (34)), or divide $J_i(\mathbf{x}_1, -\mathbf{x}_2) = |c_{10}|^2 g(\mathbf{x}_1) g^*(-\mathbf{x}_2) + c_{01} J_{iin}(\mathbf{x}_1, -\mathbf{x}_2)$ (see (36)), into two parts for the coherent and incoherent components. Then we can use the solutions to the inverse problems for the coherent and incoherent components obtained in Yuen (2011t), Sect. 2.2 (C), and Yuen (2011u), Sect. 2.2, (B).

We may use an analytical method. We make use of the fact that there are non-denumerable numbers of partially coherent waves, while there are only one coherent wave and one incoherent wave; there is only one $f(\mathbf{x}_o)$ and one $\rho(\mathbf{x}_o)$. We use one more quasi-monochromatic partially coherent wave

$$W = c_{10}^{'} W_c + c_{01}^{'} W_i \tag{39}$$

with mean wave number $\bar{k}$, $\bar{k} \le \bar{k}_{\max}$. $\left|c_{10}^{'}\right|$ and $c_{01}^{'}$ have been determined.

By means of iterative algorithms with oversampling in Yuen (2011k), $J'(\varsigma_1, \varsigma_2)$ is obtained in a way similar to Sect. 2.2, (B). Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011q)).

$$J'(\varsigma_1, \varsigma_2) = \left|c_{10}^{'}\right|^2 J_{co}(\varsigma_1, \varsigma_2) + c_{01}^{'} J_{in}(\varsigma_1, \varsigma_2) \tag{40}$$

From (34) and (40), $J_{co}(\varsigma_1, \varsigma_2)$ and $J_{in}(\varsigma_1, \varsigma_2)$ are obtained.
$J_i'(\mathbf{x}_1, -\mathbf{x}_2)$ is obtained in a similar way.

$$J_i'(\mathbf{x}_1 - \mathbf{x}_2) = \left|c_{10}^{'}\right|^2 g(\mathbf{x}_1) g^*(-\mathbf{x}_2) + c_{01}^{'} J_{iin}(\mathbf{x}_1, -\mathbf{x}_2) \tag{41}$$

From (36) and (41), $g(\mathbf{x}_1) g^*(-\mathbf{x}_2)$ and $J_{iin}(\mathbf{x}_1, -\mathbf{x}_2)$ are obtained.

For the coherent component, $g(\mathbf{x}_1) g^*(-\mathbf{x}_2)$, the results of Yuen (2011t), Sect. 2.2, (C) can then be obtained: the radiant intensity in the far field of the image,
the mutual intensity of the image,
intensity of the image,

$|g(\mathbf{x})|$,

mutual intensity of the object,
intensity of the object,

$|f(\mathbf{x}_o)|$.

For the incoherent component, $J_{iin}(\mathbf{x}_1,-\mathbf{x}_2)$, the results of Yuen (2011u), Sect. 2.2, (B) can then be obtained: the radiant intensity in the far field of the image,

mutual intensity of the image,
intensity of the image,
$\rho(\mathbf{x}_o)$,
mutual intensity of the object.

$|f(\mathbf{x}_o)|$ and $\rho(\mathbf{x}_o)$ obtained should satisfy (21).

Add the results for the coherent and incoherent components, the results for the partially coherent object illumination are obtained.

Natural resolution of the object is dealt with. Empirical amplitude transfer function $H(\mathbf{K})$ and empirical optical transfer function $\Theta(\mathbf{K})$ determined in Sect. 2.3 of Yuen (2008) will be used. They take into account aberrations of the lens and other defects of the system. We have by-passed the Sparrow criterion of resolution (or alike) of the image, and the corresponding angular resolution.

2.4. Lenslets microscope and lenslets telescope. Direct and inverse problems

(A) Direct problem

Follow Sect. 2.2 and Yuen (2011v), Sect. 2.3. Two-dimensional array of lenslets can be used to form lenslets microscope or lenslets telescope. The array of lenslets forms a linear system. Its impulse response is formed by adding replica of $h(\mathbf{x}) \equiv h(x_1, x_2)$ for one lenslet successfully in x_1, x_2 to give a periodic function, the extended $h(\mathbf{x})$. The periods are chosen such that the Fourier coefficients of this extended $h(\mathbf{x})$ are nonzero (Yuen (2008), (D)). The focal length of the lenslets, the object distance and the unit cell constants a, b of the array of lenslets are chosen so that the image predicted by geometrical optics in one lenslet can be contained in one unit cell (Yuen (2008), (D), constraint (i)).

Assume that $|c_{10}|$ and c_{01} have been determined.

In the far field, the radiant intensities and mutual intensities of the images in the lenslets are identical. For an incident wave of wave number $\bar{k}$ upon the nonperiodic object, the weak radiant intensities and mutual intensities of the two-dimensional array of images of the object are collected to form strong diffraction rods; it is enhanced by the array of images. The radiant intensity and the magnitude of the mutual intensity $|J(\varsigma_1,\varsigma_2)|$ of the diffraction rods of the image in all the lenslets are measured. For a three-dimensional object, use all orientations of the object. Depending on the chosen resolution, we use a maximum mean wave number $\bar{k}_{\max}$. For a planar or three-dimensional object, use separately all incident quasi-monochromatic waves of same intensity with mean wave number $\bar{k}$, $\bar{k} \le \bar{k}_{\max}$.

(B) Inverse problem using the mutual intensity of the diffraction rods of the image in all the lenslets

Follow Sect. 2.2 and Yuen (2011v), Sect. 2.3. Iterative algorithms with oversampling in Yuen (2011n) are applied to obtain the phase of the mutual intensity of the diffraction rods $J(\varsigma_1,\varsigma_2)$ from the magnitude. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011s)). Use one more quasi-monochromatic partially coherent waves

$$W = c_{10}^{'}W_c + c_{01}^{'}W_i \tag{39}$$

Assume that $|c_{10}^{'}|$ and $c_{01}^{'}$ have been determined

We obtain $J_{co}(\varsigma_1,\varsigma_2)$ and $J_{in}(\varsigma_1,\varsigma_2)$ of the diffraction rods.

Consider the coherent component $J_{co}(\varsigma_1,\varsigma_2)$. From (26), apart from the factor $(2\pi)^6\{\exp[i\bar{k}(\varsigma_1-\varsigma_2)]\}/(\varsigma_1\varsigma_2)$, the mutual intensity in the far field

$J_{co}(\varsigma_1,\varsigma_2)$ is the six-dimensional (or four-dimensional for planar object) spatial Fourier transform of the $g_a(\mathbf{x}_1)g_a{}^*(-\mathbf{x}_2)$ of the image in all lenslets (Yuen (2011t), Sect. 2.3, (C)). Hence $g_a(\mathbf{x}_1)g_a{}^*(-\mathbf{x}_2)$ is obtained. The results of Yuen (2011t), Sect. 2.3, (C) can then be used. Approximate value of the intensity of the image in one lenslet is obtained, in particular, if the unit cell is sufficiently large.

Consider the incoherent component $J_{in}(\varsigma_1,\varsigma_2)$. In (30), apart from the factor $(2\pi)^6\{\exp[i\bar{k}(\varsigma_1-\varsigma_2)]\}/(\varsigma_1\varsigma_2)$, the mutual intensity $J_{in}(\varsigma_1,\varsigma_2)$ in the far field is the six-dimensional (or four-dimensional for planar object) spatial Fourier transform of $J_{il}(\mathbf{x}_1,-\mathbf{x}_2)$ of the image in all lenslets (Yuen (2011u), Sect. 2.3, (B). Hence $J_{il}(\mathbf{x}_1,-\mathbf{x}_2)$ is obtained. The results of Yuen (2011u), Sect. 2.3, (B) can then be used. The object $\rho(\mathbf{x}_o)$ is determined. The intensity of the image in one lenslet

$$\psi(\mathbf{x})=\rho(\mathbf{x})\otimes\left|h(\mathbf{x})\right|^2,$$

(see (28)).

For the partially coherent wave, the intensity of the image in one lenslet is given by the sum of the approximate intensity of the image in one lenslet for the coherent component and $\psi(\mathbf{x})$. The Sparrow criterion of resolution of the approximate intensity of the image in one lenslet (or alike) and the corresponding angular resolution are applied. Natural resolution of the object is not dealt with.

2.5. Crystal diffraction for quasi-monochromatic partially coherent wave. Direct and inverse problems

(A) Direct problem

Sect. 2.1 can be applied to diffraction of partially coherent wave by crystals, namely, Yuen (2011a, 2011h, 2011i), (B), (D). The total diffraction intensity and the magnitude of the total mutual intensity of the diffraction points are measured.

(B) Inverse problem using the diffraction intensity of the diffraction points

For phase-retrieval method, see Yuen (2011a, 2011h, 2011i), (B), (D). For iterative algorithms with oversampling, see Yuen (2011b or 2011j). Powerful methods in structure determination in X-ray crystallography, such as sharpening of Patterson function, direct methods, point-atom model, accelerated convergence of extension of Bragg intensities and structure factors, and sharpening in reciprocal space can also be applied (Yuen (2011o, 2011p)). $f(\mathbf{x}_o)$ of the crystal is determined.

(C) Inverse problem using the mutual intensity of the diffraction points

For phase-retrieval method, see Yuen (2011h, 2011i), (B), (D). For iterative algorithms with oversampling, see Yuen (2011j). Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011o, 2011p)). $\rho(\mathbf{x}_o)$ of the crystal is determined.

2.6. Fibre diffraction for quasi-monochromatic partially coherent wave.

Consider a fibre (freely rotating) periodic in x_3 with period c. Sect. 2.1 is applied. Because of the one-dimensional periodic structure, the diffraction intensity and mutual intensity are greatly enhanced to form strong diffraction planes which can be measured accurately. In the following about projections, we will apply Yuen (2011d), modified for projections which may be non-centrosymmetric or complex.

2.6.1. Projection of one period of a fibre (freely rotating) to a plane normal to the fibre axis

These are inverse problems. Diffraction intensity or magnitude of the mutual intensity of the diffraction plane for $\omega_3 = 0$ is used. For phase-retrieval method, see Yuen (2011d), Sect. 2.1. For iterative algorithms with oversampling, see Yuen (2011d), Sect. 2.2.

(A) Diffraction intensity of the diffraction plane for $\omega_3 = 0$ is used

Ab initio $f_p(x_{o1}, x_{o2})$ of the projection is determined.

(B) Magnitude of the mutual intensity of the diffraction plane for $\omega_3 = 0$ is used

Ab initio $\rho_p(x_{o1}, x_{o2})$ of the projection is determined

2.6.2. Projection to a plane containing the fibre axis.

Diffraction intensity or magnitude of the mutual intensity of the diffraction lines for $\omega_1 = 0$ is used. For phase-retrieval method, see Yuen (2011d), Sect. 2.3. For iterative algorithms with oversampling, see Yuen (2011d), Sect. 2.4.

(A) Diffraction intensity of the diffraction lines for $\omega_1 = 0$ is used

Ab initio $f_h(x_{o2}, x_{o3})$ of the projection is determined.

(B) Magnitude of the mutual intensity of the diffraction lines for $\omega_1 = 0$ is used

Ab initio $\rho_h(x_{o2}, x_{o3})$ of the projection are determined.

2.6.3. Projection to an axis normal to the fibre axis

Diffraction intensity or magnitude of the mutual intensity of the diffraction line for $\omega_2 = \omega_3 = 0$ is used. For phase-retrieval method, see Yuen (2011d), Sect. 2.5. For iterative algorithms with oversampling, see Yuen (2011d), Sect. 2.6.

(A) Diffraction intensity of the diffraction line for $\omega_2 = \omega_3 = 0$ is used

Ab initio $f_n(x_{o1})$ of the projection is determined.

B) Magnitude of the mutual intensity of the diffraction line for $\omega_2 = \omega_3 = 0$ is used

Ab initio $\rho_n(x_{o1})$ of the projection is determined.

2.6.4. Projection to the fibre axis

Diffraction intensity or magnitude of the mutual intensity of the diffraction line for $\omega_1 = \omega_2 = 0$ is used. For phase-retrieval method, see Yuen (2011d), Sect. 2.7. For iterative algorithms with oversampling, see Yuen (2011d), Sect. 2.8.

(A) Diffraction intensity of the diffraction line for $\omega_1 = \omega_2 = 0$ is used

Ab initio $f_c(x_{o3})$ of the projection is determined.

(B) Magnitude of the mutual intensity of the diffraction line for $\omega_1 = \omega_2 = 0$ is used

Ab initio $\rho_c(x_{o3})$ of the projection are determined.

Treatment for projections of a fixed fibre can also be obtained.

2.6.5. Diffraction of partially coherent wave by a fibre (freely rotating) or a fixed fibre

(A) Direct problem

Sect. 2.1 can be applied to diffraction of partially coherent wave by a fibre (freely rotating) or a fixed fibre, namely, Yuen (2011e, 2011m), (1), (3).

(B) Inverse problem using the diffraction intensity of the diffraction planes

For phase-retrieval method, see Yuen (2011e, 2011m), Sect. 2.1, 3.1. For iterative algorithms with oversampling, see Yuen (2011e), Sect. 2.2, 3.2, and Yuen (2011m), Sect. 2.2, 3.2 – Sect. 5. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011r)). *Ab initio* $f(\mathbf{x}_o)$ of the fibre is determined.

(C) Inverse problem using the magnitude of the mutual intensity of the diffraction planes

For phase-retrieval method, see Yuen (2011m), Sect. 2.1, 3.1. For iterative algorithms with oversampling, see Yuen (2011m), Sect. 2.2, 3.2 – Sect. 5. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011r)). *Ab initio* $\rho(\mathbf{x}_o)$ is determined.

2.7. Membrane diffraction for quasi-monochromatic partially coherent wave.

Consider a planar membrane, periodic in x_1 and x_2, with periods a and b. Sect. 2.1 is applied. Because of the two-dimensional periodic structure, the diffraction intensity and mutual intensity are greatly enhanced to form strong diffraction rods which can be measured accurately. In the following about projections, we will apply Yuen (2011f), modified for projections which may be non-centrosymmetric or complex.

2.7.1. Projection of one period of the membrane upon an axis normal to the membrane.

These are inverse problems. Diffraction intensity or magnitude of the mutual intensity of the diffraction rod for $\omega_1 = \omega_2 = 0$ is used. For phase-retrieval method, see Yuen (2011f), Sect. 2.1. For iterative algorithms with oversampling, see Yuen (2011f), Sect. 2.2.

(A) Diffraction intensity of the diffraction rod for $\omega_1 = \omega_2 = 0$ is used

Ab initio $f_n(x_{o3})$ of the projection is determined.

(B) Magnitude of the mutual intensity of the diffraction rod for $\omega_1 = \omega_2 = 0$ is used

Ab initio $\rho_n(x_{o3})$ of the projection is determined.

2.7.2. Projection to the plane of the membrane

Diffraction intensity or magnitude of the mutual intensity of the diffraction points for $\omega_3 = 0$ is used. For phase-retrieval method, see Yuen (2011f), Sect. 2.3. For iterative algorithms with oversampling, see Yuen (2011f), Sect. 2.4.

(A) Diffraction intensity of the diffraction points for $\omega_3 = 0$ is used

Ab initio $f_h(x_{o1}, x_{o2})$ of the projection is determined.

(B) Magnitude of the mutual intensity of the diffraction points for $\omega_3 = 0$ is used

Ab initio $\rho_h(x_{o1}, x_{o2})$ of the projection is determined.

2.7.3. Diffraction of partially coherent wave by a planar periodic membrane

(A) Direct problem

Sect. 2.1 can be applied to diffraction of partially coherent wave by a planar periodic membrane, namely, Yuen (2011g, 2011n), (1).

(B) Inverse problem using the diffraction intensity of the diffraction rods

For phase-retrieval method, see Yuen (2011g, 2011n), Sect. 2.1. For iterative algorithms with oversampling, see Yuen (2011g), Sect. 2.2, and Yuen (2011n), Sect. 2.2 – Sect. 3. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011s)). *Ab initio* $f(\mathbf{x}_o)$ of the membrane is determined.

(C) Inverse problem using the magnitude of intensity of the diffraction rods

For phase-retrieval method, see Yuen (2011n), Sect. 2.1. For iterative algorithms with oversampling, see Yuen (2011n), Sect. 2.2 – Sect. 3. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011s)). *Ab initio* $\rho(\mathbf{x}_o)$ of the membrane is determined.

3. Discussions and conclusion

This article deals with the direct and inverse problems of imaging of object irradiated with a quasi-monochromatic partially coherent light wave, which is a combination of a coherent wave and an incoherent wave. Diffraction intensity, mutual intensity or radiant intensity is dealt with. The coherent and incoherent components of a partially coherent wave are used.

In lensless imaging, for coherent component and diffraction intensity of incoherent component, the first Born approximation is used. For all other cases, the first Born approximation is not used.

For lensless imaging, in terms of $f(\mathbf{x}_o)$ and $\rho(\mathbf{x}_o)$, analytical expressions of the intensity $I_o(\mathbf{x}_o)$ and mutual intensity $J_o(\mathbf{x}_{o1}, \mathbf{x}_{o2})$ of the object are obtained. For the inverse problem, all orientations of the object are used to get all diffraction intensity and magnitude of mutual intensity in the far field of the object. Iterative algorithms with oversampling are applied to obtain the phase from the magnitudes of the diffraction intensity and mutual intensity. $c_{10} f(\mathbf{x}_o)$ and $c_{01}\rho(\mathbf{x}_o)$ are determined. In Yuen (2011v), $|f(\mathbf{x}_o)|$ is determined.

For imaging of an object in a lens with partially coherent illumination, in the direct problem, the radiant intensity and the magnitude of the mutual intensity in the far field of the image are measured. Mutual intensity is used in the inverse problem. Use separately all incident quasi-monochromatic waves of same intensity, c_{10} and c_{01}, with mean wave number $\bar{k}$, $\bar{k} \le \bar{k}_{\max}$. Magnitude of the mutual intensity in the far field is measured. Iterative algorithms with oversampling are applied to obtain the phase from the magnitude of the mutual intensity. Using a numerical method, $|f(\mathbf{x})|$

and $\rho(\mathbf{x})$ can be determined.

A conceptually better, analytical method is to use one more quasi-monochromatic partially coherent wave, the mutual intensity obtained is divided into coherent and incoherent components. For the coherent component, the results of Yuen (2011t), Sect. 2.2, (C) can be adopted. $|f(\mathbf{x}_o)|$ is determined. For the incoherent component, the results of Yuen (2011u), Sect. 2.2, (B) can be adopted. $\rho(\mathbf{x}_o)$ is determined. These results can then be added for the partially coherent illumination. Natural resolution of the object is dealt with. Empirical amplitude transfer function $H(\mathbf{K})$ and empirical optical transfer function $\Theta(\mathbf{K})$ are employed. These take into account aberrations of the lens and other defects of the system. We have by-passed the Sparrow criterion of resolution (or alike) of the image, and the corresponding angular resolution. In Yuen (2011v), only the mutual intensity and the intensity of the object are obtained.

For lenslets microscope or lenslets telescope, the radiant intensity and the magnitude of the mutual intensity of the diffraction rods of the image in all the lenslets are measured. For the inversion problem, mutual intensity is used. The total intensity of the image in one lenslet is given by the sum of the approximate intensity of the image for the coherent component and the intensity of the image for the incoherent component The Sparrow criterion of resolution of the approximate total intensity of the image (or alike) is applied. Natural resolution of the object is not dealt with.

Diffraction of partially coherent wave by a nonperiodic object, or crystal diffraction, fibre diffraction, and membrane diffraction are dealt with. Powerful methods in X-ray crystallography can also be applied.

The intensity at the surface of the object obtained from (20) should be the same as the intensity at the surface of the object obtained in Yuen (2011v), (4). The intensity of the object obtained in Sect. 2.2, (B) and Sect. 2.3, (B) should be the same as the intensity of the object obtained in Yuen (2011v), (9) and Sect. 2.3, (B). In lensless imaging in Yuen (2011v), $|f(\mathbf{x}_o)|$ is obtained. In this article, we obtain the object $f(\mathbf{x}_o)$ and $\rho(\mathbf{x}_o)$. As mentioned in Yuen (2011w), Sect. 4, this provides simplifications and more insight. In fact, the approaches of Yuen (2011v) and this article are complementary to each other. As in Yuen (2011v), Sect. 2.1, (B), it is stated that ‘a three-dimensional object can be rotated so that the magnitude of all mutual intensity for the chosen resolution can be measured. This statement is true because it is true when the partially coherent wave is treated as a combination of coherent and incoherent components (Yuen (2011u), Sect. 2.1, (A))’. In Sect. 2.1, (C) of this article, it is stated that ‘However, according to Yuen (2011v), Sect. 2.1, (B), a phase-retrieval method can, in fact, be applied to obtain the phase of the mutual intensity, $J(\varsigma_1,\varsigma_2)$ in the far field of the object.’

In Sect 2.1, (C), we have followed Yuen (2011v), Sect. 2.1, (B), and apply the phase-retrieval method. If this is not known to us, we may assume that $J(\varsigma_1,\varsigma_2)$ is the Fourier transform of an unknown object X. Then we try to apply iterative algorithms with oversampling. If phase of $J(\varsigma_1,\varsigma_2)$ can be obtained, we can then proceed as in Sect. 2.1, (C). Whether this assumption is correct or not depends on the results obtained.

Wolf (2007), Sect. 6.2, (6) use an object $f(\mathbf{x}_o)$ (see Yuen (2011v), (1)). In this article, we use two representations, $f(\mathbf{x}_o)$ and $\rho(\mathbf{x}_o)$, of an object. Because of the relation $\left|f(\mathbf{x}_o)\right|^2 = \rho(\mathbf{x}_o)$ in (21), we, in fact, use only one representation $f(\mathbf{x}_o)$, just like what is done in Wolf (2007).

This article illustrates that in dealing with partially coherent wave in an imaging system, the coherent component and the incoherent component can be treated separately. The results can then be combined to give the result for partially coherent wave (Yuen (2011w), Sect. 4). There is always incoherent superposition of the coherent component and the incoherent component or their evolutions. Their intensities, mutual coherence functions, mutual intensities, cross-spectral density functions and radiant intensities add. The problem of partially coherent wave in an imaging system reduces to the problems of coherent wave and incoherent wave in the imaging system. For the inverse problem in lensless imaging, the approaches of Yuen (2011v) and this article are complementary to each other. Results obtained in the previous articles about coherent imaging and incoherent imaging can be used. More results and insight are obtained. Experiments about quasi-monochromatic partially coherent light wave which is a combination of a coherent wave and an incoherent wave, in optical systems are needed.

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Nonperiodic object emitting polychromatic wave

7

Imaging of nonperiodic object emitting polychromatic coherent wave.

Direct and inverse problems, lensless imaging, phase-retrieval microscope, phase-retrieval telescope, lenslets microscope and lenslets telescope

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This article deals with the direct and inverse problems of imaging of nonperiodic object emitting polychromatic coherent light wave. Radiant intensity, cross-spectral density function and mutual intensity are dealt with.

Keywords: Phase retrieval; iterative algorithms with oversampling; first Born approximation; mutual coherence function; mutual intensity; cross-spectral density function; spectral degree of coherence; radiant intensity; natural resolution; empirical amplitude transfer function; Sparrow criterion of resolution; Rayleigh criterion of resolution; optical wave, acoustic wave and matter wave imaging, including electromagnetic wave, X-ray, electron, ion and neutron waves.

1. Introduction

This is sequel to Yuen (2011g). This article deals with the direct and inverse problems of imaging of nonperiodic object emitting polychromatic coherent light wave. Radiant intensity, cross-spectral density function and mutual intensity are dealt with. The first Born approximation is not used. For the inverse problem, iterative algorithms with oversampling are applied to the radiant intensity, spectral degree of coherence or mutual intensity in the far field.

Sect. 2.1 presents lensless imaging. In the inverse problem, the object $\hat{f}(\mathbf{x}_o, \omega)$, the intensity and intensity distribution of the object are obtained.

Sect. 2.2 presents imaging via a lens. In the inverse problem using the radiant intensity in the far field of the image, $\hat{f}(\mathbf{x}_o,\varpi)$, the mutual intensity and the intensity of the object are obtained. Natural resolution of the object is dealt with. In the inverse problem using the spectral degree of coherence in the far field of the image, the intensity distribution of the object is obtained. Natural resolution of the object is dealt with. Empirical amplitude transfer function is used. It takes into account aberrations of the lens and other defects of the system. We have by-passed the Sparrow or Rayleigh criterion of resolution (or alike) of the image, and the corresponding angular resolution.

In Sect. 2.3, in the inverse problem for lenslets microscope or lenslets telescope using the radiant intensity of the diffraction rods of the image in all the lenslets, the extended $\hat{g}(\mathbf{x},\varpi)$ is determined. The object $\hat{f}(\mathbf{x}_o,\varpi)$ is determined by means of periodic convolution. Natural resolution of the object is dealt with. In the inverse problem using mutual intensity of the diffraction rods, approximate value of the intensity of the image is obtained, in particular, for sufficiently large unit cell. The Sparrow criterion of resolution of the approximate intensity of the image (or alike) is applied. Natural resolution of the object is not dealt with.

In combination with iterative algorithms and oversampling, powerful methods in structure determination in X-ray crystallography, such as sharpening of Patterson function, direct methods, point-atom model, accelerated convergence of extension of Bragg intensities and structure factors, and sharpening in reciprocal space can also be applied.

The spectral degree of coherence is measured in Arimoto & Ohtsuka (1998).

Imaging of an object emitting polychromatic plane wave by means of correlation-induced spectral changes is presented in Gbur & Wolf (1999).

This article may be applied to acoustic wave and matter wave, including electromagnetic wave, X-ray, electron, ion and neutron waves. For optical wave, acoustic wave and matter wave, including electromagnetic wave, X-ray, electron, ion and neutron waves, use Zarubin (1993), (4.2) for three-dimensional object and (5.9) for planar object.

2. Method

We will use the notation:

For $f(\mathbf{x}_o, t)$, use $\tilde{f}$ for the spatial Fourier transform,

$\hat{f}$ for the temporal Fourier transform,

$\breve{f}$ for the spatial and temporal Fourier transform.

