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Polaronic effects in semiconductor quantum dots

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Abstract
We discuss the role of excitonic polarons on the interband optical properties of single self-assembled semiconductor quantum dots. We show in particular that the strong coupling to optical phonons leads to fundamental modifications in the description of the excitation, relaxation and luminescence processes, as compared to previous schemes, and that the use of excitonic polaron states provides a coherent model for the description of various interband optical processes.

1. Introduction

Various experimental and theoretical results demonstrate that electrons confined in quantum dots (QDs) are strongly coupled to the longitudinal optical (LO) vibrations of the underlying semiconductor lattice [1–3]. This coupling leads to the formation of the so-called electronic quantum dot polarons, which are the true excitations of a charged dot. Infrared absorption probes directly the polaron levels instead of the purely electronic ones. Interband transitions are also expected to be related to polaron states involving electron/hole states coupled to optical phonons (excitonic polarons, for brevity [4]). In the following, we review different results related to the coupling of electron/hole pairs with optical phonons, and their consequences on the interband optical response of semiconductor quantum dots [4–6]. We shall focus primarily on the fact that the polaron framework leads to a coherent description of the ensemble of processes governing the interband spectrum of single QDs, namely, the creation, relaxation and luminescence processes, including the appearance of different kinds of replica.

2. Quantum dot polarons eigenstates

Dot excitonic polarons correspond to the actual interband excitations of quantum dots. Let us initially briefly describe these states. To start with, we assume in the following bulk-like monochromatic LO states (energy $\hbar\omega_{\mathrm{LO}}$). We denote by $|n\rangle$ the different electron/hole levels we are interested in, and the bulk lattice states by $|0\rangle$ (lattice ground state), $|1; q\rangle$ (one bulk phonon with wavevector q), $|2; q_1, q_2\rangle$ (two bulk phonons with wavevectors q_1 and q_2), and so on. In the absence of electron/hole–phonon coupling the decoupled dot eigenstates read

$$|n; p, Q_p\rangle = |n\rangle \otimes |p; Q_p\rangle \quad (1)$$

with $p = 0, 1, \ldots$ and $Q_p = (q_1, \ldots, q_p)$ a p-dimensional vector containing p bulk wavevectors. Since we are interested in the low lying dot states at low temperatures, let us disregard any continuum of electron/hole levels. Let us finally denote by $H = H_0 + V_{\mathrm{F}}$ the total Hamiltonian and by $|N\rangle$ the polaron wavefunction,

$$|N\rangle = \sum_{n,p,Q_p} C_N(n, p, Q_p)|n; p, Q_p\rangle \quad (2a)$$

$$H_0|n; p, Q_p\rangle = (e_n + p\hbar\omega_{\mathrm{LO}})|n; p, Q_p\rangle. \quad (2b)$$

V_F is the Fröhlich interaction which couples only states differing by one phonon occupation. The coefficients $C_N(n, p, Q_p)$ incorporate the mixings introduced by the carrier–phonon interactions among the decoupled $|n\rangle \otimes |p; Q_p\rangle$ states. It is worth recalling that when only one electron/hole level is retained, the diagonalization can be done analytically (see e.g. [7, 8]), and it leads to the so-called adiabatic coupling regime. More generally, different electronic levels should be retained in the basis, essentially because the characteristic inter-level spacing is of the same order of magnitude as the optical phonon energy [1–4, 9, 10].

3. Optical properties of single QDs in the polaron framework

An interband optical experiment probes the polaron states $|N\rangle$, instead of the electron/hole states $|n\rangle$. Let us define in the following the optical response function for polaron states of a single QD:

$$\eta_{PO}(\omega_{in}, \omega_{out}) = \sum_{N\geqslant 0} T_{cre}[\hbar\omega_{in}; |N\rangle] T_{rel}[|N\rangle; |G\rangle] T_{lum}[|G\rangle; \hbar\omega_{out}] \quad (3a)$$

$$T_{cre}[\hbar\omega_{in}; |N\rangle] = \sum_{p\geqslant 0} N_{BE}(p\hbar\omega_{ph}) F_{N,p} \delta[\hbar\omega_{in} - E_N + p\hbar\omega_{ph}] \quad (3b)$$

$$T_{lum}[|G\rangle; \hbar\omega_{out}] = \sum_{p\geqslant 0} \{1 + N_{BE}(p\hbar\omega_{ph})\} F_{0,p} \delta[\hbar\omega_{out} - E_0 + p\hbar\omega_{ph}] \quad (3c)$$

