Theory of relaxation and decoherence of intersublevel transitions in semiconductor quantum dots

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Abstract. We present calculations of intersublevel population and phase relaxation in semiconductor quantum dots. Anharmonic polaron decay is shown to be responsible for population relaxation over a large spectral range. Additional pure dephasing processes are caused by acoustic phonons, which cause the appearance of peaked sidebands and induce broadening through real and virtual transitions between the excited states.

1. Introduction

Energy relaxation in quasi zero-dimensional systems has been the subject of debates for nearly two decades. It was first pointed out that a phonon bottleneck should occur in these nanostructures [1, 2, 3]: the emission of acoustic phonon should be strongly inhibited as soon as the energy separation between the levels is higher than a few meV, while LO-phonon emission should not be possible unless the intersublevel energy separation match the LO-phonon energy.

Later, it was found that a strong coupling regime occurs between confined electronic states and LO-phonon [4, 5, 6]. This is due to the fact that the Fröhlich matrix elements (of the order of a few meV) are much larger than both the electronic linewidth (discussed below) and the dispersion of long-wavelength LO-phonon involved (typically 0.1meV). A striking consequence is the observation of anticrossing when two confined states (electron, hole, or exciton) have energies differing by a multiple of LO-phonon energy [5, 6, 7, 8, 9, 10]. The eigenstates are quasiparticles called polarons. This polaronic nature of the intersublevel transitions have important consequences for energy relaxation.

Recently we have proposed a model for energy relaxation in semiconductor quantum dots (QDs) that rely on a microscopic description of polaron anharmonicity [11]. This approach results in dramatic differences compared to previous established models [12, 13]. We show that this mechanism is responsible for energy relaxation in QDs in a large spectral range, in good agreement with pump-probe measurements [14, 15, 16, 17].

We also study the coherence of such QDs intersublevel transitions. At low temperature, dephasing is limited by the anharmonic polaron decay to the ground state ($T_2 = 2T_1$), while at higher temperature additional mechanisms of dephasing appear. We show below that acoustic phonons induce (i) peaked acoustic phonon sidebands and (ii) homogeneous broadening of the intersublevel transition due to both real and virtual transitions towards the second excited p-state.
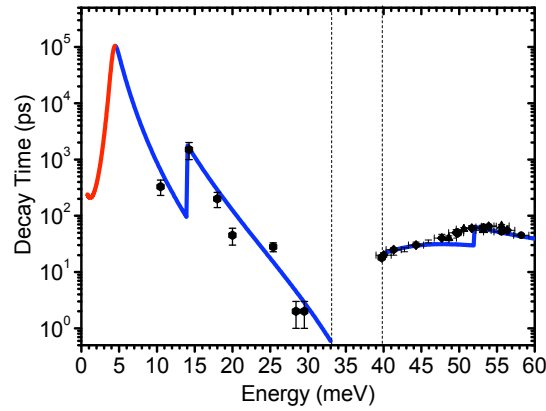
Figure 1. Intersublevel relaxation time as a function of the transition energy: Calculation in solid line and measurements from the references [15] and [17] in symbols. The transition energy on the abscissa corresponds to the energy difference between the ground and excited polaron state with dominant electronic component. The blue line corresponds to the domain where the relaxation is dominated by anharmonic relaxation decay while the red one corresponds to acoustic phonon emission. The dashed lines indicate the polaron splitting at the LO-phonon resonance.

2. Energy relaxation by anharmonic polaron decay

It was first pointed out by Li and co-workers [12] that Fröhlich couplings combined to LO-phonon instability allow electronic relaxation. In their model, a constant phonon damping was added phenomenologically in the coupled equation of the electron-phonon system. This semi-classical model was used in order to explain the lifetime increases for energies comprise between 40 and 50 meV [14, 15] but failed in explaining behaviour at higher energies [15]. More recently, in [11], we have presented a microscopic model of anharmonicity induced relaxation. We have shown that this approach is mandatory: the efficiency of anharmonic processes varies strongly with the energy to dissipate. These relative variations are of the same order, or even larger that the relative variations of the LO-phonon component with energy. As a consequence, the semi-classical model fails in predicting, even qualitatively, the variations of the intersublevel lifetimes with energy.

The microscopic theory for anharmonic polaron relaxation was presented in [11], and has been recently extended to take into account electronic states far detuned from the LO-phonon resonance, in particular in the THz range [17]. The polaron relaxation rate between two QDs levels reads:

$$\Gamma_i = \frac{4\Delta^2}{(\hbar\omega_{\mathrm{LO}} + \Delta)^2} \frac{V_F^2}{(\Delta - \hbar\omega_{\mathrm{LO}})^2 + V_F^2} \Gamma^{ph}(\Delta) \tag{1}$$

where V_F is the Fröhlich matrix element involved, ω_{LO} is the bulk LO-phonon frequency, Δ is the transition energy between polaron states, and the function Γ^{ph} was defined in [11]. $\Gamma^{ph}(\Delta)$ can be seen as the decay rate of a LO-phonon vibration that would be driven at the polaron frequency $\Delta/\hbar$. The first term in the above formula is needed only for transitions well detuned from the resonance $\hbar\omega_{\mathrm{LO}}$, and accounts for interferences between resonant and non-resonant Fröhlich interactions. The second term corresponds to the weight of the LO-phonon component in the excited polaron (if the polaron wavefunction is of the form $|\phi_i\rangle = \alpha_i|p, 0\rangle + \beta_i|s, 1\rangle$, where 0 and 1 refer to LO-phonon occupation numbers, this quantity reads $|\beta_i|^2$).