2.1. Lensless imaging. Direct and inverse problems

(A) Direct problem

Consider a real or complex finite-size planar or three-dimensional object $f(\mathbf{x}_o, t)$ emitting a polychromatic coherent wave. As a function of $\mathbf{x}_o$, $f(\mathbf{x}_o, t)$ satisfies the constraints in Yuen (2011a, 2011c or 2011d).

Consider the space-frequency domain. The temporal Fourier transform of $f(\mathbf{x}_o, t)$ is $\hat{f}(\mathbf{x}_o, \omega)$.

The intensity of the object $\hat{I}_o(\mathbf{x}_o, \omega) = \left|\hat{f}(\mathbf{x}_o, \omega)\right|^2$. (1)

The intensity distribution of the object is given by $\int_0^\infty \left|\hat{f}(\mathbf{x}_o, \omega)\right|^2 d\omega$ (2)

The cross-spectral density function of the object

$$\hat{\Gamma}_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}, \omega) = \hat{f}(\mathbf{x}_{o1}, \omega)\hat{f}^*(\mathbf{x}_{o2}, \omega), \quad (3)$$

(Carter & Wolf (1981), (4.1), Mandel & Wolf (1995), (4.5-73)).

The mutual coherence function of the object $\Gamma_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}, \tau)$ is given by the inverse temporal Fourier transform of the cross-spectral density function of the object $\hat{\Gamma}_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}, \omega)$ (Mandel & Wolf (1995), (4.5-77)).

The mutual intensity of the object $J_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}) = \Gamma_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}, 0)$. (4)

The intensity distribution of the object is given by $J_o(\mathbf{x}_o, \mathbf{x}_o)$. (5)

This is equal to that in (2).

The cross-spectral density function in the far field of the object is given in Carter & Wolf (1981), (4.8) for three-dimensional object (or Wolf & Carter (1978), (3.16) for planar object),

$$\hat{\Gamma}(\varsigma_1\mathbf{u}_1, \varsigma_2\mathbf{u}_2, \omega) = (2\pi)^6\, \breve{f}(k\mathbf{u}_1, \omega)\, \breve{f}^*(k\mathbf{u}_2, \omega)\{\exp[ik(\varsigma_1 - \varsigma_2)]\}/(\varsigma_1\varsigma_2) \tag{6}$$

$\mathbf{u}_1$, $\mathbf{u}_2$ are unit vectors.

$\varsigma_1\mathbf{u}_1$ and $\varsigma_2\mathbf{u}_2$ denote points in the far field.

The wave number $k = \omega / c$.

The radiant intensity in the far field of the object

$$\Lambda(\mathbf{u}, \omega) = (2\pi)^6 \left| \breve{f}(k\mathbf{u}, \omega) \right|^2, \tag{7}$$

(Carter & Wolf (1981), (4.9) for three-dimensional object, or Wolf & Carter (1978), (3.17) for planar object). $\mathbf{u}$ is a unit vector.

The radiant intensity and the magnitude of the spectral degree of coherence in the far field are measured (Carter & Wolf (1981), (4.10), (5.8), Mandel & Wolf (1995), pp. 173-176, Wolf & James (1996), Sect. 4.3, Wolf (2007), pp. 68-69).

(B) Inverse problem using the radiant intensity in the far field of the object

According to the chosen resolution, $\omega_{\max}$ is employed. Narrow-band filters are used separately to transmit narrow bands of the spectrum for all ω with $\omega \le \omega_{\max}$ (Wolf & James (1996), Sect. 7.1, 7.2, Wolf (2007), p. 68-69). That is, each time, we deal with one quasi-monochromatic wave. The radiant intensity in (7) is measured for all ω with $\omega \le \omega_{\max}$ and all $\mathbf{u}$. Hence $\left|\breve{f}(k\mathbf{u},\omega)\right|$ is determined. Iterative algorithms with oversampling in Yuen (2011b or 2011e) are applied to obtain the phase of $\breve{f}(k\mathbf{u},\omega)$ from the magnitude. Powerful methods in structure determination in X-ray crystallography, such as sharpening of Patterson function, direct methods, point-atom model, accelerated convergence of extension of Bragg intensities and structure factors, and sharpening in reciprocal space can also be applied (Yuen (2011f)). The solution $\hat{f}(\mathbf{x}_o,\omega)$ is obtained. The intensity of the object is obtained (see (1)). The inverse temporal Fourier transform of $\hat{f}(\mathbf{x}_o,\omega)$ is $f(\mathbf{x}_o,t)$.

(C) Inverse problem using the spectral degree of coherence in the far field of the object

In (6), apart from the factor $(2\pi)^6\left\{\exp[i\bar{k}(\varsigma_1-\varsigma_2)]\right\}/(\varsigma_1\varsigma_2)$, the cross-spectral density function in the far field of the object $\hat{\Gamma}(\varsigma_1\mathbf{u}_1,\varsigma_2\mathbf{u}_2,\omega)$ is the six-dimensional (or four-dimensional for planar object) spatial Fourier transform of $\hat{f}(\mathbf{x}_{o1},\omega)\hat{f}^*(-\mathbf{x}_{o2},\omega)$. The magnitude of the spectral degree of coherence in the far field $\left|\hat{\Gamma}(\varsigma_1\mathbf{u}_1,\varsigma_2\mathbf{u}_2,\omega)\right|$ is measured for $\omega \le \omega_{\max}$ and all $\mathbf{u}_1$ and $\mathbf{u}_2$.

$\hat{f}(\mathbf{x}_{o1},\omega)\hat{f}^*(-\mathbf{x}_{o2},\omega)$ is a six-dimensional complex body (or four-dimensional for planar object) with each of its real and imaginary components satisfying the constraints (Yuen (2011d), (C)):

(i) finite size
(ii) non-negative
(iii) bounded

(iv) sectionally continuous

(v) sharp boundary. The component function $\neq 0$ almost everywhere at the boundary surface of the six-dimensional body (or four-dimensional for planar object)

The real and imaginary components satisfy the constraint

(vi) the size (shape and dimensions), and the regions of zeros (not isolated points) of the real part and the imaginary part are the same.

Iterative algorithms with oversampling in Yuen (2011e) are applied to obtain the phase of $\hat{\Gamma}(\varsigma_1\mathbf{u}_1, \varsigma_2\mathbf{u}_2, \omega)$ from the magnitude. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011f)). The solution $\hat{f}(\mathbf{x}_{o1}, \omega)\hat{f}^*(-\mathbf{x}_{o2}, \omega)$ is obtained.

Replace $-\mathbf{x}_{o2}$ by $\mathbf{x}_{o2}$ in $\hat{f}(\mathbf{x}_{o1}, \omega)\hat{f}^*(-\mathbf{x}_{o2}, \omega)$. The cross-spectral density function of the object $\hat{\Gamma}_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}, \omega) = \hat{f}(\mathbf{x}_{o1}, \omega)\hat{f}^*(\mathbf{x}_{o2}, \omega)$ in (3) is obtained.

For $\mathbf{x}_{o1} = \mathbf{x}_{o2} = \mathbf{x}_o$, $\left|\hat{f}(\mathbf{x}_o, \omega)\right|$ is obtained. This will agree with that obtained from the radiant intensity in (B).

The mutual coherence function of the object $\Gamma_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}, \tau)$ is given by the inverse temporal Fourier transform of the cross-spectral density function of the object $\hat{\Gamma}_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}, \omega)$.

The mutual intensity of the object $J_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}) = \Gamma_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}, 0)$.
The intensity distribution of the object is given by $J_o(\mathbf{x}_o, \mathbf{x}_o)$.
This is equal to that in (2).

Remark: For object emitting only one quasi-monochromatic coherent wave, the measured radiant intensity and the magnitude of the mutual intensity in the far field are not sufficient for the application of phase-retrieval method.

2.2. Image of object in a lens. Direct and inverse problems. Phase-retrieval microscope, phase-retrieval telescope

(A) Direct problem

This is a continuation of Sect. 2.1, Lensless imaging, (A) Direct problem.

A lens is used to achieve superresolution.

Use a generalized transfer function (Beran & Parrent (1964), (7-24), (7-25)):

$$L(\mathbf{K}_1, \mathbf{K}_2, \omega) = \tilde{h}(\mathbf{K}_1, \omega)\tilde{h}^*(-\mathbf{K}_2, \omega) \tag{8}$$

$$\breve{\Gamma}_i(\mathbf{K}_1, \mathbf{K}_2, \omega) = L(\mathbf{K}_1, \mathbf{K}_2, \omega)\breve{\Gamma}_o(\mathbf{K}_1, \mathbf{K}_2, \omega) \tag{9}$$

where $\tilde{h}(\mathbf{K}, \omega)$ is the spatial Fourier transform of the impulse response $h(\mathbf{x}, \omega)$ of the lens. $\tilde{h}(\mathbf{K}, \omega)$ is the amplitude transfer function.

$\breve{\Gamma}_i(\mathbf{K}_1, \mathbf{K}_2, \omega)$ and $\breve{\Gamma}_o(\mathbf{K}_1, \mathbf{K}_2, \omega)$ are the spatial Fourier transforms of the cross-spectral density functions of the image $\hat{\Gamma}_i(\mathbf{x}_1, \mathbf{x}_2, \omega)$ and the object $\hat{\Gamma}_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}, \omega)$, respectively.

$\hat{\Gamma}_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}, \omega)$ is given by (3). Hence from (9), $\breve{\Gamma}_i(\mathbf{K}_1, \mathbf{K}_2, \omega)$ is obtained. $\hat{\Gamma}_i(\mathbf{x}_1, \mathbf{x}_2, \omega)$ is then obtained.

From Beran & Parrent (1964), (7-44), for $\mathbf{x}_1 = \mathbf{x}_2 = \mathbf{x}$, the intensity of the image

$$\hat{I}_i(\mathbf{x}, \omega) = \hat{\Gamma}_i(\mathbf{x}, \mathbf{x}, \omega) \tag{10}$$

The intensity distribution of the image is given by $\int_0^\infty \hat{I}_i(\mathbf{x}, \omega)d\omega$ (11)

The mutual coherence function of the image $\Gamma_i(\mathbf{x}_1, \mathbf{x}_2, \tau)$ is given by the inverse temporal Fourier transform of $\hat{\Gamma}_i(\mathbf{x}_1, \mathbf{x}_2, \omega)$.

The mutual intensity of the image $J_i(\mathbf{x}_1, \mathbf{x}_2) = \Gamma_i(\mathbf{x}_1, \mathbf{x}_2, 0)$. (12)

The intensity distribution of the image is given by $J_i(\mathbf{x}, \mathbf{x})$. (13)

This is equal to that in (11).

The real image

$$\hat{g}(\mathbf{x}, \omega) = \hat{f}(\mathbf{x}, \omega) \otimes h(\mathbf{x}, \omega) \tag{14}$$

The real image is coherent (Beran & Parrent (1964), p. 109, (7-33)).

The intensity of the image $\hat{I}_i(\mathbf{x}, \omega) = |\hat{g}(\mathbf{x}, \omega)|^2$. (15)

which is the same as that in (10).

The intensity distribution of the image is given by $\int_0^\infty |\hat{g}(\mathbf{x}, \omega)|^2 \, d\omega$ (16)

which is the same as that in (11).

The intensity distribution and the magnitude of the mutual intensity of the image can be measured. By means of narrow-band filters transmitting a narrow band of the spectrum, intensity of the image can be measured.

The image acts as a secondary source. From Carter & Wolf (1981), (3.5), Mandel & Wolf (1995), (5.2-25) for three-dimensional object, or Wolf (2007), p.84, (16) for planar object, the cross-spectral density function in the far field of the image,

$$\hat{\Gamma}(\varsigma_1 \mathbf{u}_1, \varsigma_2 \mathbf{u}_2, \omega) = (2\pi)^6 \breve{\Gamma}_i(k\mathbf{u}_1, -k\mathbf{u}_2, \omega) \{\exp[ik(\varsigma_1 - \varsigma_2)]\} / (\varsigma_1 \varsigma_2) \tag{17}$$

From Carter & Wolf (1981), (3.9), Mandel & Wolf (1995), (5.2-15) or (5.2-19), (5.2-22) for three-dimensional object, or Wolf (2007), p.86, (20) for planar object, the radiant intensity in the far field of the image,

$$\Lambda(\mathbf{u}, \omega) = (2\pi)^6 \breve{\Gamma}_i(k\mathbf{u}, -k\mathbf{u}, \omega) \tag{18}$$

For a three-dimensional object, use all orientations of the object. The radiant intensity and the magnitude of the spectral degree of coherence in the far field can be measured.

(B) Inverse problem using the radiant intensity in the far field of the image

According to the chosen resolution, $\omega_{\max}$ is employed. Narrow-band filters are used separately to transmit narrow bands of the spectrum for all ω with $\omega \le \omega_{\max}$.

Consider each quasi-monochromatic wave of mean angular frequency $\overline{\omega}$ transmitted by a filter. The radiant intensity in the far field of the image is used. Use Yuen (2011g), Sect. 2.2 (B). The object $\hat{f}(\mathbf{x}_o, \overline{\omega})$ is obtained. The mutual intensity and the intensity of the object are obtained. Natural resolution of the object is dealt with. Empirical amplitude transfer function $\tilde{h}(\mathbf{K}, \overline{\omega})$ is used. We have by-passed the Sparrow or Rayleigh criterion of resolution (or alike) of the image, and the corresponding angular resolution.

(C) Inverse problem using the spectral degree of coherence in the far field of the image

In (17), apart from the factor $(2\pi)^6\{\exp[ik(\varsigma_1-\varsigma_2)]\}/(\varsigma_1\varsigma_2)$, the cross-spectral density function in the far field of the image $\hat{\Gamma}(\varsigma_1\mathbf{u}_1, \varsigma_2\mathbf{u}_2, \omega)$ is the six-dimensional (or four-dimensional for planar object) spatial Fourier transform of $\hat{\Gamma}_i(\mathbf{x}_1, -\mathbf{x}_2, \omega)$ with the image regarded as a source. The magnitude of the spectral degree of coherence in the far field $\left|\hat{\Gamma}(\varsigma_1\mathbf{u}_1, \varsigma_2\mathbf{u}_2, \omega)\right|$ is measured for $\omega \le \omega_{\max}$ and all $\mathbf{u}_1$ and $\mathbf{u}_2$. $\hat{\Gamma}_i(\mathbf{x}_1, -\mathbf{x}_2, \omega)$ is a six-dimensional complex body (or four-dimensional for planar object) with each of its real and imaginary components satisfying the constraints in Sect. 2.1, (C)). Iterative algorithms with oversampling in

Yuen (2011e) are applied to obtain the phase of $\hat{\Gamma}(\varsigma_1\mathbf{u}_1, \varsigma_2\mathbf{u}_2, \omega)$ from the magnitude. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011f)). The solution $\hat{\Gamma}_i(\mathbf{x}_1, -\mathbf{x}_2, \omega)$ is obtained. Substitute this into (18). The radiant intensity obtained should agree with the experimentally measured values.

Replace $-\mathbf{x}_2$ by $\mathbf{x}_2$ in $\hat{\Gamma}_i(\mathbf{x}_1, -\mathbf{x}_2, \omega)$. The cross-spectral density function of the image $\hat{\Gamma}_i(\mathbf{x}_1, \mathbf{x}_2, \omega)$ is obtained.

The intensity of the image $\hat{I}_i(\mathbf{x}, \omega) = \hat{\Gamma}_i(\mathbf{x}, \mathbf{x}, \omega)$.

This is equal to that in (10).

The intensity distribution of the image is given by $\int_0^\infty \hat{I}_i(\mathbf{x}, \omega) d\omega$.

This is equal to that in (11).

This intensity distribution should agree with the value measured in (A).

The mutual coherence function of the image $\Gamma_i(\mathbf{x}_1, \mathbf{x}_2, \tau)$ is given by the inverse temporal Fourier transform of $\hat{\Gamma}_i(\mathbf{x}_1, \mathbf{x}_2, \omega)$.

The mutual intensity of the image $J_i(\mathbf{x}_1, \mathbf{x}_2) = \Gamma_i(\mathbf{x}_1, \mathbf{x}_2, 0)$. (19)

The intensity distribution of the image is given by $J_i(\mathbf{x}, \mathbf{x})$. (20)

From (9), $\breve{\Gamma}_o(\mathbf{K}_1, \mathbf{K}_2, \omega)$ of the object is obtained. Its inverse spatial Fourier transforms gives the cross-spectral density functions of the object $\hat{\Gamma}_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}, \omega)$.

Its inverse temporal Fourier transform gives the mutual coherence function of the object $\Gamma_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}, \tau)$.

The mutual intensity of the object $J_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}) = \Gamma_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}, 0)$.
The intensity distribution of the object is given by $J_o(\mathbf{x}_o, \mathbf{x}_o)$
This is equal to that in (2).

Natural resolution of the object is dealt with. Empirical amplitude transfer function $\tilde{h}(\mathbf{K}, \omega)$ is used. We have by-passed the Sparrow or Rayleigh criterion of resolution (or alike) of the image, and the corresponding angular resolution.

2.3. Lenslets microscope and lenslets telescope. Direct and inverse problems

According to the chosen resolution, $\omega_{\max}$ is employed. Narrow-band filters are used separately to transmit narrow bands of the spectrum for all ω with $\omega \leq \omega_{\max}$.

Consider each quasi-monochromatic wave of mean angular frequency $\overline{\omega}$ transmitted by a filter. From Wolf & James (1996), (7.8),

$$\mu(P_1, P_2, \overline{\omega}) = \gamma(P_1, P_2, 0) \tag{21}$$

where $\mu(P_1, P_2, \overline{\omega})$ is the spectral degree of coherence at frequency $\overline{\omega}$,
$\gamma(P_1, P_2, 0)$ is the complex degree of coherence for zero time delay.

Hence we can use measurement of the magnitude of the mutual intensity in the far field $|J(\varsigma_1, \varsigma_2)|$.

Two-dimensional array of lenslets can be used to form lenslets microscope or lenslets telescope. The array of lenslets forms a linear system. Its impulse response is formed by adding replica of $h(\mathbf{x}, \overline{\omega})$ for one lenslet successfully in x_1, x_2 to give a periodic function, the extended $h(\mathbf{x}, \overline{\omega})$. The periods are chosen such that the Fourier coefficients of this extended $h(\mathbf{x}, \overline{\omega})$ are nonzero (Yuen (2008), (D)). The focal length of the lenslets, the object distance and the unit cell constants a, b of the array of lenslets are chosen so that the image predicted by geometrical optics in one lenslet, $\hat{f}(\mathbf{x}, \overline{\omega})$, can be contained in one unit cell (Yuen (2008), (D), constraint (i)).

(A) Direct problem

Use Yuen (2011g), Sect. 2.3, (A). The radiant intensity and the magnitude of the mutual intensity $\left|J(\varsigma_1, \varsigma_2)\right|$ of the diffraction rods of the image in all the lenslets are measured.

(B) Inverse problem using the radiant intensity of the diffraction rods of the image in all the lenslets

Use Yuen (2011g), Sect. 2.3, (B), Sect. 2.4, (A). The extended $\hat{g}(\mathbf{x}, \overline{\omega})$ is determined. It is formed by adding replica of $\hat{g}(\mathbf{x}, \overline{\omega})$ in one lenslet successfully in x_1, x_2 to give a periodic function with same unit cell constants as the array of lenslets.

The intensity of this extended $\hat{g}(\mathbf{x}, \overline{\omega})$ = | extended $\hat{g}(\mathbf{x}, \overline{\omega})$ | ^{2}.

The object $\hat{f}(\mathbf{x}_o, \overline{\omega})$ is determined by means of periodic convolution.

Natural resolution of the object is dealt with. The image $\hat{g}(\mathbf{x}, \overline{\omega})$ is not used explicitly. The Sparrow criterion of resolution (or alike) of the image and the corresponding angular resolution are by-passed. The empirical impulse response $h(\mathbf{x}, \overline{\omega})$ contains the aberrations of the lenslets and other defects of the system.

(C) Inverse problem using the mutual intensity of the diffraction rods of the image in all the lenslets

Use Yuen (2011g), Sect. 2.3, (C), Sect. 2.4, (B). The magnitude of the mutual intensity $\left|J(\varsigma_1, \varsigma_2)\right|$ of the diffraction rods is used. Approximate value of the intensity of the image in one lenslet is obtained, in particular, for sufficiently large unit cell. The Sparrow criterion of resolution of the approximate intensity of the image (or alike) and the corresponding angular resolution are applied. Natural resolution of the object is not dealt with.

3. Discussions and conclusion

This article deals with the direct and inverse problems of imaging of object emitting polychromatic coherent light wave. Radiant intensity, cross-spectral density function and mutual intensity are dealt with.

In lensless imaging, the inverse problem uses radiant intensity or spectral degree of coherence in the far field of the object. $\hat{f}(\mathbf{x}_o, \overline{\omega})$, $\left|\hat{f}(\mathbf{x}_o, \omega)\right|$ and intensity distribution of the object is obtained.

In the inverse problem of imaging of object in a lens using the radiant intensity in the far field of the image, the object $\hat{f}(\mathbf{x}_o, \overline{\omega})$ is obtained. The mutual intensity and the intensity of the object are obtained. Natural resolution of the object is dealt with. Empirical amplitude transfer function $\tilde{h}(\mathbf{K}, \overline{\omega})$ is used. We have by-passed the Sparrow or Rayleigh criterion of resolution (or alike) of the image, and the corresponding angular resolution. In the inverse problem using spectral degree of coherence in the far field of the image, intensity and intensity distribution of the image and intensity distribution of the object are obtained. Natural resolution of the object is dealt with.

In the inverse problem for lenslets microscope or lenslets telescope using the radiant intensity of the diffraction rods of the image in all the lenslets, the extended $\hat{g}(\mathbf{x}, \overline{\omega})$ is determined. The object $\hat{f}(\mathbf{x}_o, \overline{\omega})$ is determined by means of periodic convolution. Natural resolution of the object is dealt with. In the inverse problem using mutual intensity of the diffraction rods, approximate value of the intensity of the image in one lenslet is obtained, in particular, for sufficiently large unit cell. The Sparrow criterion of resolution of the approximate intensity of the image (or alike) is applied. Natural resolution of the object is not dealt with.

In combination with iterative algorithms and oversampling, powerful methods in X-ray crystallography can also be applied.

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8

Imaging of nonperiodic object emitting polychromatic incoherent wave.

Direct and inverse problems, lensless imaging, phase-retrieval microscope, phase-retrieval telescope, lenslets microscope and lenslets telescope

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This article deals with the direct and inverse problems of imaging of nonperiodic object emitting polychromatic incoherent light wave. Radiant intensity, cross-spectral density function or mutual intensity is dealt with.

Keywords: Phase retrieval; iterative algorithms with oversampling; first Born approximation; cross-spectral density function; mutual coherence function; mutual intensity; spectral degree of coherence; radiant intensity; Michelson stellar interferometer; natural resolution; empirical amplitude transfer function; empirical optical transfer function; Rayleigh criterion of resolution; optical wave, acoustic wave and matter wave imaging, including electromagnetic wave, X-ray, electron, ion and neutron waves.

1. Introduction

This is sequel to Yuen (2011f). For lensless imaging of nonperiodic object emitting quasi-monochromatic incoherent wave, see Yuen (2011f), Sect. 2.1. This case is the Michelson stellar interferometer for incoherent source.

Correlation-induced spectral changes is presented Mandel & Wolf (1995), Wolf & James (1996), Wolf (2007), Ch. 4, 5, 6.

An important method of determining the angular separation of double stars is presented in James et al. (1995). More results and discussions of imaging of an object by means of this method is presented in Kandpal et al. (1996), Vicalvi et al. (1996), Wolf & James (1996), Sect. 7, Arimoto & Ohtsuka (1998), Santarsiero et al. (1998), Anand & Kandpal (2002), Itoh et al. (2006) and Wolf (2007), p. 66. This is treated in Sect. 2.1 of this article.

This article deals with the direct and inverse problems of imaging of self-illuminating object emitting polychromatic incoherent light wave. Radiant intensity, cross-spectral density function or mutual intensity is dealt with. The first Born approximation is not used. For the inverse problems, iterative algorithms with oversampling are applied to the spectral degree of coherence or mutual intensity.

Sect. 2.1 presents lensless imaging. In the inverse problem, the intensity $\hat{\rho}(\mathbf{x}_o, \omega)$ and intensity distribution of the object are obtained.

Sect. 2.2 presents imaging via a lens. The inverse problem uses the spectral degree of coherence in the far field of the image. The intensity and intensity distribution of the object are obtained. Natural resolution of the object is dealt with. Empirical amplitude transfer function and empirical optical transfer function are used. They take into account aberrations of the lens and other defects of the system. We have by-passed the two-point Rayleigh criterion of resolution (or alike) of the image, and the corresponding angular resolution. These also apply to Sect. 2.3.

In Sect. 2.3, for lenslets microscope or lenslets telescope, the inverse problem uses the mutual intensity of the diffraction rods in the far field of the image in all the lenslets. The intensity and intensity distribution of the object are obtained. Lenslets microscope is useful for determining structure of a cell or molecule, and lenslets telescope for structure of an astronomical object.

In combination with iterative algorithms and oversampling, powerful methods in structure determination in X-ray crystallography, such as sharpening of Patterson function, direct methods, point-atom model, accelerated convergence of extension of Bragg intensities and structure factors, and sharpening in reciprocal space can also be applied.

This article may be applied to acoustic wave and matter wave, including electromagnetic wave, X-ray, electron, ion and neutron waves. For optical wave, acoustic wave and matter wave, including electromagnetic wave, X-ray, electron, ion and neutron waves, use Zarubin (1993), (4.2) for three-dimensional object and (5.9) for planar object.

2.1. Lensless imaging. Direct and inverse problems

(A) Direct problem

Consider a finite-size, non-negative, planar or three-dimensional object $\rho(\mathbf{x}_o, t)$ emitting a polychromatic incoherent wave.

Consider the space-frequency domain. The temporal Fourier transform of $\rho(\mathbf{x}_o, t)$ is $\hat{\rho}(\mathbf{x}_o, \omega)$. As a function of $\mathbf{x}_o$, $\hat{\rho}(\mathbf{x}_o, \omega)$ satisfies the constraints in Yuen (2011a or 2011c). $\hat{\rho}(\mathbf{x}_o, \omega)$ is a non-negative function.