where $\hbar\omega_{in}$ and $\hbar\omega_{out}$ are the incoming and outgoing photon energies, N_{BE} the equilibrium phonon population, $\omega_{ph} = \omega_{LO}$, $|N\rangle$ the Nth *polaron* state (with $|G\rangle$ the ground one for $N = 0$) of the QD, E_N its energy and $F_{N,p}$ its oscillator strength for a transition involving the vacuum with p phonons, which is proportional to the square of the dipolar matrix element,

$$\langle p, Q_p|V_{dipo}|N\rangle = \sum_n CN(n, p, Q_p) f_n \quad (4)$$

where f_n is the oscillator strength for the electron/hole state $|n\rangle$. The three terms in the definition of $\eta_{PO}(\omega_{in}, \omega_{out})$ correspond respectively to the photoexcitation, energy relaxation and radiative recombination processes. This form for the optical response function assumes decoupled (and sequential) processes of absorption, relaxation and luminescence, and thus excludes coherent contributions such as resonant Raman and Rayleigh-like processes. In addition, it assumes that all the luminescence is related to the ground level $|G\rangle$. The use of this restriction, which can be lifted to obtain a more general response function, is actually guided by different experimental work, which associate as a rule the luminescence measured at low temperature and weak excitation intensity with $|G\rangle$ (and its replica, see below). We note that because of the deltas, and disregarding the relaxation term T_{rel}, the contribution of the bound polaron levels to the optical spectrum of a single dot is a series of sharp resonances (of different intensities) at

$$\hbar\omega_{in} - \hbar\omega_{out} = E_N - E_0 + m\hbar\omega_{ph} \quad (5)$$

with m an arbitrary integer. The phonon contributions to T_{cre} and T_{lum} arise from the fact that the light couples the dot excitonic *polarons* to the empty dot, which can be in a zero-phonon, one-phonon, etc state. We quote in the following various consequences of the use of the polaron formalism on the interband optical processes.

The optical response function $\eta_{PO}(\omega_{in}, \omega_{out})$ incorporates naturally the contributions of the so-called phonon replica to the luminescence and absorption processes. A replica of order $p_r = |p_{POL} - p_{VAC}| > 0$ is usually defined as a transition between a polaron state with a dominant component with p_{POL} phonons and the vacuum with p_{VAC} phonons. The $p_{POL} - p_{VAC} = 0$ case is usually called the zero-phonon (or 0th order) line (ZPL). Only the 0th order term is expected to be significant in T_{cre} at low temperatures ($K_B T \ll \hbar\omega_{ph}$; see however [11]), while the contribution of the p_rth replica in the photoluminescence at $\hbar\omega_{out} = E_0 - p_r\hbar\omega_{ph}$ is governed uniquely by the oscillator strength $F_{0,pr}$. Note that this definition holds for polaron states with a well-defined dominant contribution, which implies necessarily weak replica intensities with respect to the ZPL. This is in particular the case arising when the well-known Huang–Rhys model is applied to a semiconductor QD, leading to the exact solution for the different transitions involving one particular electron/hole state (e.g. the ground one $|g\rangle$ for $n = 0$) and all the phonon states. The electron/hole–phonon coupling is characterized by the so-called Huang–Rhys parameter S_{HR}, and the replica intensities decrease rapidly with increasing order p_r for small S_{HR} values [7, 8]. However, the true polaron levels of a QD do not have as a rule a predominant phonon component, even if the Huang–Rhys parameter is small. This is for instance the situation when the first optically active electron/hole P-like transition is detuned from the ground S-like one by about $\hbar\omega_{LO}$: indeed, in this case their resonant coupling will result in polaron states with, for each of them, two components of equal weights but different phonon numbers. Thus, transitions involving these highly mixed polarons cannot be properly assigned as ZPL or replica. In the previous example one would have, in the adiabatic approximation for the S and P states (i.e., when the V_F induced inter-level couplings are neglected), an absorption spectrum with a ZPL for the P-like transition and a S-related high energy first order replica. Moreover, these two transitions would have very different intensities. When the inter-level coupling is taken into account, the two resulting polaron states are mixed states which couple to vacuum (the electronic vacuum with, e.g., zero phonons) with the same strength, leading to two new transitions of the same intensity. Finally, it is worth stressing that the polaron transitions are not located at the same energies as the independent S-related and P-related ones, because of the inter-level Frohlich coupling.