The above formula can be split into two contributions: the first two terms describe the variations of the polaronic nature of the excitation while the term Γ^{ph} account for the variations in the efficiency of anharmonic mechanism. As shown in [11], $\Gamma^{ph}(E)$ is made of a sum of different anharmonic channels,
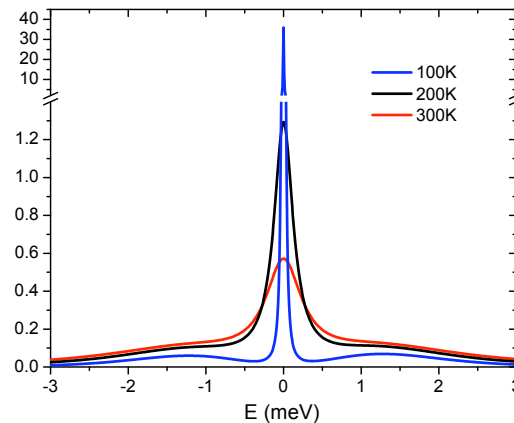
Figure 2. Intersublevel absorption from s to first p excited state of a QD for different temperatures (100K, 200K, 300K) assuming a splitting of 5 meV between the p-states. The absorption exhibits zero-phonon lines (ZPL) and acoustic phonon sidebands on both sides of the ZPL.

which consist of two-phonon combinations in the cubic approximation for crystal anharmonicity. Each of these channels efficiency shows dramatic variations with energy. As a consequence, one cannot approximate Γ^{ph} by a constant Γ_{LO} corresponding to LO-phonon relaxation in bulk. In the semiclassical model, the polaron relaxation rate was governed by the sole LO-phonon component $|\beta_i|^2$ and thus its behaviour was expected to be symmetric with respect to the LO-phonon resonance. Here, in strong contrast, a dramatic increase of intersublevel times is found below the LO-phonon energy when reducing the transition energy [17].

3. Acoustic phonon induced decoherence
Beyond the knowledge of population relaxation mechanism, it is important to know if there is additional dephasing processes. For example, Sherwin and co-workers proposed that intersublevel transitions in QDs in the terahertz range could be used a quantum bit [18]. Homogeneous linewidth are also important for optoelectronic applications such as polaron laser proposed by Sauvage and Boucaud [19].

Acoustic phonon plays a role not only by providing a population relaxation mechanism at low transition energy, but also by inducing pure dephasing processes. As shown in [20] and [21], acoustic phonon brings two kinds of contributions to the dephasing of intersublevel transitions. The diagonal part of the acoustic phonon-electron interaction is responsible for acoustic phonon sidebands. On the other hand, non-diagonal elements can induce real or virtual transitions between the excited levels.

The calculated absorption of a single quantum dot is presented on figure 2. In this calculation, the transition energy is 55 meV, and a splitting of 5 meV is assumed between the p-states. In the lineshape calculation, three mechanisms are taken into account: (i) anharmonic polaron decay, (ii) acoustic phonon sidebands, (iii) acoustic phonon induced real and virtual transitions towards the higher p-states.

For interband transitions, it was already shown that acoustic phonon causes the appearance of sidebands. However, in the latter case, these sidebands consist at non-zero temperature of a broad pedestal centered near the zero-phonon line (ZPL) [22, 23]. In the time-domain, these sidebands are responsible for a partial decay of coherence on the ps time scale [24, 25]. In contrast, for intersublevel transitions, these sidebands are instead peaked on both sides of the transition (see figure 2), which results in an oscillatory behaviour in photon echo experiments [20].

The ZPL broadening with temperature is essentially attributed to real and virtual transitions between the p-states. Acoustic phonons can induce efficient one-phonon absorption or emission only if the energy separations between the p-levels is of the order of a few meV (with a maximum of efficiency for typically

1-2 meV). Hence it is crucial to take also into account two phonon processes. Note that virtual transitions have also been proposed in order to explain broadening of the ZPL for interband transitions [26, 27]. In [20] and [21], we have calculated real and virtual transitions, taking into account up to two-phonon processes in a non-perturbative manner. A very good agreement between calculations and experiments is obtained. In particular, virtual transitions plays a dominant role in decoherence at high temperature.

4. Conclusion

We have presented theoretical investigations of intersublevel population and phase relaxation in QDs. Population relaxation is due to anharmonic polaron decay in a large spectral range, and a microscopic model of crystal anharmonicity is mandatory in order to calculate relaxation times. Additional dephasing processes are caused by interaction with acoustic phonon, resulting in the appearance of acoustic phonon sidebands and broadening of the ZPL due to real and virtual transitions.

Acknowledgments

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