Intensity of the object $= \hat{\rho}(\mathbf{x}_o, \omega)$ (1)

The intensity distribution of the object is given by $\int_0^\infty \hat{\rho}(\mathbf{x}_o, \omega) d\omega$ (2)

From Carter & Wolf (1981), (5.1), the cross-spectral density function of the object

$$\hat{\Gamma}_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}, \omega) = \hat{\rho}(\mathbf{x}_{o1}, \omega)\delta(\mathbf{x}_{o1} - \mathbf{x}_{o2}) \tag{3}$$

The mutual coherence function of the object $\Gamma_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}, \tau)$ is given by the inverse temporal Fourier transform of the cross-spectral density function of the object $\hat{\Gamma}_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}, \omega)$.

The mutual intensity of the object $J_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}) = \Gamma_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}, 0)$. (4)

The intensity distribution of the object is given by $J_o(\mathbf{x}_o, \mathbf{x}_o)$. (5)

This is equal to that in (2).

The cross-spectral density function in the far field of the object is given in Carter & Wolf (1981), (5.6) for three-dimensional object (or Wolf & Carter (1978), (3.12) for planar object),

$$\hat{\Gamma}(\varsigma_1\mathbf{u}_1, \varsigma_2\mathbf{u}_2, \omega) = (2\pi)^3 \breve{\rho}[k(\mathbf{u}_1 - \mathbf{u}_2), \omega]\{\exp[ik(\varsigma_1 - \varsigma_2)]\}/(\varsigma_1\varsigma_2)$$

(6)

$\mathbf{u}_1$, $\mathbf{u}_2$ are unit vectors.

$\varsigma_1\mathbf{u}_1$ and $\varsigma_2\mathbf{u}_2$ denote points in the far field.

The wave number $k = \omega / c$.

The radiant intensity in the far field of the object,

$$\Lambda(\mathbf{u},\omega) = \int \hat{\rho}(\mathbf{x}_o,\omega) d\mathbf{x}_o \,, \tag{7}$$

which is independent of $\mathbf{u}$ (Carter & Wolf (1981), (5.7b) for three-dimensional object, or Wolf & Carter (1978), (3.13) for planar object). $\mathbf{u}$ is a unit vector.

(B) Inverse problem using the spectral degree of coherence in the far field of the object

In (6), apart from the factor $(2\pi)^6 \{\exp[ik(\varsigma_1 - \varsigma_2)]\}/(\varsigma_1\varsigma_2)$, the cross-spectral density function in the far field of the object $\hat{\Gamma}(\varsigma_1\mathbf{u}_1, \varsigma_2\mathbf{u}_2, \omega)$ is the three-dimensional (or two-dimensional for planar object) spatial Fourier transform of $\hat{\rho}(\mathbf{x}_o, \omega)$ (generalization of the van Cittert-Zernike theorem, Wolf (2007), p. 71). According to the chosen resolution, $\omega_{\max}$ is employed. The magnitude of the spectral degree of coherence in the far field $\left|\hat{\Gamma}(\varsigma_1\mathbf{u}_1, \varsigma_2\mathbf{u}_2, \omega)\right|$ is measured for $\omega \le \omega_{\max}$ and all $\mathbf{u}_1 - \mathbf{u}_2$ (Carter & Wolf (1981), (5.8), Mandel & Wolf (1995), pp. 173-176, Wolf & James (1996), Sect. 4.3, Wolf (2007), pp. 68-69). Iterative algorithms with oversampling in Yuen (2011b or 2011d) is applied to obtain the phase of $\hat{\Gamma}(\varsigma_1\mathbf{u}_1, \varsigma_2\mathbf{u}_2, \omega)$ from the magnitude. Powerful methods in structure determination in X-ray crystallography, such as sharpening of Patterson function, direct methods, point-atom model, accelerated convergence of extension of Bragg intensities and structure factors, and sharpening in reciprocal space can also be applied (Yuen (2011e)). The solution $\hat{\rho}(\mathbf{x}_o, \omega)$ is obtained.

The intensity of the object is obtained (see (1)). The inverse temporal Fourier transform of $\hat{\rho}(\mathbf{x}_o, \omega)$ is $\rho(\mathbf{x}_o, t)$.

The intensity distribution is given in (2).

2.2. Image of object in a lens. Phase-retrieval microscope, phase-retrieval telescope. Direct and inverse problems

(A) Direct problem

This is a continuation of Sect. 2.1, Lensless imaging, (A) Direct problem.

A lens is used to achieve superresolution.

The image is partially coherent. Use the generalized transfer function Yuen (2011g), (8), (9):

$$L(\mathbf{K}_1, \mathbf{K}_2, \omega) = \tilde{h}(\mathbf{K}_1, \omega)\tilde{h}^*(-\mathbf{K}_2, \omega) \tag{8}$$

$$\breve{\Gamma}_i(\mathbf{K}_1, \mathbf{K}_2, \omega) = L(\mathbf{K}_1, \mathbf{K}_2, \omega)\breve{\Gamma}_o(\mathbf{K}_1, \mathbf{K}_2, \omega) \tag{9}$$

where $\tilde{h}(\mathbf{K}, \omega)$ is the spatial Fourier transform of the impulse response $h(\mathbf{x}, \omega)$ of the lens. $\tilde{h}(\mathbf{K}, \omega)$ is the amplitude transfer function.

$\breve{\Gamma}_i(\mathbf{K}_1, \mathbf{K}_2, \omega)$ and $\breve{\Gamma}_o(\mathbf{K}_1, \mathbf{K}_2, \omega)$ are the spatial Fourier transforms of the cross-spectral density functions of the image $\hat{\Gamma}_i(\mathbf{x}_1, \mathbf{x}_2, \omega)$ and the object $\hat{\Gamma}_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}, \omega)$, respectively.

$\hat{\Gamma}_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}, \omega)$ is given by (3). Hence from (9), $\breve{\Gamma}_i(\mathbf{K}_1, \mathbf{K}_2, \omega)$ is obtained. $\hat{\Gamma}_i(\mathbf{x}_1, \mathbf{x}_2, \omega)$ is then obtained.

The mutual coherence function of the image $\Gamma_i(\mathbf{x}_1, \mathbf{x}_2, \tau)$ is given by the inverse temporal Fourier transform of $\hat{\Gamma}_i(\mathbf{x}_1,\mathbf{x}_2,\omega)$.

The mutual intensity of the image $J_i(\mathbf{x}_1,\mathbf{x}_2) = \Gamma_i(\mathbf{x}_1,\mathbf{x}_2,0)$. (10)

The intensity distribution of the image is given by $J_i(\mathbf{x},\mathbf{x})$. (11)

From Beran & Parrent (1964), (7-44), (7-45), the intensity of the image,

$$\hat{I}_i(\mathbf{x},\omega) = \hat{\rho}(\mathbf{x},\omega) \otimes \left|h(\mathbf{x},\omega)\right|^2 \tag{12}$$

$$\breve{I}_i(\mathbf{K},\omega) = \breve{\rho}(\mathbf{K},\omega)\Theta(\mathbf{K},\omega) \tag{13}$$

The optical transfer function $\Theta(\mathbf{K},\omega)$ is the spatial Fourier transform of $\left|h(\mathbf{x},\omega)\right|^2$.

The intensity distribution and the magnitude of the mutual intensity of the image are measured.

From (13), the spatial spectral density of the image (Beran & Parrent (1964), (7-48)) is $\tilde{I}_i(\mathbf{K},0) = \int_0^\infty \breve{\rho}(\mathbf{K},\omega)\Theta(\mathbf{K},\omega)d\omega$ (14)

The intensity distribution in the image is given by the spatial inverse transform of $\tilde{I}_i(\mathbf{K},0)$ in (14). (15)

The image acts as a secondary source. From Carter & Wolf (1981), (3.5), Mandel & Wolf (1995), (5.2-25) for three-dimensional object, or Wolf (2007), p.84, (16) for planar object, the cross-spectral density function in the far field of the image,

$$\hat{\Gamma}(\varsigma_1\mathbf{u}_1,\varsigma_2\mathbf{u}_2,\omega) = (2\pi)^6\breve{\Gamma}_i(k\mathbf{u}_1,-k\mathbf{u}_2,\omega)\left\{\exp[ik(\varsigma_1-\varsigma_2)]\right\}/(\varsigma_1\varsigma_2) \tag{16}$$

From Carter & Wolf (1981), (3.9), Mandel & Wolf (1995), (5.2-15) or (5.2-19), (5.2-22) for three-dimensional object, or Wolf (2007), p.86, (20) for planar object, the radiant intensity in the far field of the image,

$$\Lambda(\mathbf{u},\omega)=(2\pi)^6\breve{\Gamma}_i(k\mathbf{u},-k\mathbf{u},\omega) \tag{17}$$

For a three-dimensional object, use all orientations of the object. According to the chosen resolution, $\omega_{\max}$ is employed. The radiant intensity and the magnitude of the spectral degree of coherence in the far field $\left|\hat{\Gamma}(\varsigma_1\mathbf{u}_1,\varsigma_2\mathbf{u}_2,\omega)\right|$ are measured for $\omega\le\omega_{\max}$.

(B) Inverse problem using the spectral degree of coherence in the far field of the image

Use (16) and Yuen (2011g), Sect. 2.2, (C). Iterative algorithms with oversampling in Yuen (2011d) are applied to obtain the phase of $\hat{\Gamma}(\varsigma_1\mathbf{u}_1,\varsigma_2\mathbf{u}_2,\omega)$ from the magnitude. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011e)). The solution $\hat{\Gamma}_i(\mathbf{x}_1,-\mathbf{x}_2,\omega)$ is obtained. Use $\hat{\Gamma}_i(\mathbf{x}_1,-\mathbf{x}_2,\omega)$ in (17). The radiant intensity obtained should agree with the experimentally measured values.

Replace $-\mathbf{x}_2$ by $\mathbf{x}_2$ in $\hat{\Gamma}_i(\mathbf{x}_1,-\mathbf{x}_2,\omega)$. The cross-spectral density function of the image $\hat{\Gamma}_i(\mathbf{x}_1,\mathbf{x}_2,\omega)$ is obtained.

From Beran & Parrent (1964), (7-44), for $\mathbf{x}_1=\mathbf{x}_2=\mathbf{x}$, the intensity of the image

$$\hat{I}_i(\mathbf{x},\omega)=\hat{\Gamma}_i(\mathbf{x},\mathbf{x},\omega)\,. \tag{18}$$

This is equal to that in (12).

The intensity distribution of the image is given by $\int_0^\infty \hat{I}_i(\mathbf{x},\omega)d\omega$ (19)

This intensity distribution should agree with the value measured in (A).

Hence from $\hat{I}_i(\mathbf{x},\omega)$ in (18) and (13), $\hat{\rho}(\mathbf{x},\omega)$ is determined. The object $\hat{\rho}(\mathbf{x}_o,\omega)$ is obtained. This should be equal to the intensity of object $\hat{\rho}(\mathbf{x}_o,\omega)$ in (1).

The mutual coherence function of the image $\Gamma_i(\mathbf{x}_1,\mathbf{x}_2,\tau)$ is given by the inverse temporal Fourier transform of $\hat{\Gamma}_i(\mathbf{x}_1,\mathbf{x}_2,\omega)$.

The mutual intensity of the image $J_i(\mathbf{x}_1,\mathbf{x}_2)=\Gamma_i(\mathbf{x}_1,\mathbf{x}_2,0)$.
The intensity distribution of the image is given by $J_i(\mathbf{x},\mathbf{x})$. This is equal to that in (19).

From (9), $\breve{\Gamma}_o(\mathbf{K}_1,\mathbf{K}_2,\omega)$ of the object is obtained. Its inverse spatial Fourier transforms gives the cross-spectral density functions of the object $\hat{\Gamma}_o(\mathbf{x}_{o1},\mathbf{x}_{o2},\omega)$.

Follow (18), the intensity of the object $\hat{\rho}(\mathbf{x}_o,\omega)=\hat{\Gamma}_o(\mathbf{x}_o,\mathbf{x}_o,\omega)$ (20)

Alternatively, from (3), the intensity of the object,

$$\hat{\rho}(\mathbf{x}_o,\omega)=\int\hat{\Gamma}_o(\mathbf{x}_{o1},\mathbf{x}_o,\omega)d\mathbf{x}_{o1} \tag{21}$$

Inverse temporal Fourier transform of $\hat{\Gamma}_o(\mathbf{x}_{o1},\mathbf{x}_{o2},\omega)$ gives the mutual coherence function of the object $\Gamma_o(\mathbf{x}_{o1},\mathbf{x}_{o2},\tau)$.
The mutual intensity of the object $J_o(\mathbf{x}_{o1},\mathbf{x}_{o2})=\Gamma_o(\mathbf{x}_{o1},\mathbf{x}_{o2},0)$.
The intensity distribution of the object is given by $J_o(\mathbf{x}_o,\mathbf{x}_o)$
This is equal to that in (2).

Natural resolution of the object is dealt with. Empirical amplitude transfer function $\tilde{h}(\mathbf{K},\omega)$ and empirical optical transfer function $\Theta(\mathbf{K},\omega)$ determined in Yuen (2008), Sect. 2.3 are used. We have by-passed the two-point Rayleigh criterion of resolution (or alike) of the image, and the corresponding angular resolution.

2.3. Lenslets microscope and lenslets telescope. Direct and inverse problems

According to the chosen resolution, $\omega_{\max}$ is employed. Narrow-band filters are used to transmit narrow bands of the spectrum for all ω with $\omega \leq \omega_{\max}$ (Wolf & James (1996), Sect. 7.1, 7.2, Wolf (2007), p. 68-69).

Consider each quasi-monochromatic wave of mean angular frequency $\overline{\omega}$ transmitted by a filter. As in Yuen (2011g), Sect. 2.3, we can use measurement of the magnitude of the mutual intensity in the far field $\left|J(\varsigma_1,\varsigma_2)\right|$.

Two-dimensional array of lenslets can be used to form lenslets microscope or lenslets telescope. The array of lenslets forms a linear system. Its impulse response is formed by adding replica of $h(\mathbf{x},\overline{\omega})$ for one lenslet successfully in x_1, x_2 to give a periodic function, the extended $h(\mathbf{x},\overline{\omega})$. The periods are chosen such that the Fourier coefficients of this extended $h(\mathbf{x},\overline{\omega})$ are nonzero (Yuen (2008), (D)). The focal length of the lenslets, the object distance and the unit cell constants a,b of the array of lenslets are chosen so that the image predicted by geometrical optics in one lenslet, $\hat{\rho}(\mathbf{x},\overline{\omega})$, can be contained in one unit cell (Yuen (2008), (D), constraint (i)).

(A) Direct problem

Use Yuen (2011f), Sect. 2.3, (A). The radiant intensity and the magnitude of the mutual intensity $\left|J(\varsigma_1,\varsigma_2)\right|$ of the diffraction rods of the image in all the lenslets are measured

(B) Inverse problem using the mutual intensity of the diffraction rods of the image in all the lenslets

Use Yuen (2011f), Sect. 2.3, (B). The magnitude of the mutual intensity $\left|J(\varsigma_1,\varsigma_2)\right|$ of the diffraction rods is used. The intensity of the object $\hat{\rho}(\mathbf{x}_o,\omega)$ is determined.

The intensity distribution of the object is given by $\int_0^\infty \hat{\rho}(\mathbf{x}_o,\omega)d\omega$.

Natural resolution of the object is dealt with. The extended intensity of the image is not used explicitly. The Rayleigh criterion of resolution (or alike) of the image and the corresponding angular resolution are by-passed. Empirical amplitude transfer function $\tilde{h}(\mathbf{K},\omega)$ and empirical optical transfer function $\Theta(\mathbf{K},\omega)$ are used. The impulse response $h(\mathbf{x},\omega)$ contains the aberrations of the lenslets and other defects of the system.

3. Discussions and conclusion

This article deals with the direct and inverse problems of imaging of object emitting polychromatic incoherent light wave. Radiant intensity, cross-spectral density function or mutual intensity is dealt with.

The inverse problems of lensless imaging and phase-retrieval microscope or phase-retrieval telescope use spectral degree of coherence in the far field of the object or image. The important problem of determining the structure of the object by means of correlation-induced spectral changes is treated. In lensless imaging, intensity $\hat{\rho}(\mathbf{x}_o,\omega)$ and intensity distribution of the object is obtained. In imaging of object in a lens, intensities and intensity distributions of the image and the object are obtained.

In the inverse problem for lenslets microscope or lenslets telescope, the mutual intensity of the diffraction rods of the image in all the lenslets is used. The intensity and intensity distribution of the object are obtained. Natural resolution of the object is dealt with.

In combination with iterative algorithms and oversampling, powerful methods in X-ray crystallography can also be applied.

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9

Imaging of nonperiodic object emitting polychromatic partially coherent wave.

Direct and inverse problems, lensless imaging, phase-retrieval microscope, phase-retrieval telescope, lenslets microscope and lenslets telescope

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This article deals with the direct and inverse problems of imaging of nonperiodic object emitting polychromatic partially coherent light wave. Radiant intensity, cross-spectral density function or mutual intensity is dealt with.

Keywords: Phase retrieval; iterative algorithms with oversampling; first Born approximation; mutual coherence function; cross-spectral density function; spectral degree of coherence; mutual intensity; radiant intensity; natural resolution; empirical amplitude transfer function; Sparrow criterion of resolution; Rayleigh criterion of resolution; optical wave, acoustic wave and matter wave imaging, including electromagnetic wave, X-ray, electron, ion and neutron waves.

1. Introduction

This is sequel to Yuen (2011f). This article deals with the direct and inverse problems of imaging of self-illuminating nonperiodic object emitting polychromatic partially coherent light wave. Radiant intensity, cross-spectral density function or mutual intensity is dealt with. The first Born approximation is not used. For the inverse problems, iterative algorithms with oversampling are applied to the spectral degree of coherence or mutual intensity.

Sect. 2.1 presents lensless imaging. In the inverse problem, the intensity and intensity distribution of the object are obtained.

Sect. 2.2 presents imaging via a lens. The inverse problem uses the spectral degree of coherence in the far field of the image. The intensities and intensity distributions of the image and the object are obtained. The empirical amplitude transfer function $\tilde{h}(\mathbf{K},\omega)$ is used. It takes into account aberrations of the lens and other defects of the system. Natural resolution of the object is dealt with. We have by-passed the Sparrow or Rayleigh criterion of resolution (or alike) of the image, and the corresponding angular resolution.

In Sect. 2.3 for lenslets microscope and lenslets telescope, the inverse problem uses the mutual intensity of the diffraction rods of the image in all the lenslets. The intensity and intensity distribution of the object are obtained. Approximate value of the intensity of the image is obtained. The Sparrow criterion of resolution of the approximate intensity of the image (or alike) and the corresponding angular resolution are applied. Natural resolution of the object is not dealt with. Lenslets microscope is useful for determining structure of a cell or molecule, and lenslets telescope for structure of an astronomical object.

In combination with iterative algorithms and oversampling, powerful methods in structure determination in X-ray crystallography, such as sharpening of Patterson function, direct methods, point-atom model, accelerated convergence of extension of Bragg intensities and structure factors, and sharpening in reciprocal space can also be applied.

This article may be applied to acoustic wave and matter wave, including electromagnetic wave, X-ray, electron, ion and neutron waves. For optical wave, acoustic wave and matter wave, including electromagnetic wave, X-ray, electron, ion and neutron waves, use Zarubin (1993), (4.2) for three-dimensional object and (5.9) for planar object.

2.1. Lensless imaging. Direct and inverse problems

(A) Direct problem

Consider a real or complex finite-size, planar or three-dimensional object $\chi(\mathbf{x}_o,t)$ emitting a polychromatic partially coherent wave. As a function of $\mathbf{x}_o$, $\chi(\mathbf{x}_o,t)$ satisfies the constraints in Yuen (2011a, 2011b or 2011c).

The mutual coherence function of the object

$$\Gamma_o(\mathbf{x}_{o1},\mathbf{x}_{o2},\tau)=\left\langle \chi(\mathbf{x}_{o1},t+\tau),\chi^*(\mathbf{x}_{o2},t)\right\rangle, \qquad (1)$$

Carter & Wolf (1981), (2.4b)).

The mutual intensity of the object $J_o(\mathbf{x}_{o1},\mathbf{x}_{o2})=\Gamma_o(\mathbf{x}_{o1},\mathbf{x}_{o2},0)$ (2)

The intensity distribution of the object is given by $J_o(\mathbf{x}_o,\mathbf{x}_o)$. (3)

Consider the space-frequency domain. The temporal Fourier transform of $\chi(\mathbf{x}_o,t)$ is $\hat{\chi}(\mathbf{x}_o,t)$.

The cross-spectral density function of the object $\hat{\Gamma}_o(\mathbf{x}_{o1},\mathbf{x}_{o2},\omega)$ is given by the temporal Fourier transform of the mutual coherence function of the object. (4)

From Beran & Parrent (1964), (7-44), the intensity of the object

$$\hat{I}_o(\mathbf{x}_o,\omega)=\hat{\Gamma}_o(\mathbf{x}_o,\mathbf{x}_o,\omega) \qquad (5)$$

The intensity distribution of the image is given by $\int_0^\infty \hat{I}_o(\mathbf{x}_o,\omega)d\omega$ (6)

which is equal to that in (3).

From Carter & Wolf (1981), (3.5), Mandel & Wolf (1995), (5.2-25) for three-dimensional object, or Wolf (2007), p.84, (16) for planar object, the cross-spectral density function in the far field of the object,

$$\hat{\Gamma}(\varsigma_1\mathbf{u}_1,\varsigma_2\mathbf{u}_2,\omega)=(2\pi)^6\breve{\Gamma}_o(k\mathbf{u}_1,-k\mathbf{u}_2,\omega)\left\{\exp[ik(\varsigma_1-\varsigma_2)]\right\}/(\varsigma_1\varsigma_2)$$

(7)

From Carter & Wolf (1981), (3.9), Mandel & Wolf (1995), (5.2-15) or (5.2-19), (5.2-22) for three-dimensional object, or Wolf (2007), p.86, (20) for planar object, the radiant intensity in the far field of the object,

$$\Lambda(\mathbf{u},\omega) = (2\pi)^6 \breve{\Gamma}_o(k\mathbf{u},-k\mathbf{u},\omega) \tag{8}$$

For a three-dimensional object, use all orientations of the object. According to the chosen resolution, $\omega_{\max}$ is employed. The radiant intensity and the magnitude of the spectral degree of coherence in the far field $\left|\hat{\Gamma}(\varsigma_1\mathbf{u}_1,\varsigma_2\mathbf{u}_2,\omega)\right|$ are measured for $\omega \le \omega_{\max}$.

(B) Inverse problem using the spectral degree of coherence in the far field of the object

Use (7) and Yuen (2011g), Sect. 2.2, (C). Iterative algorithms with oversampling in Yuen (2011d) are applied to obtain the phase of $\hat{\Gamma}(\varsigma_1\mathbf{u}_1,\varsigma_2\mathbf{u}_2,\omega)$ from the magnitude. Powerful methods in structure determination in X-ray crystallography, such as sharpening of Patterson function, direct methods, point-atom model, accelerated convergence of extension of Bragg intensities and structure factors, and sharpening in reciprocal space can also be applied (Yuen (2011e)). The solution $\hat{\Gamma}_o(\mathbf{x}_{o1},-\mathbf{x}_{o2},\omega)$ is obtained. Substitute this into (8). The radiant intensity obtained should agree with the experimentally measured values.

Replace $-\mathbf{x}_{o2}$ by $\mathbf{x}_{o2}$ in $\hat{\Gamma}_o(\mathbf{x}_{o1},-\mathbf{x}_{o2},\omega)$. The cross-spectral density function of the object $\hat{\Gamma}_o(\mathbf{x}_{o1},\mathbf{x}_{o2},\omega)$ is obtained.

For $\mathbf{x}_{o1} = \mathbf{x}_{o2} = \mathbf{x}_o$, the intensity of the object $\hat{I}_o(\mathbf{x}_o,\omega) = \hat{\Gamma}_o(\mathbf{x}_o,\mathbf{x}_o,\omega)$ is obtained. It should be equal to that in (5).

The intensity distribution of the object is given by $\int_0^\infty \hat{I}_o(\mathbf{x}_o,\omega)d\omega$ which should be equal to that in (6).

The mutual coherence function of the object $\Gamma_o(\mathbf{x}_{o1},\mathbf{x}_{o2},\tau)$ is given by the inverse temporal Fourier transform of the cross-spectral density function of the object $\hat{\Gamma}_o(\mathbf{x}_{o1},\mathbf{x}_{o2},\omega)$. It should be equal to that in (1).

The mutual intensity of the object $J_o(\mathbf{x}_{o1},\mathbf{x}_{o2})=\Gamma_o(\mathbf{x}_{o1},\mathbf{x}_{o2},0)$.

It should be equal to that in (2).

The intensity distribution of the object is given by $J_o(\mathbf{x}_o,\mathbf{x}_o)$ which should be equal to that in (3).

2.2. Image of object in a lens. Phase-retrieval microscope, phase-retrieval telescope. Direct and inverse problems

(A) Direct problem

This is a continuation of Sect. 2.1, Lensless imaging, (A) Direct problem.

A lens is used to achieve superresolution.

The image is partially coherent. Use the generalized transfer function Yuen (2011g), (8), (9):

$$L(\mathbf{K}_1,\mathbf{K}_2,\omega)=\tilde{h}(\mathbf{K}_1,\omega)\tilde{h}^*(-\mathbf{K}_2,\omega) \tag{9}$$

$$\breve{\Gamma}_i(\mathbf{K}_1,\mathbf{K}_2,\omega)=L(\mathbf{K}_1,\mathbf{K}_2,\omega)\breve{\Gamma}_o(\mathbf{K}_1,\mathbf{K}_2,\omega) \tag{10}$$

where $\tilde{h}(\mathbf{K},\omega)$ is the spatial Fourier transform of the impulse response $h(\mathbf{x},\omega)$ of the lens. $\tilde{h}(\mathbf{K},\omega)$ is the amplitude transfer function.