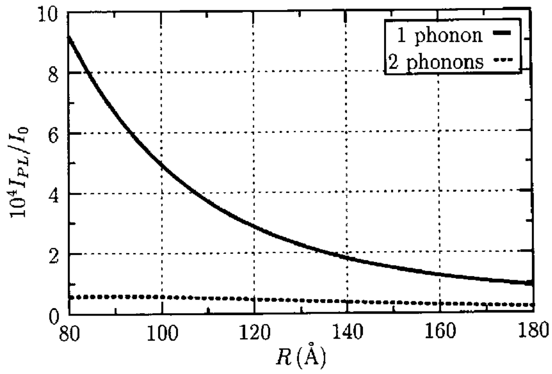

Figure 1. Relative intensities of the first- and second-order replicas as a function of the dot radius, for a dot modelled by a truncated cone of fixed height-to-radius ratio.

Another important consequence of the polaron coupling is the existence of many more optically active states than expected within the purely electron/hole scheme [4]. This can be more clearly illustrated for dots which have one dark state (when, e.g., the electron and the hole are in states of different symmetries, like the S_e–P_h transition) separated from the ground S transition by about $\hbar\omega_{LO}$: indeed, in this case their resonant coupling will result in *two* polaron states that both have an optically active component. This multiplication of the optically active lines is thus inherent to the polaron levels of QDs.

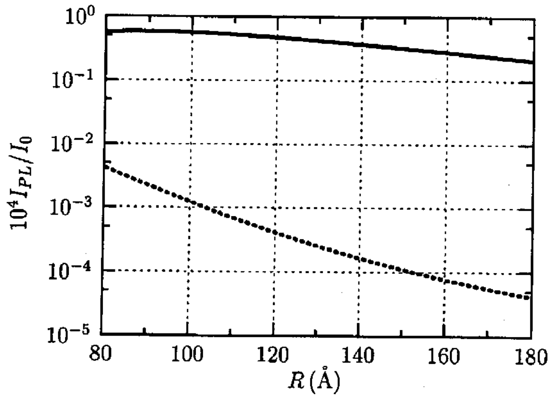

Figure 2. Relative intensities of the second-order replica as a function of the dot radius, for a dot modelled by a truncated cone of fixed height-to-radius ratio, for polaron states (solid curve) and for the Huang–Rhys model (dotted curve).

Let us briefly discuss the importance of polaron coupling in the low energy replica of luminescence. Such replicas arise naturally in the polaron framework. Moreover, their relative intensities as a function of p_r no longer follow the Huang–Rhys law [4, 5, 7, 8]. This is illustrated in figures 1 and 2. Figure 1 shows the intensities (relative to the fundamental one) of the first and second replicas as a function of the dot radius (for GaAs/Ga(In)As dots modelled by truncated cones with fixed height-to-radius ratio and basis angle). We found that the relative intensity of the first replica is not significantly modified, as compared to the Huang–Rhys value, while that of the second replica becomes enhanced by at least two orders of magnitude, as shown in figure 2. This can be understood within a perturbation model that accounts for both the intra-level phonon couplings (which leads to the Huang–Rhys result for the ground transition) and for the inter-level couplings.

To qualitatively demonstrate this result, and for simplicity, let us denote concisely by $|n\rangle \otimes |p\rangle$ a decoupled state with the electron/hole pair in state $|n(X)\rangle$ and the phonons in some proper state with occupancy p $(= 0, 1, 2, \ldots)$. The first-order correction involves the coupling of $|S_e S_h\rangle \otimes |0\rangle$ with the high energy states $|S_e S_h\rangle \otimes |1\rangle$, $|S_e P_h\rangle \otimes |1\rangle$ and $|S_h P_e\rangle \otimes |1\rangle$ states. The latter two are both non-radiative and thus do not lead to an additional radiative contribution, while the first one gives the Huang–Rhys result (to this order). At the second order, one should consider excursions towards $|S_e S_h\rangle \otimes |2\rangle$ and $|P_e P_h\rangle \otimes |2\rangle$ states, via any of the intermediate $|S_e S_h\rangle \otimes |1\rangle$, $|S_e P_h\rangle \otimes |1\rangle$ and $|S_h P_e\rangle \otimes |1\rangle$ states involved in the first-order term. All the states reached by two excursions are optically active, leading to additional (with respect to the Huang–Rhys case) contributions to the second order replica.

Note finally that, even if we neglect resonant processes ($\omega_{in} = \omega_{out}$), the sum in the definition of $\eta_{POL}(\omega_{in}, \omega_{out})$ keeps the $N = 0$ term. This is again a consequence of the fact that the *same* polaron state $|N\rangle$ (and in particular $|G\rangle$) can couple to the empty dot with different phonon occupancies and thus contribute to non-resonant low energy replicas.

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