$\breve{\Gamma}_i(\mathbf{K}_1,\mathbf{K}_2,\omega)$ and $\breve{\Gamma}_o(\mathbf{K}_1,\mathbf{K}_2,\omega)$ are the spatial Fourier transforms of the cross-spectral density functions of the image $\hat{\Gamma}_i(\mathbf{x}_1,\mathbf{x}_2,\omega)$

and the object $\hat{\Gamma}_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}, \omega)$, respectively.

$\hat{\Gamma}_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}, \omega)$ is obtained in Sect. 2.1, (A). Hence from (10), $\breve{\Gamma}_i(\mathbf{K}_1, \mathbf{K}_2, \omega)$ is obtained. $\hat{\Gamma}_i(\mathbf{x}_1, \mathbf{x}_2, \omega)$ is then obtained.

The mutual coherence function of the image $\Gamma_i(\mathbf{x}_1, \mathbf{x}_2, \tau)$ is given by the inverse temporal Fourier transform of $\hat{\Gamma}_i(\mathbf{x}_1, \mathbf{x}_2, \omega)$. (11)

The mutual intensity of the image $J_i(\mathbf{x}_1, \mathbf{x}_2) = \Gamma_i(\mathbf{x}_1, \mathbf{x}_2, 0)$. (12)

The intensity distribution of the image is given by $J_i(\mathbf{x}, \mathbf{x})$. (13)

The intensity and the magnitude of the mutual intensity of the image are measured.

The image acts as a secondary source. From Carter & Wolf (1981), (3.5), Mandel & Wolf (1995), (5.2-25) for three-dimensional object, or Wolf (2007), p.84, (16) for planar object, the cross-spectral density function in the far field of the image,

$$\hat{\Gamma}(\varsigma_1\mathbf{u}_1, \varsigma_2\mathbf{u}_2, \omega) = (2\pi)^6 \breve{\Gamma}_i(k\mathbf{u}_1, -k\mathbf{u}_2, \omega)\{\exp[ik(\varsigma_1 - \varsigma_2)]\}/(\varsigma_1\varsigma_2) \tag{14}$$

From Carter & Wolf (1981), (3.9), Mandel & Wolf (1995), (5.2-15) or (5.2-19), (5.2-22) for three-dimensional object, or Wolf (2007), p.86, (20) for planar object, the radiant intensity in the far field of the image,

$$\Lambda(\mathbf{u}, \omega) = (2\pi)^6 \breve{\Gamma}_i(k\mathbf{u}, -k\mathbf{u}, \omega) \tag{15}$$

For a three-dimensional object, use all orientations of the object. According to the chosen resolution, $\omega_{\max}$ is employed. The radiant intensity and the magnitude of the spectral degree of coherence in the far field $\left|\hat{\Gamma}(\varsigma_1\mathbf{u}_1, \varsigma_2\mathbf{u}_2, \omega)\right|$ are measured for $\omega \le \omega_{\max}$.

(B) Inverse problem using the spectral degree of coherence in the far field of the

image

Use (14) and Yuen (2011g), Sect. 2.2, (C). Iterative algorithms with oversampling in Yuen (2011d) are applied to obtain the phase of $\hat{\Gamma}(\varsigma_1\mathbf{u}_1, \varsigma_2\mathbf{u}_2, \omega)$ from the magnitude. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011e)). The solution $\hat{\Gamma}_i(\mathbf{x}_1, -\mathbf{x}_2, \omega)$ is obtained. Use $\hat{\Gamma}_i(\mathbf{x}_1, -\mathbf{x}_2, \omega)$ in (15). The radiant intensity obtained should agree with the experimentally measured values.

Replace $-\mathbf{x}_2$ by $\mathbf{x}_2$ in $\hat{\Gamma}_i(\mathbf{x}_1, -\mathbf{x}_2, \omega)$. The cross-spectral density function of the image $\hat{\Gamma}_i(\mathbf{x}_1, \mathbf{x}_2, \omega)$ is obtained. (16)

The intensity of the image $\hat{I}_i(\mathbf{x}, \omega) = \hat{\Gamma}_i(\mathbf{x}, \mathbf{x}, \omega)$ (17)

The intensity distribution of the image is given by $\int_0^\infty \hat{I}_i(\mathbf{x}, \omega) d\omega$ (18)

This intensity distribution should agree with the value measured in (A).

The mutual coherence function of the image $\Gamma_i(\mathbf{x}_1, \mathbf{x}_2, \tau)$ is given by the inverse temporal Fourier transform of $\hat{\Gamma}_i(\mathbf{x}_1, \mathbf{x}_2, \omega)$.

The mutual intensity of the image $J_i(\mathbf{x}_1, \mathbf{x}_2) = \Gamma_i(\mathbf{x}_1, \mathbf{x}_2, 0)$.
The intensity distribution of the image is given by $J_i(\mathbf{x}, \mathbf{x})$. This is equal to that in (18).

$\breve{\Gamma}_i(\mathbf{K}_1, \mathbf{K}_2, \omega)$ is the spatial Fourier transform of $\hat{\Gamma}_i(\mathbf{x}_1, \mathbf{x}_2, \omega)$. From (10), $\breve{\Gamma}_o(\mathbf{K}_1, \mathbf{K}_2, \omega)$ of the object is obtained. Its inverse spatial Fourier transforms gives the cross-spectral density functions of the object $\hat{\Gamma}_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}, \omega)$.

The intensity of the object $\hat{I}_o(\mathbf{x}_o, \omega) = \hat{\Gamma}_o(\mathbf{x}_o, \mathbf{x}_o, \omega)$

Inverse temporal Fourier transform of $\hat{\Gamma}_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}, \omega)$ gives the mutual coherence function of the object $\Gamma_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}, \tau)$.
The mutual intensity of the object $J_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}) = \Gamma_o(\mathbf{x}_{o1}, \mathbf{x}_{o2}, 0)$.
The intensity distribution of the object is given by $J_o(\mathbf{x}_o, \mathbf{x}_o)$
This is equal to that in (3).

In (9), the empirical amplitude transfer function $\tilde{h}(\mathbf{K}, \omega)$ determined in Yuen (2008), Sect. 2.3 is used. Natural resolution of the object is dealt with. We have by-passed the Sparrow or Rayleigh criterion of resolution (or alike) of the image, and the corresponding angular resolution.

2.3. Lenslets microscope and lenslets telescope. Direct and inverse problems

According to the chosen resolution, $\omega_{\max}$ is employed. Narrow-band filters are used to transmit narrow bands of the spectrum for all ω with $\omega \le \omega_{\max}$ (Wolf & James (1996), Sect. 7.1, 7.2, Wolf (2007), p. 68-69).

Consider each quasi-monochromatic wave of mean angular frequency $\overline{\omega}$ transmitted by a filter. As in Yuen (2011g), Sect. 2.3, we can use measurement of the magnitude of the mutual intensity in the far field $|J(\varsigma_1, \varsigma_2)|$.

Two-dimensional array of lenslets can be used to form lenslets microscope or lenslets telescope. The array of lenslets forms a linear system. The focal length of the lenslets, the object distance and the unit cell constants a, b of the array of lenslets are chosen so that the image predicted by geometrical optics in one lenslet can be contained in one unit cell (Yuen (2008), (D), constraint (i)).

(A) Direct problem

Use Yuen (2011f), Sect. 2.3, (A). The radiant intensity and the magnitude of the mutual intensity $\left|J(\varsigma_1,\varsigma_2)\right|$ of the diffraction rods of the image in all the lenslets are measured

(B) Inverse problem using the mutual intensity of the diffraction rods of the image in all the lenslets

Use Yuen (2011f), Sect. 2.3, (B). The magnitude of the mutual intensity $\left|J(\varsigma_1,\varsigma_2)\right|$ of the diffraction rods is used. Approximate value of the intensity of the image in one lenslet is obtained, in particular, for sufficiently large unit cell. The Sparrow criterion of resolution of the approximate intensity of the image (or alike) and the corresponding angular resolution are applied. Natural resolution of the object is not dealt with.

3. Discussions and conclusion

This article deals with the direct and inverse problems of imaging of object emitting polychromatic partially coherent light wave. Radiant intensity, cross-spectral density function or mutual intensity is dealt with.

In imaging via a lens (Sect. 2.1) and imaging via a lens (Sect. 2.2), the inverse problems use spectral degree of coherence. The important problem of determining the structure of the object by means of correlation-induced spectral changes is treated. In lensless imaging, intensity and intensity distribution of the object is obtained. In imaging of object in a lens, intensities and intensity distributions of the image and the object are obtained. Natural resolution of the object is dealt with.

In Sect. 2.3, for lenslets microscope or lenslets telescope, the inverse problem uses the mutual intensity of the diffraction rods of the image in all the lenslets. Approximate value of the intensity of the image in one lenslet is obtained. The Sparrow criterion of resolution of the approximate intensity of the image (or alike) is applied. Natural resolution of the object is not dealt with.

In combination with iterative algorithms and oversampling, powerful methods in X-ray crystallography can also be applied.

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10

Unique decomposition of polychromatic partially coherent wave in a polychromatic coherent wave and a polychromatic incoherent wave

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A polychromatic partially coherent radiation from a self-illuminating source of size within certain finite limits is a combination of a polychromatic coherent wave and a polychromatic incoherent wave. The decomposition is unique. This applies to sources emitting light wave, electromagnetic wave, acoustic wave, and matter wave (particles). These include chemical-reaction source (including fluorescence, phosphorescence, luminescence) and nuclear-reaction source (including alpha-decay, beta-decay gamma-decay). In a region of size within certain finite limits, every polychromatic partially coherent wave is a combination of a polychromatic coherent wave and a polychromatic incoherent wave.

Keywords: Polychromatic coherent wave; polychromatic incoherent wave; decomposition factor.

1. Introduction

Yuen (2011a), Sect. 2 presents a unique decomposition of a quasi-monochromatic partially coherent wave in coherent and incoherent components. The superposition of the coherent component and the incoherent component or their evolutions, is always incoherent. Their intensities, mutual coherence functions, mutual intensities, cross-spectral density functions and radiant intensities add. In dealing with partially coherent wave in an imaging system, the coherent component and the incoherent component can be treated separately. The results can then be combined to give the result for partially coherent wave. The problem of partially coherent wave in an imaging system reduces to mainly the problems of coherent wave and incoherent wave in the imaging system.

This is illustrated in Yuen (2011b).

This article investigates whether these can be generalized to polychromatic partially coherent wave (Sect. 2).

2. Decomposition of polychromatic partially coherent wave

Consider the space-frequency domain. We start with the hypothesis that every polychromatic partially coherent wave is a combination of quasi-monochromatic partially coherent waves.

Since these quasi-monochromatic partially coherent waves are not correlated (cf. Mandel & Wolf (1995), p. 63), their intensities, mutual coherence functions, mutual intensities, cross-spectral density functions and radiant intensities add.

Yuen (2011a), Sect. 2 shows that in a region of size within certain finite limits, a quasi-monochromatic partially coherent wave is a combination of a coherent wave and an incoherent wave. The decomposition is unique.

Hence every polychromatic partially coherent wave is a combination of quasi-monochromatic coherent waves and quasi-monochromatic incoherent waves. The quasi-monochromatic coherent waves can be grouped together, and the quasi-monochromatic incoherent waves grouped together. The group of quasi-monochromatic coherent waves forms a polychromatic coherent wave. The group of quasi-monochromatic incoherent waves forms a polychromatic incoherent wave. Hence, in a region of size within certain finite limits, every polychromatic partially coherent wave is a combination of a polychromatic coherent wave and a polychromatic incoherent wave. This decomposition of the polychromatic partially coherent wave is unique. This applies to scalar and vector waves (optical wave, electromagnetic wave, acoustic wave and matter wave).

From Yuen (2011a), Sect. 2, in a region of size within certain finite limits, a quasi-monochromatic partially coherent wave of mean angular frequency ω is a combination of $c_{10}(\omega)W_c(\omega)$ and $c_{01}(\omega)W_i(\omega)$. $W_c(\omega)$ and $W_i(\omega)$ are respectively coherent wave and incoherent wave of mean angular frequency ω and of unit quantities. $c_{10}(\omega)$ is real or complex. $c_{01}(\omega)$ is non-negative. Denote the polychromatic coherent wave by $\{c_{10}(\omega)W_c(\omega)\}$, and the polychromatic incoherent wave by $\{c_{01}(\omega)W_i(\omega)\}$.

For the polychromatic partially coherent wave, the spectral density (power spectrum)

$$S(\omega)=\left|c_{10}(\omega)\right|^2+c_{01}(\omega) \tag{1}$$

The total energy of the polychromatic coherent wave

$$E_c=\int_0^\infty \left|c_{10}(\omega)\right|^2 d\omega \tag{2}$$

The total energy of the polychromatic incoherent wave

$$E_i=\int_0^\infty c_{01}(\omega)d\omega \tag{3}$$

The total energy of the polychromatic partially coherent wave

$$E=\int_0^\infty S(\omega)d\omega=E_c+E_i \tag{4}$$

Analogous to the degree of polarization for partially polarized light (Wolf (2007), p. 163, (26)), we can define a decomposition factor α as the ratio of the intensity of the polychromatic coherent wave to the total intensity of the polychromatic partially coherent wave.

$$\alpha=\frac{E_c}{E} \tag{5}$$

From Yuen (2011a), Sect. 3, we have: For a quasi-monochromatic partially coherent wave, the superposition of the coherent component and the incoherent component or their evolutions, is always incoherent. Their intensities, mutual coherence functions, mutual intensities, cross-spectral density functions and radiant intensities add. Hence, the superposition of the polychromatic coherent wave and the polychromatic incoherent wave or their evolutions of a polychromatic partially coherent wave is always incoherent. Their intensities, mutual coherence functions, mutual intensities, cross-spectral density functions and radiant intensities add. In dealing with polychromatic partially coherent wave in an imaging system, the polychromatic coherent wave and the polychromatic incoherent wave can be treated separately. The results can then be combined to give the result for the polychromatic partially coherent wave. The problem of polychromatic partially coherent wave in an imaging system reduces to mainly the problems of polychromatic coherent wave and polychromatic incoherent wave in the imaging system.

An object is represented by two functions. It exhibits a real or complex $\hat{f}(\mathbf{x}_o, \omega)$ under unit quantity of quasi-monochromatic coherent illumination of mean angular frequency ω, and exhibits a non-negative function $\hat{\rho}(\mathbf{x}_o, \omega)$ under unit quantity of quasi-monochromatic incoherent illumination of mean angular frequency ω.

When the object is illuminated by the polychromatic coherent wave $\{c_{10}(\omega) W_c(\omega)\}$, it exhibits $\{c_{10}(\omega)\hat{f}(\mathbf{x}_o, \omega)\}$ (6)

The intensity of the object $\hat{I}_{oco}(\mathbf{x}_o, \omega) = \left|c_{10}(\omega)\hat{f}(\mathbf{x}_o, \omega)\right|^2$. (7)

The intensity distribution of the object is $\int_0^\infty \left|c_{10}(\omega)\hat{f}(\mathbf{x}_o, \omega)\right|^2 d\omega$. (8)

When the object is illuminated by the polychromatic incoherent wave $\{c_{01}(\omega) W_i(\omega)\}$, it exhibits $\{c_{01}(\omega)\hat{\rho}(\mathbf{x}_o, \omega)\}$. (9)

The intensity of the object $\hat{I}_{oin}(\mathbf{x}_o, \omega) = c_{01}(\omega)\hat{\rho}(\mathbf{x}_o, \omega)$ (10)

Intensity distribution of the object is $\int_0^\infty c_{01}(\omega)\hat{\rho}(\mathbf{x}_o, \omega) d\omega$. (11)

When the object is illuminated by a combination of $\{c_{10}(\omega) W_c(\omega)\}$ and $\{c_{01}(\omega) W_i(\omega)\}$, it exhibits $\{c_{10}(\omega)\hat{f}(\mathbf{x}_o, \omega)\}$ and $\{c_{01}(\omega)\hat{\rho}(\mathbf{x}_o, \omega)\}$.

(12)

The intensity of the object

$$\hat{I}_o(\mathbf{x}_o,\omega)=\hat{I}_{oco}(\mathbf{x}_o,\omega)+\hat{I}_{oin}(\mathbf{x}_o,\omega) \tag{13}$$

$$=\left|c_{10}(\omega)\hat{f}(\mathbf{x}_o,\omega)\right|^2+c_{01}(\omega)\hat{\rho}(\mathbf{x}_o,\omega)\,. \tag{14}$$

Intensity distribution of the object is

$$\int_0^\infty\left|c_{10}(\omega)\hat{f}(\mathbf{x}_o,\omega)\right|^2 d\omega+\int_0^\infty c_{01}(\omega)\hat{\rho}(\mathbf{x}_o,\omega)d\omega \tag{15}$$

To determine $c_{10}(\omega)$ and $c_{01}(\omega)$, use an object of known $\hat{f}(\mathbf{x}_o,\omega)$ under unit quantity of quasi-monochromatic coherent illumination of mean angular frequency ω, and known $\hat{\rho}(\mathbf{x}_o,\omega)$ under unit quantity of quasi-monochromatic incoherent illumination of mean angular frequency ω. Use a polychromatic partially coherent radiation as the object illumination. From the intensity of the object (14), a relation of $|c_{10}(\omega)|$ and $c_{01}(\omega)$ is found. Use the same polychromatic partially coherent radiation and a large number of objects with known $\hat{f}(\mathbf{x}_o,\omega)$ and $\hat{\rho}(\mathbf{x}_o,\omega)$. There are many relations and two unknowns. The system is over-determined. If consistent values of $|c_{10}(\omega)|$ and $c_{01}(\omega)$ are obtained, this also serves as an experimental verification for the decomposition of polychromatic partially coherent radiations. A direct verification is to use filters which absorb coherent radiation or incoherent radiation.

Remark: An object illumination is the wave incident on an object. The wave is emitted from a self-illuminating source. (For light wave, see also Wolf (2007), p. 113, ‘the process of scattering on a static linear medium and the process of radiation from a localized source distribution are mathematically equivalent to each other.', and Carter & Wolf (1981), p. 228). Hence this article also applies to source emitting polychromatic partially coherent wave: A polychromatic partially coherent radiation from a self-illuminating source of size within certain finite limits is a combination of a polychromatic coherent wave and a polychromatic incoherent wave. The decomposition is unique. This applies to sources emitting light wave, electromagnetic wave, acoustic wave, and matter wave (particles). These include chemical-reaction source (including fluorescence, phosphorescence, luminescence) and nuclear-reaction source (including alpha-decay, beta-decay gamma-decay).

3. Discussions and conclusion

This article shows that in a region of size within certain finite limits, every polychromatic partially coherent wave is a combination of a polychromatic coherent wave and a polychromatic incoherent wave. This decomposition of the polychromatic partially coherent wave is unique. This applies to source emitting polychromatic partially coherent wave. The superposition of the polychromatic coherent wave and the polychromatic incoherent wave or their evolutions is always incoherent. Their intensities, mutual coherence functions, mutual intensities, cross-spectral density functions and radiant intensities add. In dealing with polychromatic partially coherent wave in an imaging system, the polychromatic coherent wave and the polychromatic incoherent wave can be treated separately. The results can then be combined to give the result for the polychromatic partially coherent wave. The problem of polychromatic partially coherent wave in an imaging system reduces to the problems of polychromatic coherent wave and polychromatic incoherent wave in the imaging system. This will be illustrated in Yuen (2011c).

A decomposition factor α is introduced as the ratio of the intensity of the polychromatic coherent wave to the total intensity of the polychromatic partially coherent wave. α can be compared with the complex degree of coherence $\gamma_{12}(\tau)$ and the spectral degree of coherence $\mu_{12}(\omega)$ (Friberg & Wolf (1995)).

It is important and useful to develop filters for absorbing coherent light and incoherent light. The decomposition of polychromatic partially coherent waves can then be verified directly. It remains to determine the phase of $c_{10}(\omega)$. Since the experimentally observed quantities, intensity, intensity distribution, mutual coherence function, mutual intensity, cross-spectral density function and radiant intensity involve $\left|c_{10}(\omega)\right|^2$, the phase of $c_{10}(\omega)$ is not used.

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11

Imaging of nonperiodic object emitting polychromatic partially coherent wave which is a combination of a polychromatic coherent wave and a polychromatic incoherent wave.

Direct and inverse problems, lensless imaging, phase-retrieval microscope, phase-retrieval telescope, lenslets microscope and lenslets telescope

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This article deals with the direct and inverse problems of imaging of nonperiodic object emitting polychromatic partially coherent light wave which is a combination of a polychromatic coherent wave and a polychromatic incoherent wave. Radiant intensity, cross-spectral density function or mutual intensity is dealt with.

Keywords: Phase retrieval; iterative algorithms with oversampling; first Born approximation; mutual coherence function; mutual intensity; cross-spectral density function; spectral degree of coherence; radiant intensity; natural resolution; empirical impulse response; Sparrow criterion of resolution; Rayleigh criterion of resolution; optical wave, acoustic wave and matter wave imaging, including electromagnetic wave, X-ray, electron, ion and neutron waves.

1. Introduction

This article is sequel to Yuen (2011h, 2011i, 2011j, 2011k, 2011m). The combination of a combination of a polychromatic coherent wave and a polychromatic incoherent wave is a polychromatic partially coherent wave. This article deals with the direct and inverse problems of imaging of nonperiodic object emitting a polychromatic partially coherent light wave in terms of the coherent and incoherent components. Radiant intensity, cross-spectral density function or mutual intensity is dealt with. The first Born approximation is not used. For the inverse problems, iterative algorithms with oversampling are applied to the spectral degree of coherence or mutual intensity. In fact, Yuen (2011m) shows: In a region of size within certain finite limits, every polychromatic partially coherent wave is a combination of a polychromatic coherent wave and a polychromatic incoherent wave. This decomposition of the polychromatic partially coherent wave is unique.

Sect. 2.1 presents lensless imaging. In the inverse problem, $c_{10}(\omega)\hat{f}(\mathbf{x}_o,\omega)$, $c_{01}(\omega)\hat{\rho}(\mathbf{x}_o,\omega)$, intensity and intensity distribution of the object are determined.

Sect. 2.2 presents imaging via a lens. The inverse problem uses the spectral degree of coherence in the far field of the image. Using a numerical method, approximate $\left|\hat{f}(\mathbf{x}_o,\omega)\right|$, $\hat{\rho}(\mathbf{x}_o,\omega)$ and intensity of the object can be determined. Natural resolution of the object is dealt with. Empirical impulse response $h(\mathbf{x},\omega)$ will be used. They take into account aberrations of the lens and other defects of the system. We have by-passed the Sparrow or Rayleigh criterion of resolution (or alike) of the image, and the corresponding angular resolution.

In Sect. 2.3 for lenslets microscope and lenslets telescope, the inverse problem uses the mutual intensity of the diffraction rods in the far field of the image in all the lenslets. Approximate intensities of the object and image for the partially coherent wave are obtained. Natural resolution of the object is dealt with. Empirical impulse response $h(\mathbf{x},\omega)$ is used. Lenslets microscope is useful for determining structure of a cell or molecule, and lenslets telescope for structure of an astronomical object.

In combination with iterative algorithms and oversampling, powerful methods in structure determination in X-ray crystallography, such as sharpening of Patterson function, direct methods, point-atom model, accelerated convergence of extension of Bragg intensities and structure factors, and sharpening in reciprocal space can also be applied.

This article illustrates that, in dealing with polychromatic partially coherent wave in an imaging system, the polychromatic coherent wave and the polychromatic incoherent wave can be treated separately. The results can then be combined to give the result for the polychromatic partially coherent wave. In the direct problem, the problem of polychromatic partially coherent wave in an imaging system reduces to the problems of polychromatic coherent wave and polychromatic incoherent wave in the imaging system. For the inverse problem in lensless imaging, information from Yuen (2011k) is used.

This article may be applied to acoustic wave and matter wave, including electromagnetic wave, X-ray, electron, ion and neutron waves. For optical wave, acoustic wave and matter wave, including electromagnetic wave, X-ray, electron, ion and neutron waves, use Zarubin (1993), (4.2) for three-dimensional object and (5.9) for planar object.

2.1. Lensless imaging. Direct and inverse problems

(A) Direct problem

Consider the space-frequency domain. Consider a finite-size, planar or three-dimensional object $\left\{c_{10}(\omega)\hat{f}(\mathbf{x}_o,\omega)\right\}$ emitting a polychromatic coherent wave $\left\{c_{10}(\omega)W_c(\omega)\right\}$, and $\left\{c_{01}(\omega)\hat{\rho}(\mathbf{x}_o,\omega)\right\}$ emitting a polychromatic incoherent wave $\left\{c_{01}(\omega)W_i(\omega)\right\}$ (Yuen (2011m), Sect. 2). $W_c(\omega)$ and $W_i(\omega)$ are respectively coherent wave and incoherent wave of mean angular frequency ω and of unit quantities. $c_{10}(\omega)$ is real or complex. $c_{01}(\omega)$ is non-negative. The resultant wave emitted is a polychromatic partially coherent scalar wave. As a function of $\mathbf{x}_o$, $\hat{f}(\mathbf{x}_o,\omega)$ satisfies the constraints in Yuen (2011a, 2011c or 2011d) and $\hat{\rho}(\mathbf{x}_o,\omega)$ satisfies the constraints in Yuen (2011a or 2011c). $\hat{\rho}(\mathbf{x}_o,\omega)$ is a non-negative function.

Yuen (2011m) shows: In a region of size within certain finite limits, every polychromatic partially coherent wave is a combination of a polychromatic coherent wave and a polychromatic incoherent wave. This decomposition of the polychromatic partially coherent wave is unique. The superposition of the polychromatic coherent wave and the polychromatic incoherent wave or their evolutions is always incoherent. Their intensities, mutual coherence functions, mutual intensities, cross-spectral density functions and radiant intensities add.

Assume that $|c_{10}(\omega)|$ and $c_{01}(\omega)$ have been determined (Yuen (2011m), Sect. 2).

For the polychromatic coherent wave $\{c_{10}(\omega)W_c(\omega)\}$, from Yuen (2011m), (6), the object exhibits $\{c_{10}(\omega)\hat{f}(\mathbf{x}_o,\omega)\}$. (1)

From Yuen (2011m), (7), the intensity of the object

$$\hat{I}_{oco}(\mathbf{x}_o,\omega)=\left|c_{10}(\omega)\hat{f}(\mathbf{x}_o,\omega)\right|^2. \tag{2}$$

From Yuen (2011m), (8), the intensity distribution of the object is

$$\int_0^\infty \left|c_{10}(\omega)\hat{f}(\mathbf{x}_o,\omega)\right|^2 d\omega \tag{3}$$

From Yuen (2011i), (3), the cross-spectral density function of the object

$$\hat{\Gamma}_{oco}(\mathbf{x}_{o1},\mathbf{x}_{o2},\omega)=|c_{10}(\omega)|^2\,\hat{f}(\mathbf{x}_{o1},\omega)\hat{f}^*(\mathbf{x}_{o2},\omega). \tag{4}$$

The mutual coherence function of the object $\Gamma_{oco}(\mathbf{x}_{o1},\mathbf{x}_{o2},\tau)$ is given by the inverse temporal Fourier transform of the cross-spectral density function of the object

$$\hat{\Gamma}_{oco}(\mathbf{x}_{o1},\mathbf{x}_{o2},\omega). \tag{5}$$

The mutual intensity of the object $J_{oco}(\mathbf{x}_{o1},\mathbf{x}_{o2})=\Gamma_{oco}(\mathbf{x}_{o1},\mathbf{x}_{o2},0)$. (6)

The intensity distribution of the object is given by $J_{oco}(\mathbf{x}_o,\mathbf{x}_o)$. (7)

This is equal to that in (3).

The cross-spectral density function in the far field of the object is given by Yuen (2011i), (6),

$$\hat{\Gamma}_{co}(\varsigma_1\mathbf{u}_1,\varsigma_2\mathbf{u}_2,\omega)$$

$$=(2\pi)^6|c_{10}(\omega)|^2\,\breve{f}(k\mathbf{u}_1,\omega)\breve{f}^*(k\mathbf{u}_2,\omega)\{\exp[ik(\varsigma_1-\varsigma_2)]\}/(\varsigma_1\varsigma_2) \tag{8}$$

$\mathbf{u}_1$, $\mathbf{u}_2$ are unit vectors.

$\varsigma_1\mathbf{u}_1$ and $\varsigma_2\mathbf{u}_2$ denote points in the far field.

The wave number $k=\omega/c$.

The radiant intensity in the far field of the object is given by Yuen (2011i), (7),

$$\Lambda_{co}(\mathbf{u},\omega)=(2\pi)^6\left|c_{10}(\omega)\,\breve{f}(k\mathbf{u},\omega)\right|^2, \tag{9}$$

$\mathbf{u}$ is a unit vector.

For the polychromatic incoherent wave $\{c_{01}(\omega)W_i(\omega)\}$, from Yuen (2011m), (9), the object exhibits $\{c_{01}(\omega)\hat{\rho}(\mathbf{x}_o,\omega)\}$. (10)

From Yuen (2011m), (10), the intensity of the object

$$\hat{I}_{oin}(\mathbf{x}_o,\omega)=c_{01}(\omega)\hat{\rho}(\mathbf{x}_o,\omega) \tag{11}$$

From Yuen (2011m), (11), intensity distribution of the object is

$$\int_0^\infty c_{01}(\omega)\hat{\rho}(\mathbf{x}_o,\omega)d\omega. \tag{12}$$

From Yuen (2011j), (3), the cross-spectral density function of the object

$$\hat{\Gamma}_{oin}(\mathbf{x}_{o1},\mathbf{x}_{o2},\omega)=c_{01}(\omega)\hat{\rho}(\mathbf{x}_{o1},\omega)\delta(\mathbf{x}_{o1}-\mathbf{x}_{o2}) \tag{13}$$

The mutual coherence function of the object $\Gamma_{oin}(\mathbf{x}_{o1},\mathbf{x}_{o2},\tau)$ is given by the inverse temporal Fourier transform of the cross-spectral density function of the object

$$\hat{\Gamma}_{oin}(\mathbf{x}_{o1},\mathbf{x}_{o2},\omega). \tag{14}$$

The mutual intensity of the object $J_{oin}(\mathbf{x}_{o1},\mathbf{x}_{o2})=\Gamma_{oin}(\mathbf{x}_{o1},\mathbf{x}_{o2},0)$. (15)

The intensity distribution of the object is given by $J_{oin}(\mathbf{x}_o,\mathbf{x}_o)$. (16)

This is equal to that in (12).

The cross-spectral density function in the far field of the object is given by Yuen (2011j), (6),

$$\hat{\Gamma}_{in}(\varsigma_1\mathbf{u}_1,\varsigma_2\mathbf{u}_2,\omega)$$

$$=(2\pi)^3 c_{01}(\omega)\breve{\rho}[k(\mathbf{u}_1-\mathbf{u}_2),\omega]\{\exp[ik(\varsigma_1-\varsigma_2)]\}/(\varsigma_1\varsigma_2) \tag{17}$$

The radiant intensity in the far field of the object is given by Yuen (2011j), (7),

$$\Lambda_{in}(\mathbf{u},\omega)=\int c_{01}(\omega)\hat{\rho}(\mathbf{x}_o,\omega)d\mathbf{x}_o \tag{18}$$

which is independent of $\mathbf{u}$

For the polychromatic partially coherent wave $\{c_{10}(\omega)W_c(\omega)\}$ and $\{c_{01}(\omega)W_i(\omega)\}$, the object exhibits $\{c_{10}(\omega)\hat{f}(\mathbf{x}_o,\omega)\}$ and $\{c_{01}(\omega)\hat{\rho}(\mathbf{x}_o,\omega)\}$.

(19)

The intensity of the object

$$\hat{I}_o(\mathbf{x}_o,\omega)=\hat{I}_{oco}(\mathbf{x}_o,\omega)+\hat{I}_{oin}(\mathbf{x}_o,\omega)$$

$$=\left|c_{10}(\omega)\hat{f}(\mathbf{x}_o,\omega)\right|^2+c_{01}(\omega)\hat{\rho}(\mathbf{x}_o,\omega) \tag{20}$$

(use (2) and (11)).

The intensity distribution of the object is given by

$$\int_0^\infty\left|c_{10}(\omega)\hat{f}(\mathbf{x}_o,\omega)\right|^2 d\omega+\int_0^\infty c_{01}(\omega)\hat{\rho}(\mathbf{x}_o,\omega)d\omega \tag{21}$$

(use (3) and (12)).

The cross-spectral density function of the object

$$\hat{\Gamma}_o(\mathbf{x}_{o1},\mathbf{x}_{o2},\omega)=\hat{\Gamma}_{oco}(\mathbf{x}_{o1},\mathbf{x}_{o2},\omega)+\hat{\Gamma}_{oin}(\mathbf{x}_{o1},\mathbf{x}_{o2},\omega)$$

$$=\left|c_{10}(\omega)\right|^2\hat{f}(\mathbf{x}_{o1},\omega)\hat{f}^*(\mathbf{x}_{o2},\omega)+c_{01}(\omega)\hat{\rho}(\mathbf{x}_{o1},\omega)\delta(\mathbf{x}_{o1}-\mathbf{x}_{o2}) \tag{22}$$

(use (4) and (13)).

The mutual coherence function of the object $\Gamma_o(\mathbf{x}_{o1},\mathbf{x}_{o2},\tau)$ is given by the inverse temporal Fourier transform of the cross-spectral density function of the object

$$\hat{\Gamma}_o(\mathbf{x}_{o1},\mathbf{x}_{o2},\omega)\,. \tag{23}$$

The total mutual intensity of the object

$$J_o(\mathbf{x}_{o1},\mathbf{x}_{o2}) = J_{oco}(\mathbf{x}_{o1},\mathbf{x}_{o2}) + J_{oin}(\mathbf{x}_{o1},\mathbf{x}_{o2})\,, \tag{24}$$

(use (6) and (15)).

The intensity distribution of the object is given by $J_{oco}(\mathbf{x}_o,\mathbf{x}_o) + J_{oin}(\mathbf{x}_o,\mathbf{x}_o)$

(25)

(use (7) and (16)). This is equal to that in (21).

The total cross-spectral density function in the far field of the object

$$\hat{\Gamma}(\varsigma\mathbf{u}_1,\varsigma\mathbf{u}_2,\omega) = \hat{\Gamma}_{co}(\varsigma_1\mathbf{u}_1,\varsigma_2\mathbf{u}_2,\omega) + \hat{\Gamma}_{in}(\varsigma_1\mathbf{u}_1,\varsigma_2\mathbf{u}_2,\omega)\,,$$

$$= (2\pi)^6\left|c_{10}(\omega)\right|^2 \breve{f}(k\mathbf{u}_1,\omega)\,\breve{f}^*(k\mathbf{u}_2,\omega)\{\exp[ik(\varsigma_1-\varsigma_2)]\}/(\varsigma_1\varsigma_2)$$

$$+ (2\pi)^3 c_{01}(\omega)\breve{\rho}[k(\mathbf{u}_1-\mathbf{u}_2),\omega]\{\exp[ik(\varsigma_1-\varsigma_2)]\}/(\varsigma_1\varsigma_2) \tag{26}$$

(use (8) and (17)).

The total radiant intensity in the far field of the object

$$\Lambda(\mathbf{u},\omega) = \Lambda_{co}(\mathbf{u},\omega) + \Lambda_{in}(\mathbf{u},\omega)$$

$$= (2\pi)^6\left|c_{10}(\omega)\,\breve{f}(k\mathbf{u},\omega)\right|^2 + \int c_{01}(\omega)\hat{\rho}(\mathbf{x}_o,\omega)d\mathbf{x}_o \tag{27}$$

(use (9) and (18)).

For a three-dimensional object, use all orientations of the object. According to the chosen resolution, $\omega_{\max}$ is employed. The radiant intensity and the magnitude of the spectral degree of coherence in the far field $\left|\hat{\Gamma}(\varsigma_1\mathbf{u}_1,\varsigma_2\mathbf{u}_2,\omega)\right|$ are measured for $\omega \le \omega_{\max}$.

(B) Inverse problem using the radiant intensity in the far field of the object

Assume that $|c_{10}(\omega)|$ and $c_{01}(\omega)$ have not been determined. In (27), the minimum value of $\Lambda(\mathbf{u},\omega)$,

$$\Lambda(\mathbf{u},\omega)_{\min} = \Lambda_{in}(\mathbf{u},\omega) = \int c_{01}(\omega)\hat{\rho}(\mathbf{x}_o,\omega)d\mathbf{x}_o \tag{28}$$

From (27) and (28), $\Lambda_{co}(\mathbf{u},\omega)$ is determined.

From (9), $\left|c_{10}(\omega)\breve{f}(k\mathbf{u},\omega)\right|$ is obtained.

As in Yuen (2011i), Sect. 2.1, (B), iterative algorithms with oversampling in Yuen (2011b or 2011e) are applied to obtain the phase of $c_{10}(\omega)\breve{f}(k\mathbf{u},\omega)$ from the magnitude. Powerful methods in structure determination in X-ray crystallography, such as sharpening of Patterson function, direct methods, point-atom model, accelerated convergence of extension of Bragg intensities and structure factors, and sharpening in reciprocal space can also be applied (Yuen (2011f)). The solution $c_{10}(\omega)\hat{f}(\mathbf{x}_o,\omega)$ is obtained. The intensity, the intensity distribution, the cross-spectral density function, the mutual coherence function and the mutual intensity of the object, and the cross-spectral density function in the far field $\hat{\Gamma}_{co}(\varsigma_1\mathbf{u}_1,\varsigma_2\mathbf{u}_2,\omega)$ for the polychromatic coherent wave are obtained (see (2), (3), (4), (5), (6), (8)).

(C) Inverse problem using the spectral degree of coherence in the far field of the object

Assume that $|c_{10}(\omega)|$ and $c_{01}(\omega)$ have not been determined. In (26), $\hat{\Gamma}(\varsigma\mathbf{u}_1,\varsigma\mathbf{u}_2,\omega)$, is the sum of a six-dimensional spatial Fourier transform and a three-dimensional spatial Fourier transform. Hence we cannot, based on (26), use our phase-retrieval method to obtain the phase of the cross-spectral density function from the magnitude. However, according to Yuen (2011k), Sect. 2.1, (B), a phase-retrieval method can, in fact, be applied to obtain the phase of the cross-spectral density function in the far field of the object.

As in Yuen (2011i), Sect. 2.2, (C), iterative algorithms with oversampling in Yuen (2011b or 2011e) is applied to obtain the phase of $\hat{\Gamma}(\varsigma_1\mathbf{u}_1,\varsigma_2\mathbf{u}_2,\omega)$ from the magnitude. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011f)). Since the cross-spectral density function in the far field $\hat{\Gamma}_{co}(\varsigma_1\mathbf{u}_1,\varsigma_2\mathbf{u}_2,\omega)$ is obtained in (B), $\hat{\Gamma}_{in}(\varsigma_1\mathbf{u}_1,\varsigma_2\mathbf{u}_2,\omega)$ is determined from (26). $c_{01}(\omega)\hat{\rho}(\mathbf{x}_o,\omega)$ is then obtained (Yuen (2011j), Sect. 2.1, (B)). Substitute this into (28), the result obtained should agree with the experimental value of $\Lambda(\mathbf{u},\omega)_{\min}$.

The total intensity of the object is given by (20). It should be the same as that in Yuen (2011k), (5).

The intensity distribution of the object is given by (21). It should be the same as that in Yuen (2011k), (6).

The cross-spectral density function of the object is given by (22). It should be the same as that in Yuen (2011k), (4).

The total mutual intensity of the object is given by (24). It should be the same as that in Yuen (2011k), (2).

As in Iizuka (2008), (10.22), and Goodman (2005), (6-14),

$$\left|\hat{f}(\mathbf{x}_o,\omega)\right|^2 = \hat{\rho}(\mathbf{x}_o,\omega)\,. \tag{29}$$

$\hat{f}(\mathbf{x}_o,\omega)$ and $\hat{\rho}(\mathbf{x}_o,\omega)$ obtained should satisfy this relation.

2.2. Image of object in a lens, phase-retrieval microscope, phase-retrieval telescope. Direct and inverse problems

(A) Direct problem

This is a continuation of Sect. 2.1, Lensless imaging, (A) Direct problem.

Assume that $|c_{10}(\omega)|$ and $c_{01}(\omega)$ have been determined. A lens is used to achieve superresolution. Use a generalized transfer function (Beran & Parrent (1964), (7-24), (7-25)):

$$L(\mathbf{K}_1,\mathbf{K}_2,\omega)=\tilde{h}(\mathbf{K}_1,\omega)\tilde{h}^*(-\mathbf{K}_2,\omega) \tag{30}$$

$$\breve{\Gamma}_i(\mathbf{K}_1,\mathbf{K}_2,\omega)=L(\mathbf{K}_1,\mathbf{K}_2,\omega)\breve{\Gamma}_o(\mathbf{K}_1,\mathbf{K}_2,\omega) \tag{31}$$

where $\tilde{h}(\mathbf{K},\omega)$ is the spatial Fourier transform of the impulse response $h(\mathbf{x},\omega)$ of the lens. $\tilde{h}(\mathbf{K},\omega)$ is the amplitude transfer function.

$\breve{\Gamma}_i(\mathbf{K}_1,\mathbf{K}_2,\omega)$ and $\breve{\Gamma}_o(\mathbf{K}_1,\mathbf{K}_2,\omega)$ are the spatial Fourier transforms of the cross-spectral density functions of the image $\hat{\Gamma}_i(\mathbf{x}_1,\mathbf{x}_2,\omega)$ and the object $\hat{\Gamma}_o(\mathbf{x}_{o1},\mathbf{x}_{o2},\omega)$, respectively.

Consider the polychromatic coherent wave $\{c_{10}(\omega)W_c(\omega)\}$. $\hat{\Gamma}_{oco}(\mathbf{x}_{o1},\mathbf{x}_{o2},\omega)$ is given by (4). Hence from (31), $\breve{\Gamma}_{ico}(\mathbf{K}_1,\mathbf{K}_2,\omega)$ of the image is obtained. $\hat{\Gamma}_{ico}(\mathbf{x}_1,\mathbf{x}_2,\omega)$ of the image is then obtained.

The intensity of the image $\hat{I}_{ico}(\mathbf{x},\omega)=\hat{\Gamma}_{ico}(\mathbf{x},\mathbf{x},\omega)$ (32)

The intensity distribution of the image is given by $\int_0^\infty \hat{I}_{ico}(\mathbf{x},\omega)d\omega$ (33)

The mutual coherence function of the image $\Gamma_{ico}(\mathbf{x}_1,\mathbf{x}_2,\tau)$ is given by the inverse temporal Fourier transform of $\hat{\Gamma}_{ico}(\mathbf{x}_1,\mathbf{x}_2,\omega)$. (34)

The mutual intensity of the image $J_{ico}(\mathbf{x}_1,\mathbf{x}_2)=\Gamma_{ico}(\mathbf{x}_1,\mathbf{x}_2,0)$. (35)

The intensity distribution of the image is given by $J_{ico}(\mathbf{x},\mathbf{x})$. (36)

The image acts as a secondary source. The cross-spectral density function in the far field of the image is given by Yuen (2011i), (17),

$$\hat{\Gamma}_{co}(\varsigma_1\mathbf{u}_1,\varsigma_2\mathbf{u}_2,\omega)=(2\pi)^6\breve{\Gamma}_{ico}(k\mathbf{u}_1,-k\mathbf{u}_2,\omega)\{\exp[ik(\varsigma_1-\varsigma_2)]\}/(\varsigma_1\varsigma_2) \tag{37}$$

The radiant intensity in the far field of the image is given by Yuen (2011i), (18),

$$\Lambda_{co}(\mathbf{u},\omega)=(2\pi)^6\breve{\Gamma}_{ico}(k\mathbf{u},-k\mathbf{u},\omega) \tag{38}$$

Consider the polychromatic incoherent wave $\{c_{01}(\omega)W_i(\omega)\}$. $\hat{\Gamma}_{oin}(\mathbf{x}_{o1},\mathbf{x}_{o2},\omega)$ is given by (13). Hence from (31), $\breve{\Gamma}_{iin}(\mathbf{K}_1,\mathbf{K}_2,\omega)$ of the image is obtained. $\hat{\Gamma}_{iin}(\mathbf{x}_1,\mathbf{x}_2,\omega)$ of the image is then obtained.

The mutual coherence function of the image $\Gamma_{iin}(\mathbf{x}_1,\mathbf{x}_2,\tau)$ is given by the inverse temporal Fourier transform of $\hat{\Gamma}_{iin}(\mathbf{x}_1,\mathbf{x}_2,\omega)$. (39)

The mutual intensity of the image $J_{iin}(\mathbf{x}_1,\mathbf{x}_2)=\Gamma_{iin}(\mathbf{x}_1,\mathbf{x}_2,0)$. (40)

The intensity distribution of the image is given by $J_{iin}(\mathbf{x},\mathbf{x})$. (41)

The intensity of the image,

$$\hat{I}_{iin}(\mathbf{x},\omega)=c_{01}(\omega)\hat{\rho}(\mathbf{x},\omega)\otimes|h(\mathbf{x},\omega)|^2\ , \tag{42}$$

(Yuen (2011j), (12)).

The image acts as a secondary source. The cross-spectral density function in the far field of the image,

$$\hat{\Gamma}_{in}(\varsigma_1\mathbf{u}_1,\varsigma_2\mathbf{u}_2,\omega)=(2\pi)^6\breve{\Gamma}_{iin}(k\mathbf{u}_1,-k\mathbf{u}_2,\omega)\{\exp[ik(\varsigma_1-\varsigma_2)]\}/(\varsigma_1\varsigma_2), \tag{43}$$

(Yuen (2011j), (16))

The radiant intensity in the far field of the image,

$$\Lambda_{in}(\mathbf{u},\omega)=(2\pi)^6\breve{\Gamma}_{iin}(k\mathbf{u},-k\mathbf{u},\omega), \tag{44}$$

(Yuen (2011j), (17)).

Consider the polychromatic partially coherent wave $\{c_{10}(\omega)W_c(\omega)\}$ and $\{c_{01}(\omega)W_i(\omega)\}$. The intensity of the image

$$\hat{I}_i(\mathbf{x},\omega)=\hat{I}_{ico}(\mathbf{x},\omega)+\hat{I}_{iin}(\mathbf{x},\omega) \tag{45}$$

(use (32) and (42)).

Use (33) and (42). The intensity distribution of the image is given by

$$\int_0^\infty \hat{I}_{ico}(\mathbf{x},\omega)d\omega+\int_0^\infty \hat{I}_{iin}(\mathbf{x},\omega)d\omega \tag{46}$$

The total cross-spectral density function of the image

$$\hat{\Gamma}_i(\mathbf{x}_1,\mathbf{x}_2,\omega)=\hat{\Gamma}_{ico}(\mathbf{x}_1,\mathbf{x}_2,\omega)+\hat{\Gamma}_{iin}(\mathbf{x}_1,\mathbf{x}_2,\omega). \tag{47}$$

The mutual coherence function of the image

$$\Gamma_i(\mathbf{x}_1,\mathbf{x}_2,\tau)=\Gamma_{ico}(\mathbf{x}_1,\mathbf{x}_2,\tau)+\Gamma_{iin}(\mathbf{x}_1,\mathbf{x}_2,\tau) \tag{48}$$

(use (34) and (39)).

From (35) and (40), the mutual intensity of the image

$$\begin{aligned}J_i(\mathbf{x}_1,\mathbf{x}_2)&=J_{ico}(\mathbf{x}_1,\mathbf{x}_2)+J_{iin}(\mathbf{x}_1,\mathbf{x}_2)\\&=\Gamma_{ico}(\mathbf{x}_1,\mathbf{x}_2,0)+\Gamma_{iin}(\mathbf{x}_1,\mathbf{x}_2,0)\end{aligned} \tag{49}$$

From (37), (43) and Yuen (2011k), (14), the total cross-spectral density function in the far field of the image,

$$\hat{\Gamma}(\varsigma_1\mathbf{u}_1,\varsigma_2\mathbf{u}_2,\omega)=\hat{\Gamma}_{co}(\varsigma_1\mathbf{u}_2,\varsigma_2\mathbf{u}_2,\omega)+\hat{\Gamma}_{in}(\varsigma_1\mathbf{u}_1,\varsigma_2\mathbf{u}_2,\omega)$$

$$= (2\pi)^6 \breve{\Gamma}_{ico}(k\mathbf{u}_1, -k\mathbf{u}_2, \omega)\{\exp[ik(\varsigma_1 - \varsigma_2)]\}/(\varsigma_1\varsigma_2)$$

$$+ (2\pi)^6 \breve{\Gamma}_{iin}(k\mathbf{u}_1, -k\mathbf{u}_2, \omega)\{\exp[ik(\varsigma_1 - \varsigma_2)]\}/(\varsigma_1\varsigma_2)$$

$$= (2\pi)^6 \breve{\Gamma}_{i}(k\mathbf{u}_1, -k\mathbf{u}_2, \omega)\{\exp[ik(\varsigma_1 - \varsigma_2)]\}/(\varsigma_1\varsigma_2)$$

(50)

From (38), (44) and Yuen (2011k), (15), the total radiant intensity in the far field of the image

$$\Lambda(\mathbf{u}, \omega) = \Lambda_{co}(\mathbf{u}, \omega) + \Lambda_{in}(\mathbf{u}, \omega)$$

$$= (2\pi)^6 \breve{\Gamma}_{ico}(k\mathbf{u}, -k\mathbf{u}, \omega) + (2\pi)^6 \breve{\Gamma}_{iin}(k\mathbf{u}, -k\mathbf{u}, \omega)$$

$$= (2\pi)^6 \breve{\Gamma}_{i}(k\mathbf{u}, -k\mathbf{u}, \omega). \qquad (51)$$

For a three-dimensional object, use all orientations of the object. According to the chosen resolution, $\omega_{\max}$ is employed. The radiant intensity and the magnitude of the spectral degree of coherence in the far field $\left|\hat{\Gamma}(\varsigma_1\mathbf{u}_1, \varsigma_2\mathbf{u}_2, \omega)\right|$ are measured for $\omega \le \omega_{\max}$.

(B) Inverse problem using the spectral degree of coherence in the far field of the image

Assume that $|c_{10}(\omega)|$ and $c_{01}(\omega)$ have been determined.

As in Yuen (2011i), (17), apart from the factor $(2\pi)^6\{\exp[ik(\varsigma_1 - \varsigma_2)]\}/(\varsigma_1\varsigma_2)$, the cross-spectral density function in the far field of the image for the coherent wave $\hat{\Gamma}_{co}(\varsigma_1\mathbf{u}_1, \varsigma_2\mathbf{u}_2, \omega)$ (see (37)) is the six-dimensional (or four-dimensional for planar object) spatial Fourier transform of $\hat{\Gamma}_{ico}(\mathbf{x}_1,-\mathbf{x}_2,\omega)$. As in Yuen (2011j), (16), apart from the factor $(2\pi)^6\{\exp[ik(\varsigma_1-\varsigma_2)]\}/(\varsigma_1\varsigma_2)$, The cross-spectral density function in the far field of the image for the incoherent wave $\hat{\Gamma}_{in}(\varsigma_1\mathbf{u}_1,\varsigma_2\mathbf{u}_2,\omega)$ (see (43)) is the six-dimensional (or four-dimensional for planar object) spatial Fourier transform of $\hat{\Gamma}_{iin}(\mathbf{x}_1,-\mathbf{x}_2,\omega)$. Hence the total cross-spectral density function in the far field $\hat{\Gamma}(\varsigma_1\mathbf{u}_1,\varsigma_2\mathbf{u}_2,\omega)$ is the six-dimensional (or four-dimensional for planar object) spatial Fourier transform of

$$(2\pi)^6[\hat{\Gamma}_{ico}(\mathbf{x}_1,-\mathbf{x}_2,\omega)+\hat{\Gamma}_{iin}(\mathbf{x}_1,-\mathbf{x}_2,\omega)]\{\exp[ik(\varsigma_1-\varsigma_2)\}/(\varsigma_1\varsigma_2)$$

$$=(2\pi)^6\hat{\Gamma}_i(\mathbf{x}_1,-\mathbf{x}_2,\omega)\{\exp i\bar{k}(\varsigma_1-\varsigma_2)]\}(\varsigma_1\varsigma_2),$$

from (50).

As in Yuen (2011i), Sect. 2.2, (C), iterative algorithms with oversampling in Yuen (2011e) are applied to obtain the phase of $\hat{\Gamma}(\varsigma_1\mathbf{u}_1,\varsigma_2\mathbf{u}_2,\omega)$ from the magnitude. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011f)). The solution $\hat{\Gamma}_i(\mathbf{x}_1,-\mathbf{x}_2,\omega)$ is obtained. Use $\hat{\Gamma}_i(\mathbf{x}_1,-\mathbf{x}_2,\omega)$ in (51). The radiant intensity obtained should agree with the experimentally measured values.

As in Yuen (2011k), Sect. 2.2, (B), replace $-\mathbf{x}_2$ by $\mathbf{x}_2$ in $\hat{\Gamma}_i(\mathbf{x}_1,-\mathbf{x}_2,\omega)$. The cross-spectral density function of the image $\hat{\Gamma}_i(\mathbf{x}_1,\mathbf{x}_2,\omega)$ is obtained.

The intensity of the image $\hat{I}_i(\mathbf{x},\omega)=\hat{\Gamma}_i(\mathbf{x},\mathbf{x},\omega)$ (52)

The intensity distribution of the image is given by $\int_0^\infty \hat{I}_i(\mathbf{x},\omega)d\omega$ (53)

We try to deal with the inverse problem in terms of the coherent and incoherent components. Since we do not have polychromatic coherent wave $\{c'_{10}(\omega)W_c(\omega)\}$, and polychromatic incoherent wave $\{c'_{01}(\omega)W_i(\omega)\}$ emitted by the object, we cannot use the analytical method in Yuen (2011h), Sect. 2.3. We can only use the numerical method in Yuen (2011h), Sect. 2.2, (B). For the coherent wave $c_{10}(\omega)W_c(\omega)$, the object exhibits $c_{10}(\omega)\hat{f}(\mathbf{x}_o,\omega)$, a coherent image $c_{10}(\omega)g(\mathbf{x})$ of the object $c_{10}(\omega)\hat{f}(\mathbf{x}_o,\omega)$ is formed (Goodman (2005), (5-38)).

$$\hat{g}(\mathbf{x},\omega) = \hat{f}(\mathbf{x},\omega)\otimes h(\mathbf{x},\omega) \tag{54}$$

Intensity of the image $\hat{I}_i(\mathbf{x},\omega) = \hat{\Gamma}_i(\mathbf{x},\mathbf{x},\omega)$

$$= |c_{10}(\omega)|^2\,\hat{g}(\mathbf{x},\omega)\hat{g}^*(\mathbf{x},\omega) + c_{01}(\omega)\hat{\rho}(\mathbf{x},\omega)\otimes|h(\mathbf{x},\omega)|^2$$

$$= |c_{10}(\omega)|^2\left|\hat{f}(\mathbf{x},\omega)\otimes h(\mathbf{x},\omega)\right|^2 + c_{01}(\omega)\hat{\rho}(\mathbf{x},\omega)\otimes|h(\mathbf{x},\omega)|^2 \tag{55}$$

(from (52), (42), (54)).

The impulse response $h(\mathbf{x})$ is close to a δ function (Goodman (2005), (5-24)). Hence

$$I_i(\mathbf{x},\omega) \approx |c_{10}(\omega)|^2\left|\hat{f}(\mathbf{x},\omega)\otimes\delta(\mathbf{x})\right|^2 + c_{01}(\omega)\hat{\rho}(\mathbf{x},\omega)\otimes|\delta(\mathbf{x})|^2 \tag{56}$$

$\delta^2(\mathbf{x})$ is undefined (Papoulis (1962), p. 275, Folland (1992), p. 319). However, we are only using an approximate δ function, and $\delta^2(\mathbf{x})$ has shape close to that of $\delta(\mathbf{x})$. Therefore

$$I_i(\mathbf{x},\omega) \approx |c_{10}(\omega)|^2 \left|\hat{f}(\mathbf{x},\omega)\right|^2 + c_{01}(\omega)\hat{\rho}(\mathbf{x},\omega)$$

$$= [|c_{10}(\omega)|^2 + c_{01}(\omega)]\left|\hat{f}(\mathbf{x},\omega)\right|^2 \tag{57}$$

from (29).

Hence an approximate $\left|\hat{f}(\mathbf{x},\omega)\right|^2$ (and $\hat{\rho}(\mathbf{x},\omega)$) is obtained. Use these as initial values and substitute into (55). By fine adjustments, subjected to the condition of (29), more accurate $\left|\hat{f}(\mathbf{x},\omega)\right|^2$ and $\hat{\rho}(\mathbf{x},\omega)$ are obtained. $\left|\hat{f}(\mathbf{x}_o,\omega)\right|$ determined should satisfy the constraints in Yuen (2011a, 2011c or 2011d). $\hat{\rho}(\mathbf{x}_o,\omega)$ determined should satisfy the constraints in Yuen (2011a or 2011c). Approximate intensity of the object is given by (20). This is an approximate value of that in Yuen (2011k), (5).

Natural resolution of the object is dealt with. Empirical impulse response $h(\mathbf{x},\omega)$ determined in Sect. 2.3 of Yuen (2008) will be used. They take into account aberrations of the lens and other defects of the system. We have by-passed the Sparrow or Rayleigh criterion of resolution (or alike) of the image, and the corresponding angular resolution.

2.3. Lenslets microscope and lenslets telescope. Direct and inverse problems

According to the chosen resolution, $\omega_{\max}$ is employed. Narrow-band filters are used to transmit narrow bands of the spectrum for all ω with $\omega \le \omega_{\max}$ (Wolf & James (1996), Sect. 7.1, 7.2, Wolf (2007), p. 68-69).

Consider each quasi-monochromatic wave of mean angular frequency $\overline{\omega}$ transmitted by a filter. As in Yuen (2011i), Sect. 2.3, we can use measurement of the magnitude of the mutual intensity in the far field $|J(\varsigma_1,\varsigma_2)|$. Assume that $|c_{10}(\overline{\omega})|$ and $c_{01}(\overline{\omega})$ have been determined.

(A) Direct problem

Two-dimensional array of lenslets can be used to form lenslets microscope or lenslets telescope. The array of lenslets forms a linear system. Its impulse response is formed by adding replica of $h(\mathbf{x},\overline{\omega})$ for one lenslet successfully in x_1, x_2 to give a periodic function, the extended $h(\mathbf{x},\overline{\omega})$. The periods are chosen such that the Fourier coefficients of this extended $h(\mathbf{x},\overline{\omega})$ are nonzero (Yuen (2008), (D)). The focal length of the lenslets, the object distance and the unit cell constants a,b of the array of lenslets are chosen so that the image predicted by geometrical optics in one lenslet can be contained in one unit cell (Yuen (2008), (D), constraint (i)).

In the far field, the radiant intensities and mutual intensities of the images in the lenslets are identical. The weak radiant intensities and mutual intensities of the two-dimensional array of images of the object are collected to form strong diffraction rods; it is enhanced by the array of images.

The radiant intensity and the magnitude of the mutual intensity $|J(\varsigma_1,\varsigma_2)|$ of the diffraction rods of the image in all the lenslets are measured. For a three-dimensional object, use all orientations of the object.

(B) Inverse problem using the mutual intensity of the diffraction rods of the image in all the lenslets

Iterative algorithms with oversampling in Yuen (2011e) are applied to obtain the phase of the mutual intensity of the diffraction rods $J(\varsigma_1,\varsigma_2)$ from the magnitude. Powerful methods in structure determination in X-ray crystallography can also be applied (Yuen (2011f)). The solution, $J_{il}(\mathbf{x}_1,-\mathbf{x}_2)$, is obtained (Yuen (2011g), Sect. 2.3, (B)). $J_{il}(\mathbf{x}_1,\mathbf{x}_2)$ is the mutual intensity of the image in all the lenslets.

Replace $-\mathbf{x}_2$ by $\mathbf{x}_2$. The mutual intensity of the image in all the lenslets is obtained.

For $\mathbf{x}_1=\mathbf{x}_2=\mathbf{x}$, the intensity of the image in all lenslets is obtained.

Intensity of the image in all lenslets
= intensity of image in all the lenslets for the coherent component
+ intensity of image in all the lenslets for the incoherent componet

From the intensity of image in all the lenslets, we obtain the approximate value of the intensity of the image in one lenslet $\hat{I}_i(\mathbf{x},\overline{\omega})$, in particular, for sufficiently large unit cell.

From (55),

$$\hat{I}_i(\mathbf{x},\overline{\omega}) = \left|c_{10}(\overline{\omega})\hat{g}(\mathbf{x},\overline{\omega})\right|^2 + c_{01}(\overline{\omega})\hat{\rho}(\mathbf{x},\overline{\omega}) \otimes \left|h(\mathbf{x},\overline{\omega})\right|^2$$

$$= \left|c_{10}(\overline{\omega})\right|^2 \left|\hat{f}(\mathbf{x},\overline{\omega}) \otimes h(\mathbf{x},\overline{\omega})\right|^2 + c_{01}(\overline{\omega})\hat{\rho}(\mathbf{x},\overline{\omega}) \otimes \left|h(\mathbf{x},\overline{\omega})\right|^2$$

Use the numerical method in Sect. 2.2, (B), approximate $\left|\hat{f}(\mathbf{x}_o,\omega)\right|$ and $\hat{\rho}(\mathbf{x}_o,\omega)$ of the object are obtained. Approximate intensity of the object is given by (20). Natural resolution of the object is dealt with. Empirical impulse response $h(\mathbf{x},\omega)$ will be used. They take into account aberrations of the lens and other defects of the system. The Sparrow or Rayleigh criterion of resolution (or alike) of the image is bypassed.

3. Discussions and conclusion

This article deals with the direct and inverse problems of imaging of object emitting polychromatic partially coherent light wave which is a combination of a polychromatic coherent wave and a polychromatic incoherent wave. Radiant intensity, cross-spectral density function or mutual intensity is dealt with.

For lensless imaging, in the inverse problem, $c_{10}(\omega)\hat{f}(\mathbf{x}_o,\omega)$, $c_{01}(\omega)\hat{\rho}(\mathbf{x}_o,\omega)$, intensity and intensity distribution of the object are determined.

For imaging of an object in a lens, in the inverse problem, spectral degree of coherence in the far field of the image is used. Using a numerical method, approximate $\left|\hat{f}(\mathbf{x}_o,\omega)\right|$, $\hat{\rho}(\mathbf{x}_o,\omega)$ and intensity of the object can be determined. Natural resolution of the object is dealt with. Empirical impulse response $h(\mathbf{x},\omega)$ will be used. They take into account aberrations of the lens and other defects of the system. We have by-passed the Sparrow or Rayleigh criterion of resolution (or alike) of the image, and the corresponding angular resolution.

The method used in the inverse problem in Sect. 2.2 (B) is not very satisfactory as it involves numerical method and in the adjustment, the phase of $f(\mathbf{x})$ is a variable. Conceptually it will be better if we can analytically divide the cross-spectral density function of the image $\hat{\Gamma}_i(\mathbf{x}_1, \mathbf{x}_2, \omega)$ into two parts for the coherent and incoherent components (cf. Yuen (2011h), Sect. 2.3). Then we can use the solutions to the inverse problems for the coherent and incoherent components obtained in Yuen (2011i, 2011j), Sect. 2.2, (B). If a filter can be developed to absorb the coherent component, then we can deal with the remaining incoherent component. Iterative algorithms with oversampling can be applied to the cross-spectral density function in the far field of the image for the incoherent component. The cross-spectral density function of the image for the coherent component is equal to the cross-spectral density function of the image minus the cross-spectral density function of the image for the incoherent component. We can then deal with the coherent component.

For lenslets microscope or lenslets telescope, the inverse problem uses the mutual intensity of the diffraction rods in the far field of the image in all the lenslets. Approximate intensities of the object and image for the partially coherent wave are obtained. Natural resolution of the object is dealt with. Empirical impulse response $h(\mathbf{x}, \omega)$ is used.

In combination with iterative algorithms and oversampling, powerful methods in X-ray crystallography can also be applied.

This article illustrates that, in dealing with polychromatic partially coherent wave in an imaging system, the polychromatic coherent wave and the polychromatic incoherent wave can be treated separately. The results can then be combined to give the result for the polychromatic partially coherent wave. In the direct problem, the problem of polychromatic partially coherent wave in an imaging system reduces to the problems of polychromatic coherent wave and polychromatic incoherent wave in the imaging system. For the inverse problem in lensless imaging, information from Yuen (2011k) is used. In Sect. 2.1, (B) of this article, it is stated that 'However, according to Yuen (2011k), Sect. 2.1, (B), a phase-retrieval method can, in fact, be applied to obtain the phase of the cross-spectral density function in the far field.' More insight is obtained

In astronomy, phase-retrieval method can also be used in interferometric imaging (Dainty & Fienup (1987)). Many methods have been devised to solve the phase problem and obtain the structure of the object (see, for example, interferometric imaging in Dainty & Fienup (1987), Sect. 7.5). Now the structure of the object can be determined by the method presented in this series of articles of imaging. May be we can work backward with these methods to solve problems encountered, for example, interferometry, coherence of waves, atmospheric disturbance and speckles.

It will be very interesting and useful if this series of articles can be applied to studies of scalar or electromagnetic waves in scattering, inverse scattering and in optical systems (Beran & Parrent (1964), Marathay & Parrent (1970), Boerner (1980), Marathay (1982), Peřina (1985), Mandel & Wolf (1995), Brosseau (1998), Wolf (2007)). Effects of coherent and incoherent superpositions and decompositions of coherent, incoherent, polarized and unpolarized components can be investigated. Both theoretical and experimental investigations are needed. In particular, experimental data of diffraction intensity, radiant intensity, mutual intensity, equal-time complex degree of coherence, cross-spectral density function and spectral degree of coherence in the far zone are needed. Many researches can be done. For example, find the sources that generate the same output intensity distribution (Martínez-Herrero & Mejídas (1982)).

Experiments about polychromatic partially coherent light wave which is a combination of a polychromatic coherent wave and a polychromatic incoherent wave, in optical systems are needed. We would like to emphasize that it is very important and useful the develop filters to absorb the coherent or incoherent components. We can then just deal with the coherent or components separately, and add the results. The problem of partial coherence can be much simplified.

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Miscellaneous

12

An approach for determining nuclear structure and force by means of phase retrieval in scattering

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Abstract

An approach is presented for determining nuclear structure and force by means of phase retrieval in scattering. An illustration is provided. It shows that phase retrieval in investigating nuclear physics is a possibility. One of the purposes of this article is to stimulate interest in phase-retrieval approach in nuclear physics, an open research area. It is direct, efficient and quantitative. The scattered intensity is transformed to a fictitious object which may be used as fundamental data for studies of nuclear physics. General functions of potential energy, charge and mass distributions and nuclear forces are used. A consistent set of nuclear charge and mass distributions (including shape and dimensions) and two-body central nuclear forces for all real centrosymmetric nuclei may be obtained. They can be verified by applying to many aspects of nuclear physics. These empirical functions help investigation of the law governing charge and mass

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1. Introduction

To determine properties of a nucleus, we can use electromagnetic wave and particles such as neutron, electron, proton and other nuclei incident on the nucleus. All wavelength and velocity from all direction of the incident radiation are used. The intensities of the scattered radiation in all directions are measured. The properties of the nucleus are to be obtained from these intensity data. These properties include the size, structure, the force between the constituents of the nucleus and the law governing the distributions of the constituents and their interactions. The size is the shape and dimensions. The constituents of the nucleus are the charge of the protons and electrons, and the mass of the neutrons, protons and electrons. To determine the properties of the nucleus from the scattered intensity is an inverse problem, or inverse scattering problem.

In the past hundred years, researchers have developed theories and models of the nucleus in order to fit to these experimental intensity data. The properties of the incident particle and target nucleus have to be determined. The interaction between the incident particle and the target nucleus is also unknown. Because of the large number of unknowns or factors involved, this is a very complicate and difficult problem. Many results have been obtained. But a theory accounting for all the intensity data and all other experimental data (see Sect. 4) remains to be completed.

An approach is to develop a method to proceed directly from the experimental intensity data and obtain the properties of the nucleus. Consider electromagnetic wave incident on an object. It is very difficult to believe that there are two different objects, other than mirror images, which show the same scattered intensity in all directions, for incident electromagnetic wave of all wavelengths in all directions. Hence the object should be uniquely determined by all the scattered intensities. Uniqueness means unique up to the mirror images. Now we have four factors: the incident electromagnetic wave, the object, the scattered wave, and an expression connecting these three factors. From theory, the scattering amplitude (magnitude and phase) is the Fourier transform of the electron density function of the object. This is the expression connecting the incident wave, the scattered wave and the object. The magnitude of the scattering amplitude is proportional to the scattered intensity. Hence the magnitude is known. In general, the phase cannot be measured, and is thought to be lost. This is the phase problem. By the uniqueness argument of the object in above, it means that the phase is uniquely determined by the magnitude; it is just hidden in the magnitude, not lost. The problem is to find the phase from the magnitude. This is the phase-retrieval problem.

For the incident wave, we use all wavelength and in all direction. For the scattered wave, the intensity in all directions is measured. The only factor that we can deal with is the object. Hence the phase-retrieval problem is reduced to the mathematical problem: What are the constraints of the object, under which the phase of its Fourier transform can be uniquely obtained from the magnitude? The constraints must be very general as we are dealing with all kinds of objects and we expect to find a solution. Then we need only consider all general and reasonable constraints such as finite size, real, symmetric, non-negative, bounded, continuous, differentiable, infinitely differentiable, analytic etc., so that they can provide a unique solution to the phase-retrieval problem. For an object subjected to the constraints, finite size, real, centrosymmetric and bounded, a unique solution is presented in Ref. [1].

For nucleus with incident electromagnetic wave of all wavelengths, and incident particles of all species with all velocities, in all directions, it is more difficult to believe that there are two different nuclei, other than mirror images, which have the same scattered intensity in all directions, and are compatible to all other experimental data. Hence the structures of the incident particle and the target nucleus, and their interactions should be uniquely determined by all the scattered intensities and all other experimental data. This means that the phase of the scattered wave is uniquely determined by the magnitude of the scattering amplitude and all other experimental data. There are five factors: the incident particle, the target nucleus, the scattered particle, the interaction between the particle and the target nucleus, and an expression connecting these four factors.

For the purpose of illustration, in this article, general functions of potential energies are employed to determine a consistent set of charge distributions and mass distributions of neutron, proton and electron in all real centrosymmetric nuclei, and the potential energy functions between these distributions (Sect. 3.2.5). The potential energy functions between different pairs of neutron, proton and electron are treated as different. These results can then be verified by applying to many aspects of nuclear physics such as energies, decay processes, isomers, reactions etc. (Sect. 4). For refinement, general functions of charge and mass distributions and nuclear forces are employed in Sect. 5.1. In Sect. 5.2, correlation between potential energies, charge and mass distributions and nuclear forces is included. This amounts to using general functions of potential energies, charge and mass distributions and nuclear forces. In Sect. 5.3, more general relation between an object and its diffraction pattern is used. In Sect. 5.4, the nuclear charge is bounded. In Sect. 6, nuclear size (shape and dimensions) and surface distributions are determined. Sect. 7 presents direct determination of the diameter and shape (and dimensions) of the convex hull (envelope) of a nucleus by means of the minimum-phase function. Various powerful methods have been developed in structure determination in X-ray crystallography. These can be applied in nuclear structure determination (Sect. 8).

Much work has been done by many researchers in nuclear physics, structure, force, reaction, etc. Here we can only give a very small number of references ([2] to [49]).

2. Scattering of electromagnetic wave by an object

The object can be a fixed or rotating nucleus about any number of axes. This is determination of the charge distribution $f_c(\mathbf{r})$ in the object by measuring the intensity of the diffraction pattern. We use an assumption

(a) the charge in the nucleus $f_c(\mathbf{r})$ is free.

When the first Born approximation is applicable ([50], Sect. 3.8), the diffraction pattern

$$F_c(\mathbf{K}) = \int f_c(\mathbf{r}) \exp(-i\mathbf{K}\cdot\mathbf{r}) d\mathbf{r} \qquad (1)$$

This amounts to determining the phase of the Fourier transform of $f_c(\mathbf{r})$ from the magnitude – the phase-retrieval problem. An analytical solution for the phase problem of multidimensional centrosymetric real object is presented in Ref. [1], case (C). The constraints of the object are: finite size, real, centrosymmetric and bounded. For efficient computations, iterative algorithms with oversampling may be employed. Apply Ref. [51] or [52] to centrosymmetric or spherically symmetric real nonperiodic objects. Hence the charge distribution, including shape and dimensions, of a nucleus is determined up to a multiplicative factor of -1. As proton charge is positive, the appropriate sign is chosen.

3. Scattering of a particle by an object

3.1. Phase-retrieval – determination of a fictitious object $v(\mathbf{r})$

We use an assumption

(b) The system of the incident particle, the target nucleus and their interaction is represented by a fictitious object $v(\mathbf{r})$. The wave with wavevector $\mathbf{k}_0$ is incident on this object and is scattered classically. The diffraction pattern

$$V(\mathbf{K}) = \int v(\mathbf{r}) \exp(-i\mathbf{K}\cdot\mathbf{r}) d\mathbf{r} \qquad (2)$$

where $\mathbf{K} = \mathbf{k} - \mathbf{k}_0$. $\mathbf{k}$ is wavevector of the scattered wave, $k = k_0$

$|V(\mathbf{K})|^2$ is proportional to the intensity which is measured experimentally. The phase of $V(\mathbf{K})$ cannot be measured. Let the object $v(\mathbf{r})$ be subjected to constraints:

(i) $v(\mathbf{r})$ has finite size from $-\mathbf{R}$ to $\mathbf{R}$ ($|\mathbf{R}|$ is a function of direction. Its value will be determined).

(ii) $v(\mathbf{r})$ is real

(iii) $v(\mathbf{r})$ is centrosymmetric, $v(\mathbf{r}) = v(-\mathbf{r})$

(iv) $v(\mathbf{r})$ is bounded

It is shown in Ref. [1], case (C), that the phase of $V(\mathbf{K})$ can be uniquely determined from $|V(\mathbf{K})|$ for all $\mathbf{K}$. Use iterative algorithms with oversampling (Ref. [51] or [52]) for centrosymmetric or spherically symmetric real nonperiodic objects. Thus $V(\mathbf{K})$ and hence $v(\mathbf{r})$ are obtained.

3.2. Nuclear structure and force

Two-body central force and non-relativistic case are considered. Consider a mass distribution $f_m(\mathbf{r})$ of the target nucleus, and a two-body potential energy function $g(r)$ of unit mass of the incident particle at $\mathbf{r}$ due to unit mass of the target nucleus at the origin. For an incident point-particle of unit mass, the potential energy of the particle due to the target nucleus, $= \int f_m(\mathbf{r}')g(|\mathbf{r}-\mathbf{r}'|)d\mathbf{r}' = f_m(\mathbf{r}) \otimes g(r)$

$\otimes$ stands for convolution.

We use an assumption

(c) The fictitious object $v(\mathbf{r})$ is equal to this potential energy,

$$v(\mathbf{r}) = f_m(\mathbf{r}) \otimes g(r)$$

Combination of (2) and assumption (c) is the first Born approximation (Ref. [31], (8.3)).

We consider a centrosymmetric nucleus. Let $f_m(\mathbf{r})$ be the mass distribution of the protons and neutrons. In order for $v(\mathbf{r})$ to satisfy the four constraints, $f_m(\mathbf{r})$ is subjected to the constraints:

(i) $f_m(\mathbf{r})$ has finite size which will be determined

(ii) $f_m(\mathbf{r})$ is real

(iii) centrosymmetric, $f_m(\mathbf{r}) = f_m(-\mathbf{r})$

$g(r)$ is spherically symmetric. The constraints for $g(r)$ are:

(i) $g(r)$ has finite range. This constraint is necessary for the notion of scattering to make sense.

(ii) $g(r)$ is real

The constraint for $f_m(\mathbf{r})$ and $g(r)$ together is:

(iv) Fourier transform of $f_m(\mathbf{r}) \otimes g(r)$ exists

The constraint (i) $g(r)$ has finite range, is used so that $v(\mathbf{r})$ has finite size. It is known that the nuclear force has short range. Here we only use the condition that $g(r)$ has finite range; the range can be very large but must be finite. Coulomb force has infinite range. If the nuclear size or force extends to infinity, though very small, or if there is Coulombic interaction between the incident particle and the target nucleus, $v(\mathbf{r})$ has infinite size. Due to experimental limitation, only a finite part of $v(\mathbf{r})$ is used. Use assumptions

(d) charge independence -- the potential energy functions between unit masses of different pairs of electron, proton and neutron are the same.

(e) Mass distribution of protons $f_p(\mathbf{r})$ is proportional to charge distribution of protons $f_c(\mathbf{r})$. Let $b =$ mass of a proton / charge of a proton.

$$f_p(\mathbf{r}) = bf_c(\mathbf{r}) \tag{3}$$

Since the charge distribution of the protons is determined in Sect. 2, the mass distribution of the protons $f_p(\mathbf{r})$ is known.

Neutron scattering

Here and in the following, by assumption (b) and the constraints for $v(\mathbf{r})$, $V(\mathbf{K})$ and $v(\mathbf{r})$ are obtained.

Consider a target nucleus with proton mass distribution $f_p(\mathbf{r})$ and neutron mass distribution $f_n(\mathbf{r})$. The initial stage is to regard the incident neutron as a point particle with mass m_n. The potential energy of the neutron due to the target nucleus

$$= m_n f_m(\mathbf{r}) \otimes g(r) \tag{4}$$

By assumption (c), $v(\mathbf{r}) = m_n f_m(\mathbf{r}) \otimes g(r)$

As an initial stage, let the target nucleus be spherically symmetric or freely rotating. (4) becomes $v(r) = m_n f_m(r) \otimes g(r)$

Its Fourier transform $V(K) = m_n F_m(K) G(K)$ (5)

where $F_m(K)$ and $G(K)$ are the Fourier transforms of $f_m(r)$ and $g(r)$ respectively.

Use assumption

(f) $\dfrac{f_n(r)}{f_p(r)} = \dfrac{N}{Z}$

where $f_n(r)$ is the mass distribution of the neutrons

N is the number of neutrons

Z is the number of protons

$f_p(\mathbf{r})$ is found in (e). $f_p(r)$ is the spherical average of $f_p(\mathbf{r})$. Hence $f_n(r)$ is known. $f_m(r) = f_p(r) + f_n(r)$ (6)

Hence $F_m(K)$ for each species of nucleus is known. In (5), $G(K)$ is obtained. If we use all species of nucleus, say N, find the 'best' fit of $G(K)$ to the N equations (5). Using this 'best' $G(K)$, find $F_m(K)$ in (5) for each species of nucleus. Its inverse Fourier transform gives $f_m(r)$. From (6), an improved value of $f_n(r)$ is obtained. The validity of assumption (f) can be examined. The inverse Fourier transform of the 'best' $G(K)$ gives $g(r)$. Thus we can obtain the charge distribution and approximate values of the mass distributions of protons and neutrons in all spherically symmetric or freely rotating nuclei, including isotopes, and the nuclear force. These will be used as initial values in the process of obtaining a consistent set in the following.

Now constraint (f) is disregarded. Consider an incident neutron with mass distribution $f_{ni}(r)$. Note that here we do not assume any shape for the protons and neutrons in the nucleus. By assumption (c),

$v(\mathbf{r})$ = potential energy of the incident neutron due to the nucleus,

$$= \iint f_p(\mathbf{r}')g(|\mathbf{r}''-\mathbf{r}'|)f_{ni}(|\mathbf{r}''-\mathbf{r}|)d\mathbf{r}'d\mathbf{r}''+\iint f_n(\mathbf{r}')g(|\mathbf{r}''-\mathbf{r}'|)f_{ni}(|\mathbf{r}''-\mathbf{r}|)d\mathbf{r}'d\mathbf{r}'' \quad (7)$$

$$= f_p(\mathbf{r})\otimes g(r)\otimes f_{ni}(r) + f_n(\mathbf{r})\otimes g(r)\otimes f_{ni}(r) \quad (8)$$

where $\mathbf{r}$ denotes the center of the incident neutron

Its Fourier transform

$$V(\mathbf{K}) = F_p(\mathbf{K})G(K)F_{ni}(K) + F_n(\mathbf{K})G(K)F_{ni}(K) \quad (9)$$

where $F_p(\mathbf{K})$, $F_{ni}(K)$ and $F_n(\mathbf{K})$ are the Fourier transforms of $f_p(\mathbf{r})$, $f_{ni}(r)$ and $f_n(\mathbf{r})$ respectively. $f_p(\mathbf{r})$ is determined in (3). Hence $F_p(\mathbf{K})$ is known. $F_p(\mathbf{K})$ is the nuclear form factor for protons, $F_n(\mathbf{K})$ for neutrons.

Electron-neutron scattering with the electron treated as a point particle

$$v(\mathbf{r}) = f_n(\mathbf{r})\otimes g(r)m_e \quad (10)$$

where m_e is the mass of the incident electron.

Its Fourier transform, $V(\mathbf{K}) = F_n(\mathbf{K})G(K)m_e$ (11)

Electron-nucleus scattering with the electron treated as a point particle

Similar to (8),

$$v(\mathbf{r}) = f_c(\mathbf{r})\otimes[-e/r] + f_p(\mathbf{r})\otimes g(r)m_e + f_n(\mathbf{r})\otimes g(r)m_e \quad (12)$$

where e is | charge of electron | .

Use a convergence factor $\exp(-r\lambda)$. The Fourier transform of (12) with $\lambda \to 0$, is

$$V(\mathbf{K}) = -\frac{4\pi}{K^2} eF_c(\mathbf{K}) + F_p(\mathbf{K})G(K)m_e + F_n(\mathbf{K})G(K)m_e \tag{13}$$

For electron-proton scattering, the neutron term in the right-hand side of (12) is absent.

Proton scattering

$$v(\mathbf{r}) = f_c(\mathbf{r}) \otimes \frac{1}{r} \otimes f_{ci}(r) + f_p(\mathbf{r}) \otimes g(r) \otimes f_{pi}(r) + f_n(\mathbf{r}) \otimes g(r) \otimes f_{pi}(r) \tag{14}$$

where $f_{ci}(r)$ is the charge distribution of the incident proton, and $f_{pi}(r)$ its mass distribution.

By assumption (e), (see (3)), $f_{pi}(r) = bf_{ci}(r)$.

$$V(\mathbf{K}) = \frac{4\pi}{K^2} F_c(\mathbf{K})F_{ci}(K) + F_p(\mathbf{K})G(K)F_{pi}(K) + F_n(\mathbf{K})G(K)F_{pi}(K) \tag{15}$$

where $F_{ci}(K)$ is the Fourier transform of $f_{ci}(r)$.

$F_{pi}(K)$ is the Fourier transform of $f_{pi}(r)$.

Scattering of an incident light or heavy nucleus, for example, alpha particle

$$v(\mathbf{r}) = f_c(\mathbf{r}) \otimes \frac{1}{r} \otimes f_{ci}(r) + f_p(\mathbf{r}) \otimes g(r) \otimes f_{pi}(r) + f_p(\mathbf{r}) \otimes g(r) \otimes f_{ni}(r)$$
$$+ f_n(\mathbf{r}) \otimes g(r) \otimes f_{pi}(r) + f_n(\mathbf{r}) \otimes g(r) \otimes f_{ni}(r) \tag{16}$$

where $f_{ci}(r)$ is the charge distribution of the incident nucleus.

$f_{pi}(r)$ is its proton mass distribution, $f_{ni}(r)$ its neutron mass distribution.

$$V(\mathbf{K}) = \frac{4\pi}{K^2} F_c(\mathbf{K}) F_{ci}(K) + F_p(\mathbf{K}) G(K) F_{pi}(K) + F_p(\mathbf{K}) G(K) F_{ni}(K)$$

$$+ F_n(\mathbf{K}) G(K) F_{pi}(K) + F_n(\mathbf{K}) G(K) F_{ni}(K) \qquad (17)$$

where $F_{ni}(K)$ is the Fourier transform of $f_{ni}(r)$.

3.2.1. Gross consistent set of mass distributions and potential energy functions

For all fixed or rotating centosymmetic nuclei including isotopes and all incident particles, the system is excessively over-determined. A gross consistent set of the unknown functions $F_n(\mathbf{K})$ and $G(K)$ can be obtained from (9), (11), (13), (15) and (17). The approximate values obtained in the initial stage can be used as initial values in this process.

3.2.2. Different potential energy functions between different pairs of masses

After a consistent set is obtained, we may then refine the treatment by disregarding the constraints in assumption (d) charge independence, aiming at obtaining different potential energy functions between different pairs of masses.

Let $g_{en}(r)$ = potential energy function of unit mass of electron due to unit mass of neutron, and similar definitions for $g_{ep}(r)$, $g_{nn}(r)$, $g_{np}(r)$, $g_{pp}(r)$, where e, n, p denote electron, neutron and proton respectively. Let $G_{en}(K)$, $G_{ep}(K)$, $G_{nn}(K)$, $G_{np}(K)$ and $G_{pp}(K)$ denote their Fourier transforms respectively. The electron is regarded as a point particle.

For neutron scattering,

$$v(\mathbf{r}) = f_p(\mathbf{r}) \otimes g_{np}(r) \otimes f_{ni}(r) + f_n(\mathbf{r}) \otimes g_{nn}(r) \otimes f_{ni}(r) \qquad (18)$$

$$V(\mathbf{K}) = F_p(\mathbf{K}) G_{np}(K) F_{ni}(K) + F_n(\mathbf{K}) G_{nn}(K) F_{ni}(K) \qquad (19)$$

For electron-neutron scattering,

$$v(\mathbf{r}) = f_n(\mathbf{r}) \otimes g_{en}(r) m_e \tag{20}$$

Its Fourier transform, $V(\mathbf{K}) = F_n(\mathbf{K}) G_{en}(K) m_e$ (21)

For electron-nucleus scattering (see (12)),

$$v(\mathbf{r}) = f_c(\mathbf{r}) \otimes [-e/r] + f_p(\mathbf{r}) \otimes g_{ep}(r) m_e + f_n(\mathbf{r}) \otimes g_{en}(r) m_e \tag{22}$$

Its Fourier transform,

$$V(\mathbf{K}) = -\frac{4\pi}{K^2} e F_c(\mathbf{K}) + F_p(\mathbf{K}) G_{ep}(K) m_e + F_n(\mathbf{K}) G_{en}(K) m_e \tag{23}$$

For scattering of an incident light or heavy nucleus (see (16)),

$$\begin{aligned} v(\mathbf{r}) & \\ &= f_c(\mathbf{r}) \otimes \frac{1}{r} \otimes f_{ci}(r) + f_p(\mathbf{r}) \otimes g_{pp}(r) \otimes f_{pi}(r) + f_p(\mathbf{r}) \otimes g_{np}(r) \otimes f_{ni}(r) \\ &\quad + f_n(\mathbf{r}) \otimes g_{np}(r) \otimes f_{pi}(r) + f_n(\mathbf{r}) \otimes g_{nn}(r) \otimes f_{ni}(r) \end{aligned} \tag{24}$$

Its Fourier transform,

$$\begin{aligned} V(\mathbf{K}) & \\ &= \frac{4\pi}{K^2} F_c(\mathbf{K}) F_{ci}(K) + F_p(\mathbf{K}) G_{pp}(K) F_{pi}(K) + F_p(\mathbf{K}) G_{np}(K) F_{ni}(K) \\ &\quad + F_n(\mathbf{K}) G_{np}(K) F_{pi}(K) + F_n(\mathbf{K}) G_{nn}(K) F_{ni}(K) \end{aligned} \tag{25}$$

Proton scattering is a special case.

A consistent set is then obtained.

3.2.3. Mass distribution of protons may not be proportional to its charge distribution

After a consistent set is obtained in Sect. 3.2.2, we may further refine the treatment by disregarding assumption (e), (Eq. (3)). $f_p(\mathbf{r})$, $f_{pi}(r)$ and hence $F_p(\mathbf{K})$, $F_{pi}(K)$ in Sect. 3.2.2 are treated as unknowns to be determined. A consistent set can be found.

3.2.4. Nuclear electron included

After a consistent set is obtained in Sect. 3.2.3, electron in nucleus can be included.

For neutron scattering,

$$v(\mathbf{r}) = f_p(\mathbf{r}) \otimes g_{np}(r) \otimes f_{ni}(r) + f_n(\mathbf{r}) \otimes g_{nn}(r) \otimes f_{ni}(r)$$

$$+ f_e(\mathbf{r}) \otimes g_{en}(r) \otimes f_{ni}(r) \tag{26}$$

where $f_e(\mathbf{r})$ is the electron mass distribution in the nucleus.

Its Fourier transform

$$V(\mathbf{K}) = F_p(\mathbf{K}) G_{np}(K) F_{ni}(K) + F_n(\mathbf{K}) G_{nn}(K) F_{ni}(K)$$

$$+ F_e(\mathbf{K}) G_{en}(K) F_{ni}(K) \tag{27}$$

where $F_e(\mathbf{K})$ is the Fourier transform of $f_e(\mathbf{r})$.

For electron scattering, similar to (22),

$$v(\mathbf{r}) = f_c(\mathbf{r}) \otimes (1/r) \otimes f_{ci}(r) + f_p(\mathbf{r}) \otimes g_{ep}(r) \otimes f_{ei}(r)$$

$$+ f_n(\mathbf{r}) \otimes g_{en}(r) \otimes f_{ei}(r) + f_e(\mathbf{r}) \otimes g_{ee}(r) \otimes f_{ei}(r) \tag{28}$$

where $f_{ci}(r)$ is the charge distribution of the incident electron, $f_{ei}(r)$ its mass distribution.

$g_{ee}(r)$ = potential energy function of unit mass of electron due to unit mass of electron

Its Fourier transform

$$V(\mathbf{K}) = \frac{4\pi}{K^2} F_c(\mathbf{K}) F_{ci}(K) + F_p(\mathbf{K}) G_{ep}(K) F_{ei}(K)$$

$$+ F_n(\mathbf{K}) G_{en}(K) F_{ei}(K) + F_e(\mathbf{K}) G_{ee}(K) F_{ei}(K) \tag{29}$$

where $F_{ci}(K)$, $F_{ei}(K)$, and $G_{ee}(K)$ are the Fourier transforms of $f_{ci}(r)$, $f_{ei}(r)$ and $g_{ee}(r)$ respectively.

For scattering of an incident light or heavy nucleus (see (24)),

$$
\begin{aligned}
v(\mathbf{r}) = {} & f_c(\mathbf{r}) \otimes \frac{1}{r} \otimes f_{ci}(r) + f_p(\mathbf{r}) \otimes g_{pp}(r) \otimes f_{pi}(r) \\
& + f_p(\mathbf{r}) \otimes g_{np}(r) \otimes f_{ni}(r) + f_n(\mathbf{r}) \otimes g_{np}(r) \otimes f_{pi}(r) \\
& + f_n(\mathbf{r}) \otimes g_{nn}(r) \otimes f_{ni}(r) + f_p(\mathbf{r}) \otimes g_{ep}(r) \otimes f_{ei}(r) \\
& + f_n(\mathbf{r}) \otimes g_{en}(r) \otimes f_{ei}(r) + f_e(\mathbf{r}) \otimes g_{ee}(r) \otimes f_{ei}(r) \\
& + f_e(\mathbf{r}) \otimes g_{ep}(r) \otimes f_{pi}(r) + f_e(\mathbf{r}) \otimes g_{en}(r) \otimes f_{ni}(r) \qquad (30)
\end{aligned}
$$

where $f_{ci}(r)$ is the (electron and proton) charge distribution of the incident nucleus,
$f_{ei}(r)$ its electron mass distribution.

Its Fourier transform

$$
\begin{aligned}
V(\mathbf{K}) = {} & \frac{4\pi}{K^2} F_c(\mathbf{K}) F_{ci}(K) + F_p(\mathbf{K}) G_{pp}(K) F_{pi}(K) \\
& + F_p(\mathbf{K}) G_{np}(K) F_{ni}(K) + F_n(\mathbf{K}) G_{np}(K) F_{pi}(K) \\
& + F_n(\mathbf{K}) G_{nn}(K) F_{ni}(K) + F_p(\mathbf{K}) G_{ep}(K) F_{ei}(K) \\
& + F_n(\mathbf{K}) G_{en}(K) F_{ei}(K) + F_e(\mathbf{K}) G_{ee}(K) F_{ei}(K) \\
& + F_e(\mathbf{K}) G_{ep}(K) F_{pi}(K) + F_e(\mathbf{K}) G_{en}(K) F_{ni}(K) \qquad (31)
\end{aligned}
$$

where $F_{ei}(K)$ is the Fourier transform of $f_{ei}(r)$.

A consistent set can be obtained.

3.2.5. General function of potential energy

After a consistent set is obtained in Sect. 3.2.4, assumption (c) may be relaxed. In assumption (c), $v(\mathbf{r}) = f_m(\mathbf{r}) \otimes g(r)$ (32)

This may be inappropriate for the treatment of, for example, strong interaction. But if the first Born approximation is applicable in some range of potential energy (in fact, high energy (see, for example, Ref. [36], p. 238)), it means that $v(\mathbf{r})$ is a function of potential energy and possibly other variables whose effect is small in the energy range where the first Born approximation is applicable. To represent the dependence of $v(\mathbf{r})$ on the potential energy, we use a power series for $V(\mathbf{K})$, which gives a general function of $v(\mathbf{r})$. Although the number of equations for all incident particles and target nuclei is very large, it is still finite. Hence a polynomial is used. From (32),

$$v(\mathbf{r}) = \sum_{j=1}^{H} c_j \left[f_m(\mathbf{r}) \otimes g(r) \right] \otimes \ldots \otimes \left[f_m(\mathbf{r}) \otimes g(r) \right] \tag{33}$$

where $c_1 = 1$. H and c_2, c_3,… are to be determined.

The convolution is repeated j -times.

The approach can be compared with the Rytov approximation (Ref. [53], p. 492).

For neutron scattering (corresponds to (26), (27)),

$$\begin{aligned} v(\mathbf{r}) = \sum_{j=1}^{H} c_j \Big\{ & \left[f_p(\mathbf{r}) \otimes g_{np}(r) \otimes f_{ni}(r) \right] \otimes \ldots \otimes \left[f_p(\mathbf{r}) \otimes g_{np}(r) \otimes f_{ni}(r) \right] \\ & + \left[f_n(\mathbf{r}) \otimes g_{nn}(r) \otimes f_{ni}(r) \right] \otimes \ldots \otimes \left[f_n(\mathbf{r}) \otimes g_{nn}(r) \otimes f_{ni}(r) \right] \\ & + \left[f_e(\mathbf{r}) \otimes g_{en}(r) \otimes f_{ni}(r) \right] \otimes \ldots \otimes \left[f_e(\mathbf{r}) \otimes g_{en}(r) \otimes f_{ni}(r) \right] \Big\} \end{aligned} \tag{34}$$

where each convolution is repeated j -times.

$$\begin{aligned} V(\mathbf{K}) = \sum_{j=1}^{H} c_j \Big\{ & \left[F_p(\mathbf{K}) G_{np}(K) F_{ni}(K) \right]^j + \left[F_n(\mathbf{K}) G_{nn}(K) F_{ni}(K) \right]^j \\ & + \left[F_e(\mathbf{K}) G_{en}(K) F_{ni}(K) \right]^j \Big\} \end{aligned} \tag{35}$$

High energy scattering enters the picture through $\mathbf{K}$ in (2). For high energy scattering, only the term in (35) with $j = 1$ is important.

For electron scattering (corresponds to (28), (29)),

$$\begin{aligned} v(\mathbf{r}) = \sum_{j=1}^{H} c_j \Big\{ & \left[f_c(\mathbf{r}) \otimes (1/r) \otimes f_{ci}(r) \right] \otimes \ldots \otimes \left[f_c(\mathbf{r}) \otimes (1/r) \otimes f_{ci}(r) \right] \\ & + \left\lfloor f_p(\mathbf{r}) \otimes g_{ep}(r) \otimes f_{ei}(r) \right\rfloor \otimes \ldots \otimes \left\lfloor f_p(\mathbf{r}) \otimes g_{ep}(r) \otimes f_{ei}(r) \right\rfloor \end{aligned}$$

$$+\left[f_n(\mathbf{r})\otimes g_{en}(r)\otimes f_{ei}(r)\right]\otimes\ldots\otimes\left[f_n(\mathbf{r})\otimes g_{en}(r)\otimes f_{ei}(r)\right]$$
$$+\left[f_e(\mathbf{r})\otimes g_{ee}(r)\otimes f_{ei}(r)\right]\otimes\ldots\otimes\left[f_e(\mathbf{r})\otimes g_{ee}(r)\otimes f_{ei}(r)\right]\Big\} \tag{36}$$

where each convolution is repeated j -times.

$$V(\mathbf{K})=\sum_{j=1}^{H}c_j\left\{\left[\frac{4\pi}{K^2}F_c(\mathbf{K})F_{ci}(K)\right]^j+\left[F_p(\mathbf{K})G_{ep}(K)F_{ei}(K)\right]^j\right.$$
$$\left.+\left[F_n(\mathbf{K})G_{en}(K)F_{ei}(K)\right]^j+\left[F_e(\mathbf{K})G_{ee}(K)F_{ei}(K)\right]^j\right\} \tag{37}$$

For scattering of an incident light or heavy nucleus (corresponds to (30), (31)),

$$v(\mathbf{r})=\sum_{j=1}^{H}c_j\left\{\left[f_c(\mathbf{r})\otimes\frac{1}{r}\otimes f_{ci}(r)\right]\otimes\ldots\otimes\left[f_c(\mathbf{r})\otimes\frac{1}{r}\otimes f_{ci}(r)\right]\right.$$
$$+\left\lfloor f_p(\mathbf{r})\otimes g_{pp}(r)\otimes f_{pi}(r)\right\rfloor\otimes\ldots\otimes\left\lfloor f_p(\mathbf{r})\otimes g_{pp}(r)\otimes f_{pi}(r)\right\rfloor$$
$$+\left\lfloor f_p(\mathbf{r})\otimes g_{np}(r)\otimes f_{ni}(r)\right\rfloor\otimes\ldots\otimes\left\lfloor f_p(\mathbf{r})\otimes g_{np}(r)\otimes f_{ni}(r)\right\rfloor$$
$$+\left\lfloor f_n(\mathbf{r})\otimes g_{np}(r)\otimes f_{pi}(r)\right\rfloor\otimes\ldots\otimes\left\lfloor f_n(\mathbf{r})\otimes g_{np}(r)\otimes f_{pi}(r)\right\rfloor$$
$$+\left[f_n(\mathbf{r})\otimes g_{nn}(r)\otimes f_{ni}(r)\right]\otimes\ldots\otimes\left[f_n(\mathbf{r})\otimes g_{nn}(r)\otimes f_{ni}(r)\right]$$
$$+\left\lfloor f_p(\mathbf{r})\otimes g_{ep}(r)\otimes f_{ei}(r)\right\rfloor\otimes\ldots\otimes\left\lfloor f_p(\mathbf{r})\otimes g_{ep}(r)\otimes f_{ei}(r)\right\rfloor$$
$$+\left[f_n(\mathbf{r})\otimes g_{en}(r)\otimes f_{ei}(r)\right]\otimes\ldots\otimes\left[f_n(\mathbf{r})\otimes g_{en}(r)\otimes f_{ei}(r)\right]$$
$$+\left[f_e(\mathbf{r})\otimes g_{ee}(r)\otimes f_{ei}(r)\right]\otimes\ldots\otimes\left[f_e(\mathbf{r})\otimes g_{ee}(r)\otimes f_{ei}(r)\right]$$
$$+\left\lfloor f_e(\mathbf{r})\otimes g_{ep}(r)\otimes f_{pi}(r)\right\rfloor\otimes\ldots\otimes\left\lfloor f_e(\mathbf{r})\otimes g_{ep}(r)\otimes f_{pi}(r)\right\rfloor$$
$$+\left[f_e(\mathbf{r})\otimes g_{en}(r)\otimes f_{ni}(r)\right]\otimes\ldots\otimes\left[f_e(\mathbf{r})\otimes g_{en}(r)\otimes f_{ni}(r)\right]\Big\} \tag{38}$$

where each convolution is repeated j -times.

$$
\begin{aligned}
V(\mathbf{K}) = \sum_{j=1}^{H} c_j \Bigg\{ & \left[\frac{4\pi}{K^2} F_c(\mathbf{K}) F_{ci}(K) \right]^j + \left[F_p(\mathbf{K}) G_{pp}(K) F_{pi}(K) \right]^j \\
& + \left[F_p(\mathbf{K}) G_{np}(K) F_{ni}(K) \right]^j + \left[F_n(\mathbf{K}) G_{np}(K) F_{pi}(K) \right]^j \\
& + \left[F_n(\mathbf{K}) G_{nn}(K) F_{ni}(K) \right]^j + \left[F_p(\mathbf{K}) G_{ep}(K) F_{ei}(K) \right]^j \\
& + \left[F_n(\mathbf{K}) G_{en}(K) F_{ei}(K) \right]^j + \left[F_e(\mathbf{K}) G_{ee}(K) F_{ei}(K) \right]^j \\
& + \left[F_e(\mathbf{K}) G_{ep}(K) F_{pi}(K) \right]^j + \left[F_e(\mathbf{K}) G_{en}(K) F_{ni}(K) \right]^j \Bigg\}
\end{aligned}
\tag{39}
$$

Use Sect. 2 and (35), (37) and (39). For all available real centrosymmetric electron, proton, neutron, nuclei, including isotopes, we can obtain a consistent set of real (electron and proton) charge distribution, electron, proton and neutron mass distributions, and potential energy functions between each pair of electron, proton and neutron, and the c_j's. The incident radiation is electromagnetic wave. The incident particles are electron, proton, neutron, light and heavy nuclei, including isotopes. The target objects are proton, neutron, and nuclei. The target nuclei can be fixed, freely rotating or rotating about any number of axes. The incident particles and target objects are large in number. Pairing the incident radiation or particles with the target objects, the number of combinations is very large. If the number of c_j used is small, the system is excessively over-determined. A consistent set can be determined. The fitting shows whether it is satisfactory or not. The consistent set obtained is $f_c(\mathbf{r})$, $f_e(\mathbf{r})$, $f_p(\mathbf{r})$, $f_n(\mathbf{r})$ of electron, proton, neutron and all nuclei, where appropriate, and $g_{ee}(r)$, $g_{ep}(r)$, $g_{en}(r)$, $g_{pp}(r)$, $g_{np}(r)$ and $g_{nn}(r)$, and c_2, c_3,….

Shapes and dimensions of the charge distributions and mass distributions, and nuclear form factors are determined.

An alternative way is to use $V_{cal}(\mathbf{K}) - V(\mathbf{K})$ where $V_{cal}(\mathbf{K})$ is the result obtained in Sect. 3.2.4. With $c_1 = 0$, (34) to (39) are then used in the fitting to this difference. A consistent set is obtained.

If better results can be obtained, different c_j can be used for different potential energy terms.

4. Verification

The charge and mass distributions and potential energy functions obtained can be verified by comparing with the experimental and theoretical results and by applying to study various properties of the nuclei, such as self energies, binding energies, the isomeric nuclei, stability, decay process and the nuclear reactions. In these investigations, the potential energy surface and the shape, height and thickness of the potential barrier, which depend on direction, are very useful.

4.1. Self energy due to masses

The self energy of a neutron due to mass,

$$U_{mn} = \frac{1}{2}\iint f_n(\mathbf{r}) g_{nn}(|\mathbf{r}-\mathbf{r}'|) f_n(\mathbf{r}') d\mathbf{r} d\mathbf{r}'$$

Here and in the following, the two volume integrals are over the finite volume of the object, in this case, a neutron, and we expect that all such integrals converge as they should be physically.

For a nucleus X, the self energy due to mass,

$$U_{mX} = \frac{1}{2}\iint f_p(\mathbf{r}) g_{pp}(|\mathbf{r}-\mathbf{r}'|) f_p(\mathbf{r}') d\mathbf{r} d\mathbf{r}' + \iint f_p(\mathbf{r}) g_{np}(|\mathbf{r}-\mathbf{r}'|) f_n(\mathbf{r}') d\mathbf{r} d\mathbf{r}'$$

$$+\frac{1}{2}\iint f_n(\mathbf{r})g_{nn}(|\mathbf{r}-\mathbf{r}'|)f_n(\mathbf{r}')d\mathbf{r}d\mathbf{r}'+\iint f_p(\mathbf{r})g_{ep}(|\mathbf{r}-\mathbf{r}'|)f_e(\mathbf{r}')d\mathbf{r}d\mathbf{r}'$$

$$+\iint f_n(\mathbf{r})g_{en}(|\mathbf{r}-\mathbf{r}'|)f_e(\mathbf{r}')d\mathbf{r}d\mathbf{r}'+\frac{1}{2}\iint f_e(\mathbf{r})g_{ee}(|\mathbf{r}-\mathbf{r}'|)f_e(\mathbf{r}')d\mathbf{r}d\mathbf{r}'$$

Denote the self energy due to mass of a proton by U_{mp}, an electron by U_{me}.

4.2. Self energies of neutron, nuclei, proton and electron

Self energy of a neutron $U_n = U_{mn} + m_n c^2$

m_n is the rest mass of a neutron.

For a nucleus X, the self-Coulomb-energy due to charge distribution**,**

$$U_{cX} = \frac{1}{2}\iint f_c(\mathbf{r})\frac{1}{|\mathbf{r}-\mathbf{r}'|}f_c(\mathbf{r}')d\mathbf{r}d\mathbf{r}'$$

Self energy of nucleus X, $U_X = U_{cX} + U_{mX} + m_X c^2$ (40)

m_X is the rest mass of nucleus X.

Self energy U_p of a proton is given by (40), $U_p = U_{cp} + U_{mp} + m_p c^2$

m_p is the rest mass of a proton.

Denote the self-Coulomb-energy of an electron by U_{ce}.

Self energy of an electron $U_e = U_{ce} + U_{me} + m_e c^2$

m_e is the rest mass of an electron.

If we just treat an electron as a point particle, U_{ce} and U_{me} are not calculated. From the experimental result of U_e, $(U_{ce} + U_{me})$ can be determined.

Assume that U_{ce}, U_{me} and U_e of the positron are equal to those of the electron.

4.3. Binding energy of nucleus

For nucleus X with N neutrons and Z protons, the binding energy $= NU_n + ZU_p - U_X$

Binding energy per nucleon can be calculated. In the literature, the binding energy and the energy release, Q of the reactions are defined as differences of masses only.

4.4. Separation energy

The separation energy of separating a neutron from a nucleus $= U\left({}^{A-1}_{Z}X^{N-1}\right) + U_n - U\left({}^{A}_{Z}X^{N}\right)$

The separating energy of separating a proton from a nucleus $= U\left({}^{A-1}_{Z-1}Y^{N}\right) + U_p - U\left({}^{A}_{Z}X^{N}\right)$

4.5. Alpha decay

Energy release in alpha decay, $Q = U\left({}^{A}_{Z}X^{N}\right) - U\left({}^{A-4}_{Z-2}Y^{N-2}\right) - U_\alpha$

where U_α is the self energy of an alpha particle.
If the energy release Q is negative, the parent nucleus is α-stable.

In the alpha decay theory of Gamow, Gurney and Condon, an α-particle is formed inside the parent nucleus. It moves, bouncing back and forth, in the field of the daughter nucleus. Then tunnels through the potential barrier by means of quantum tunneling. If this model is true, we expect the structure of the parent nucleus resembles the structure of the daughter nucleus plus an average of charge and mass of an α-particle. This will be shown in the result obtained in Sect. 2. The nuclear structure affects the formation of the α -particle. The potential barrier which can be determined from the consistent set in Sect. 3.2.5, including, shape, height, width and direction dependence will be very useful in the studies of α -decay theory. This is related to the half-life of the decaying process.

Along the same line, proton emission and the possibility of two-proton emission can also be investigated.

4.6. Beta decay

For negative β -decay, energy release, $Q = U\left({}^{A}_{Z}X^{N}\right) - U\left({}^{A}_{Z+1}Y^{N-1}\right) - U_e$

Assume that the masses of neutrino and anti-neutrino are negligible.

For positive β -decay, energy release, $Q = U\left({}^{A}_{Z}X^{N}\right) - U\left({}^{A}_{Z-1}Y^{N+1}\right) - U_e$

For electron capture, energy release, $Q = U\left({}^{A}_{Z}X^{N}\right) + U_e - U\left({}^{A}_{Z-1}Y^{N+1}\right) - b_e$

where b_e is the binding energy of the electron captured.

For double β -decay, energy release, $Q = U\left({}^{A}_{Z}X^{N}\right) - U\left({}^{A}_{Z+2}Y^{N-2}\right) - 2U_e$

The nucleus of the single β -decay product of the parent nucleus forms the energy barrier of the double β -decay.

If the energy release Q is negative, the process is not energetically favorable. The parent nucleus is β -stable. If nuclear electron does exist, it helps studies of β -decay theory.

4.7. Electric multipoles

From the charge distribution of the nucleus determined in Sect. 2, the electric multipole moments, for example, dipole and quadrupole can be calculated. If the electrical dipole moment is zero, it will be shown in the result.

4.8. Isomeric nuclei

Nucleus with a given neutron number N and proton number Z may form different structures, isomers. A special case is that two nuclei which are mirror images of each other. From the neutrons and protons (charge distribution, mass distributions, and potential energy functions between masses) determined in Sect. 2, calculate the potential energy surface for each configuration of neutrons and protons. Find the local and global energy minima and the energy barrier between them. These give the possible structures of the nucleus and their statistical probability of occurrence. A local energy minimum is an excited state of the nucleus. The transition from one local minimum to another proceeds by means of γ-decay or internal conversion.

4.9. Stability

Stability and the decay products of each nucleus can be studied.

4.10. Nuclear reaction.

The self energies of the reactants and products are used to investigate nuclear reactions. In a reaction, compound nucleus may be formed. When a low energy, positively charged particle is incident on a charged nucleus, it has high probability to be elastically scattered. At higher energy, the incident particle has higher probability to tunnel through the Coulomb barrier and enters the nucleus. A charged particle inside a charged nucleus can tunnel through the barrier and is emitted, for example, α-decay. The potential barrier including shape, height, width and direction dependence determined in Sect. 2 can be used in the calculation of reaction cross sections, excitation functions, and their angular dependence. The probabilities of occurrence of various reactions as a function of kinetic energy of the incident nucleus can also be calculated.

4.11. Fission reaction

In fission reaction, a heavy nucleus separates into two fragments of similar masses. The Coulomb barrier for such fission can be calculated. During the fission process, the structure and shape of the nucleus change. Hence the shape, height and width of the Coulomb barrier also change. $X \rightarrow W + Y$

The energy release, $Q = U_X - (U_W + U_Y)$

If Q is positive, the fission is spontaneous.
If Q is higher than the Coulomb barrier, the life-time of the spontaneous fission is very short.
If Q is lower than the Coulomb barrier, tunneling occurs.

Fission can be induced by absorption of an incident particle.

4.12. Fusion reaction

Two light nuclei can combine to form a heavier nucleus with a release in energy, a fusion reaction. The shape, height and width of the Coulomb barrier can be calculated.

4.13. Superheavy elements and other unknown elements

For possible existence of superheavy elements, the proton and neutron obtained in Sect. 2 can be used to build up these elements and calculate the self energy for each configuration. Local and global minima will show the possible existence of these elements. In fact, a systematic approach can be used to investigate the stability, decay energy and potential energy barrier of nuclei within a range of atomic number and mass number. This reveals possible existence of other unknown elements.

5. Refinement

5.1. General functions of charge and mass distributions and nuclear forces

If the fitting to $V(\mathbf{K})$ in Sect. 3.2.5, and the verification in Sect. 4 are not satisfactory, we may refine the treatment by allowing $v(\mathbf{r})$ to depend on the charge and mass distributions and nuclear forces separately. General functions of charge and mass distributions and nuclear forces are employed. Add to (34), (36) and (38), where appropriate, terms like

$$\sum_{j=1}^{L}\alpha_j\big[f_c(\mathbf{r})\otimes\ldots\otimes f_c(\mathbf{r})+f_p(\mathbf{r})\otimes\ldots\otimes f_p(\mathbf{r})+f_n(\mathbf{r})\otimes\ldots\otimes f_n(\mathbf{r})$$

$$f_e(\mathbf{r})\otimes\ldots\otimes f_e(\mathbf{r})\big]$$

$$+\sum_{j=1}^{N}\beta_j\big[g_{pp}(r)\otimes\ldots\otimes g_{pp}(r)+g_{np}(r)\otimes\ldots\otimes g_{np}(r)+g_{nn}(r)\otimes\ldots\otimes g_{nn}(r)$$

$$+g_{ep}(r)\otimes\ldots\otimes g_{ep}(r)+g_{en}(r)\otimes\ldots\otimes g_{en}(r)+g_{ee}(r)\otimes\ldots\otimes g_{ee}(r)\big]$$

where $\alpha_1=\beta_1=1$. L, N and α_2, $\alpha_3,\ldots$, β_2, $\beta_3,\ldots$ are to be determined.

Each convolution is repeated j-times.

Add to (35), (37) and (39), where appropriate, terms like

$$\sum_{j=1}^{L}\alpha_j\Big\{[F_c(\mathbf{K})]^j+[F_p(\mathbf{K})]^j+[F_n(\mathbf{K})]^j+[F_e(\mathbf{K})]^j\Big\}$$

$$+\sum_{j=1}^{N}\beta_j\Big\{[G_{pp}(K)]^j+[G_{np}(K)]^j+[G_{nn}(K)]^j$$

$$+[G_{ep}(K)]^j+[G_{en}(K)]^j+[G_{ee}(K)]^j\Big\}$$

All the parameters are adjusted to fit to $V(\mathbf{K})$. An alternative way is to use the calculated $V(\mathbf{K})$ obtained in Sect. 3.2.5, and adjust the newly introduced parameters α_j 's and β_j 's to fit to the difference . In either way, a consistent set is obtained. Verification as in Sect. 4 is again applied.

If better results can be obtained, different α_j can be used for different charge-and mass-distribution terms, and different β_j for different nuclear-force terms.

5.2 Correlation between potential energies, charge and mass distributions and nuclear forces

If the result in Sect. 5.1 is not satisfactory, correlation between the potential energies, charge and mass distributions and nuclear forces may be included. For example, add to equations in Sect. 5.1 terms like

$$\gamma\lfloor f_p(\mathbf{r})\otimes g_{pp}(r)\otimes f_{pi}(r)\rfloor\otimes\ldots\otimes\lfloor f_p(\mathbf{r})\otimes g_{pp}(r)\otimes f_{pi}(r)\rfloor\otimes$$

$$\lfloor f_n(\mathbf{r})\otimes g_{np}(r)\otimes f_{pi}(r)\rfloor\otimes\ldots\otimes\lfloor f_n(\mathbf{r})\otimes g_{np}(r)\otimes f_{pi}(r)\rfloor\otimes$$

$$[f_c(\mathbf{r})\otimes\ldots\otimes f_c(\mathbf{r})]\otimes[g_{en}(r)\otimes\ldots\otimes g_{en}(r)]$$

where the convolution in the first line is repeated h-times, the convolution in the second line, j-times, in the first square brackets in the third line, l-times, and in the last square brackets, m-times,

γ, h, j, l, m are to be determined in fitting,

and its Fourier transform

$$\gamma\left[F_p(\mathbf{K})G_{pp}(K)F_{pi}(K)\right]^h\left[F_n(\mathbf{K})G_{np}(K)F_{pi}(K)\right]^j\left[F_c(\mathbf{K})\right]^l\left[G_{en}(K)\right]^m$$

Adjust all the parameters to fit to $V(\mathbf{K})$. This amounts to expressing $v(\mathbf{r})$ as general functions of the potential energies, the charge and mass distributions and nuclear forces. The coefficients of the power series for $V(\mathbf{K})$ are obtained by fitting. An alternative way is to adjust the newly introduced parameters to fit to the difference. In either way, a consistent set is obtained. Verification as in Sect. 4 is applied.

5.3. Assumption (b) relaxed

Analogous to Thomson scattering and Rayleigh scattering, instead of (2), we may employing an expression

$$V(\mathbf{K}) = \int (\eta_0 + \eta_1 k_0 + \eta_2 k_0{}^2 + ...) v(\mathbf{r}) \exp(-i\mathbf{K} \cdot \mathbf{r}) d\mathbf{r} \tag{41}$$

$|V(\mathbf{K})|$ is measured experimentally. For a set of η_0, η_1,…, the phase of $V(\mathbf{K})$ is determined from $|V(\mathbf{K})|$ for all $\mathbf{K}$ with $K \le 2k_0$. $v(\mathbf{r})$ can then be obtained. η_0, η_1,… are parameters to be determined in the fitting. A consistent set is obtained. Verification as in Sect. 4 is applied.

5.4. Assumption (a) relaxed

In the scattering of electromagnetic wave in Sect. 2, the charge is assumed to be free. But in the nucleus, the proton charge is bounded to the proton mass, and the motion of a proton is not free. Instead of (1), it may be more appropriate to use an expression

$$F_c(\mathbf{K}) = \int (\xi_0 + \xi_1 k_0 + \xi_2 k_0{}^2 + ...) f_c(\mathbf{r}) \exp(-i\mathbf{K} \cdot \mathbf{r}) d\mathbf{r} \tag{42}$$

When $\xi_0 = 1$, $\xi_1 = \xi_2 = ... = 0$, (42) reduces to (1) for Thomson scattering for free electron. When $\xi_0 = \xi_1 = \xi_3 = \xi_4 = ... = 0$, $\xi_2 = 1$, (42) reduces to the case of Rayleigh scattering for bounded electron.

$|F_c(\mathbf{K})|$ is measured experimentally. For a set of ξ_0, ξ_1,…, the phase of $F_c(\mathbf{K})$ is determined from $|F_c(\mathbf{K})|$ for all $\mathbf{K}$ with $K \le 2k_0$. $f_c(\mathbf{r})$ can then be obtained. ξ_0, ξ_1,… are parameters to be determined in the fitting. A consistent set is obtained. Verification as in Sect. 4 is applied.

The proton charge is bounded to the proton mass, and the electron charge in the nucleus is bounded to the electron mass. In allocating $f_c(\mathbf{r})$ into charges associated with the protons and electrons, we may find that assumption (e) holds and, similarly, mass distribution of electron is proportional to charge distribution of electron.

6. Determination of nuclear size (shape and dimensions) and surface distribution

In (1) and (2), if we use $F_c(\mathbf{K})$ and $V(\mathbf{K})$ with $K \le K_{\max}$ and $K_{\max}$ is small, the incident radiation or particle has low energy. It is not very penetrating. Only the surface or surface layer is investigated. The target object is mainly a surface or a membrane. Following Sect. 2, 3 and 5, the size (shape and dimensions) of the charge distribution, electron, proton and neutron mass distributions of the nuclear surface can be determined. A consistent set pertaining to the surface layer can be obtained.

A special case is a freely rotating nucleus. Its diameter will be determined.

7. Direct determination of the diameter and shape (and dimensions) of the convex hull (envelope) of a nucleus by means of the minimum-phase function

If we intend to determine only the diameter or the shape (and dimensions) of the convex hull of a nucleus, and not its internal structure, a direct and simple method is to use the minimum-phase function ([54], [55]). By means of this method, the shape (and dimensions) of the surface of revolution of a nucleus spinning about an axis can be determined.

8. Application of methods in X-ray crystallography to determination of nuclear structure

Use a cubic unit cell to contain the nucleus. Mathematically add replicas of the unit cell successively to give a three-dimensional crystal. $|$Fourier coefficients$|$ and in-between $|$Fourier coefficients$|$ are obtained from the experimentally measured diffraction intensity. Various methods have been developed in structure determination in X-ray crystallography, such as sharpening of Patterson function, direct methods, point-atom model, accelerated convergence of extension of Bragg intensities and structure factors, and sharpening in reciprocal space. These powerful methods can be applied to determination of nuclear structure ([56]).

For example, if we use a unit cell with edge length = 1Å. The large empty space outside the small nucleus provides a strong support for the iterative algorithms with oversampling, like solvent flattening in [57], [58].

9. Discussion and conclusion

Determination of nuclear charge distribution by means of electromagnetic-wave scattering is affected by the atomic electrons. If, at present, it is easier to determine the nuclear charge distribution by electron scattering, Sect. 2 can be disregarded. Electron scattering is used to determine nuclear charge distribution. Here electron is regarded as a point particle and the nuclear force between the incident electron and the nucleus is negligible. $f_c(\mathbf{r})$ is obtained. It is used as initial value in obtaining a consistent set which includes $f_c(\mathbf{r})$ as value to be determined.

In the past, many data have been collected for electron scattering. The nuclear charge distribution can be determined immediately. Combining with the many data for neutron, proton and nucleus scattering, a gross consistent set of mass distributions and potential energy functions can be obtained immediately (Sect. 3.2.1). This can be compared with experimental and theoretical results. Then proceed to Sect. 3.2.2, so on.

In Sect. 5.4, a fictitious object $v(\mathbf{r})$ is obtained from the scattered intensity. The scattered intensity has been transformed to $v(\mathbf{r})$ which may be treated as fundamental data. Instead of requiring theories and models to reproduce the scattered intensity, they may be required to reproduce this $v(\mathbf{r})$. This is more direct, has clearer physical meaning, easier to visualize and perform, saves much computation, and has deeper understanding of the physics involved.

The phase-retrieval problem is solved by employing finite size of $v(\mathbf{r})$. This in turn requires that the mass distribution of the nucleus $f_m(\mathbf{r})$ has finite size. It may be true that a nucleus does have finite size, or it is just due to experimental limitation. The result should be able to show which is true. Section 6 presents an accurate method of determining nuclear size (shape and dimensions). This can be used in the application of experimental electric and magnetic multipoles in the investigation of nuclear structure. For the solution of the phase- retrieval problem, the constraints employed do not include continuity of the object. It is very interesting to see whether a nucleus has sharp boundary or taper to zero.

The consistent set obtained in Sect. 3.2.5 or Sect. 5.1, 5.2, 5.3, 5.4 includes the charge distribution and electron, proton and neutron mass distributions, and nuclear forces. These are empirical functions. They can be compared with the experimental or theoretical results. For a freely rotating centrosymmetric or non-centrosymmetric nucleus, the charge or mass distribution obtained is the spherical average. Models and theories of the nucleus can be used to fit to this spherical average. As mentioned in above, such fitting is more direct, has clearer physical meaning and easier to perform. The potential energy functions $g_{pp}(r)$, $g_{np}(r)$,… can be decomposed into different components according to theory. These empirical functions help the investigation of the law governing the charge and mass distributions and nuclear forces.

In (2), all $\mathbf{K}$ with K up to infinity are used. Hence incident particle with very high energy is used. The first Born approximation may be applicable. In practice, we use $|V(\mathbf{K})|$ with $K \le K_{\max}$. If we use $|V(\mathbf{K})|$ with $K_{\max}$ up to infinity, we obtain $v(\mathbf{r})$ with ultimate resolution. But this may not be needed. In the computation, we can gradually increase $K_{\max}$ and examine the results. A consistent set pertaining to the surface layer can be determined (Sect. 6). $K_{\max}$ is further increased until the desired resolution is reached, and the first Born approximation is applicable.

The validity of assumptions (f), (e), (d), (c), (b) and (a) can be examined. They are disregarded or relaxed successively. The individual effect can be examined. The fitting to $F_c(\mathbf{K})$ and $V(\mathbf{K})$, the comparison of the consistent set with the experimental and theoretical results, and the verification of all other experimental results in Sect. 4, show whether the assumptions are valid or not. In fact, these are the only criteria for determining the correctness of a phase-retrieval method.

In Sect. 5.4, general functions of potential energy, charge and mass distributions and two-body central nuclear forces are used. If the fitting to $V(\mathbf{K})$ or the verification in Sect. 4 is not satisfactory, the discrepancy may be due to factors such as many-body forces which have not been included. The fitting shows whether systematic discrepancy exists. Since the result obtained is quantitative. This helps in finding the factors neglected.

In the Introduction there are arguments: "It is very difficult to believe that there are two different objects, other than mirror images, which show the same scattered intensity in all directions, for incident electromagnetic wave of all wavelengths in all directions. Hence the object should be uniquely determined by all the scattered intensities," and "For nucleus with incident electromagnetic wave of all wavelengths, and incident particles of all species with all velocities, in all directions, it is more difficult to believe that there are two different nuclei, other than mirror images, which have the same scattered intensity in all directions, and are compatible to all other experimental data. Hence the structures of the incident particle and the target nucleus, and their interactions should be uniquely determined by all the scattered intensities and all other experimental data. This means that the phase of the scattered wave is uniquely determined by the magnitude the phase of the scattered wave is uniquely determined by the magnitude of the scattering amplitude and all other experimental data." When the first Born approximation is applicable, a solution is presented in Sect. 2 and Sect. 3.2.4 for real centrosymmetric nuclei. When the first Born approximation is not applicable, it is still plausible that the phase of the scattered wave is determined by the magnitude. Instead of the empirical approach in Sect. 3.2.5 and Sect. 5, a direct way is to use a scattering theory and solve the mathematical problem: What are the general and reasonable constraints of all target nuclei, under which the phase of the scattered waves of all incident waves and particles can be obtained from the magnitude? The result obtained should be consistent with the result in Sect. 2 and Sect. 3.2.4 when the first Born approximation is applicable.

In the present article, a classical approach is adopted. Except the wave nature of the incident particle is employed, no quantum theory including Schrödinger equation is used. One of the reasons is that a purpose of this article is an illustration for the phase-retrieval approach. The classical method has the advantage of simplicity and many quantitative results can be obtained. A very simple and clear picture can be presented, as shown in the article. Assumptions encountered in the literature

(d) charge independence -- the potential energy functions between unit masses of different pairs of electron, proton and neutron are the same

(e) mass distribution of protons $f_p(\mathbf{r})$ / charge distribution of protons $f_c(\mathbf{r})$

= mass of a proton / charge of a proton, for all $\mathbf{r}$

(f) mass distribution of neutron $f_n(r)$ / mass distribution of proton $f_p(r)$

= number of neutrons/number of protons, for all r

can be examined. Nuclear electron can be studied. Empirical functions for the charge and mass distributions and nuclear forces can be obtained. From these, we can calculate numerical values of many nuclear properties such as self energies, binding energies, Q of reactions, electric multipoles, etc.

Another reason is that academically, it is meaningful to increase the value of $K_{\max}$ gradually and investigate the range of K where the classical method is applicable. Then researchers can incorporate the phase-retrieval approach to the quantum theory of scattering. Researchers may apply the phase-retrieval method in quantum theory of scattering to all centrosymmetric real nuclei subjected to the constraints of $v(\mathbf{r})$, (i), (ii), (iii) and (iv) in Sect. 3.1.

It is reasonable to obtain empirical functions from experimental data. The basis of the validity of any theory is its agreement with experimental results, and it should be able to reproduce these empirical functions.

This article presents an approach for determining nuclear structure and nuclear force in all centrosymmetric real nuclei (for example, freely rotating), including isotopes, by means of phase retrieval in scattering. The incident radiations are electromagnetic wave and electron, proton, neutron and other nuclei. All the scattered intensity is transformed to a fictitious object which may be used as fundamental data for studies of nuclear physics. A consistent set of the nuclear structures, including shapes and dimensions, and two-body central nuclear forces may be obtained. They can be verified by applying to many aspects of nuclear physics, such as self energies due to mass and charge, binding energy, potential energy surface, isomeric nuclei, decay, nuclear reaction, etc.

If we intend to determine only the diameter or the shape (and dimensions) of the convex hull of a nucleus, and not its internal structure, a direct and simple method is to use the minimum-phase function (Sect. 7). By means of this method, the shape (and dimensions) of the surface of revolution of a nucleus spinning about an axis can be determined.

Various powerful methods have been developed in structure determination in X-ray crystallography. These methods can be used in the determination of nuclear structure (Sect. 8). The nuclear structure is not as complicate as the structure of macromolecular crystal. For example, the Patterson function will have less overlap. The methods in X-ray crystallography may be very useful in nuclear structure determination.

One of the purposes of this article is to stimulate interest in the phase-retrieval approach. An illustration is provided and it shows that phase retrieval in investigating nuclear physics is a possibility. Determining nuclear structure and nuclear force is very important and very complicate. Every approach is invaluable and may be attempted. The phase-retrieval approach is direct, efficient and quantitative.

The present article deals with centrosymmetric real nuclei. How to apply phase retrieval approach to non-centrosymmetric nuclei and complex nuclei which are employed in optical model ([2])?

Determining nuclear structure and nuclear force by means of phase retrieval in scattering is a wide open area for research. Much work can be done. There will be many theoretical and experimental difficulties; much research is needed. Phase retrieval is another entrance to the mystery of nuclear physics.

The problem discussed in above is an active problem; there is an incident radiation or particle. For the passive problem such as α-, β- and γ-decay, the phase-retrieval approach is applicable. See, for example, Ref. [59]. From the intensity emitted, the phase can be found. The solution of the phase-retrieval method is the decay-source structure inside the nucleus. Such approach is useful in the studies of astro-nuclear physics. Similarly for nuclear reactions (inelastic reaction and others), the phase-retrieval approach is applicable. From the intensities of the product particles or radiations, the phases can be found, and these give the reaction-source structure inside the nucleus. Considering the large number of nuclear reactions, this is a vast area for investigation.

Experimentally, for scattering of electromagnetic wave or particle by a nucleus, a collection of these nuclei, for example, gas or a crystal, can be used. The wavelength of the incident wave is of the order of nuclear size, and much less than the internuclear distance. For a gas of N nuclei, the scattering is incoherent. The diffraction intensity is N times that of one nucleus.

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Index

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