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Non-perturbative optical response of a single quantum dot in a fluctuating environment

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Abstract
We present a model which treats non-perturbatively the effects of both dipolar coupling and reservoir fluctuations on the interband optical response of a single quantum dot. We are able to study the transition to a Rabi-like situation with increasing excitation power. The incorporation of an energy relaxation process (for instance, due to polaron relaxation among the dot polaron levels) in a three-level system allows us to simulate both photoluminescence (PL) and photoluminescence excitation experiments. The PL spectrum displays one or a few lines depending on the excitation intensity, relaxation efficiency and reservoir fluctuation rates.

1. Introduction

Semiconductor quantum dots have been pictured as isolated solid-state atoms. The dot eigenstates are however sensitive to 'external' perturbations because of their localized nature, which prevents any kind of dynamical averaging. In particular, the photoluminescence (PL) profile of a single dot differs in many aspects from a Lorentzian with a purely radiative broadening. The effects of various phonon couplings [1] and random reservoir fluctuations [2] on the ground state PL have already been discussed. Even though the the coupling to light is often assumed linear and/or the analysis restricted to the optical response of the ground dot transition [1, 2], non-linear optical effects have been observed recently [3], demonstrating that the dot states can couple strongly to light and that a perturbative treatment of the dipolar interaction fails when the excitation power increases. So we need a non-perturbative framework in order to account properly for the different kinds of influences on the spectral characteristics of the dot exciton luminescence.

2. Theoretical model

In order to evaluate the effect of charges trapped in the vicinity of the dot on the optical response of the confined electron–hole pair, we consider a simple model which accounts for the random population (which we assume to be a random telegraph process) of a one-level trap. In the following, we solve for the average time evolution of the density matrix of the confined carriers in the presence of randomly fluctuating perturbation $V_{\mathrm{per}}(t)$ and possibly also of a coherent laser excitation. The Hamiltonian of the system then reads

$$H = H_0 + V_{\mathrm{per}}(t) + V_{\mathrm{dip}}(t) + V_{\mathrm{res}}(t) \qquad (1)$$

where H_0 is the stationary dot Hamiltonian for a specific configuration of the dot environment, $V_{\mathrm{dip}}(t)$ is the dipolar coupling to the exciting laser (treated within the rotating frame approximation), $V_{\mathrm{per}}(t)$ is the element of randomness introduced by the trap charging and discharging and $V_{\mathrm{res}}(t)$ accounts for different reservoirs which couple irreversibly the eigenstates of H_0 among them (for instance, to describe the radiative lifetime or phonon-assisted processes among the bound dot levels). We consider that the different time-dependent perturbations $V(t)$ add their contributions to the evolution of the density matrix but do not interfere. Let us assume that the changes in V_{per} are due to instantaneous variations of the trap population. Let us denote by $\tau_\downarrow$ (respectively $\tau_\uparrow$) the average time interval that the trap remains occupied (respectively empty), and correspondingly by $\rho_\downarrow(t)$ and $\rho_\uparrow(t)$ we denote the density matrices which describe the carrier evolution when the trap is occupied and empty, respectively. Following the lines of [4], we obtain the coupled system of equations for the average time evolution $\overline{\rho_{\downarrow,\uparrow}}(t)$:

$$\dot{\overline{\rho_\uparrow}}(\tau) = -\frac{1}{\tau_\uparrow}\overline{\rho_\uparrow}(\tau) + \frac{1}{\tau_\downarrow}\overline{\rho_\downarrow}(\tau) + \frac{1}{\mathrm{i}\hbar}[H_\uparrow, \overline{\rho_\uparrow}(\tau)] + \left(\frac{\partial \rho_\uparrow}{\partial t}\right)_{\mathrm{rel}} \qquad (2)$$

$$\dot{\overline{\rho_\downarrow}}(\tau) = -\frac{1}{\tau_\downarrow}\overline{\rho_\downarrow}(\tau) + \frac{1}{\tau_\uparrow}\overline{\rho_\uparrow}(\tau) + \frac{1}{i\hbar}[H_\downarrow, \overline{\rho_\downarrow}(\tau)] + \left(\frac{\partial \rho_\downarrow}{\partial t}\right)_{\text{rel}} \tag{3}$$

where $H_\sigma(t) = H_0 + V_\sigma + V_{\text{dip}}(t)$ with V_σ being the electrostatic perturbation to the confined carriers when the trap is in the $\sigma = \uparrow$ or $\downarrow$ configuration. The relaxation terms account for the irreversible exchanges with phonons and photons, which affect both the populations and the coherencies of the density matrices $\overline{\rho_\uparrow}$ and $\overline{\rho_\downarrow}$.

From the evolution of the density matrix, we are able to calculate the optical response of the dot exciton in a fluctuating electrostatic environment by using the quantum regression theorem [5].

In the following, we shall restrict our results to two-level and three-level systems, by retaining the ground $|g\rangle$ (empty dot : $\langle g|V_\sigma|g\rangle = 0$) and at most two excited dot levels (one electron–hole pair in either the $|e_1\rangle$ or $|e_2\rangle$ pair states).

3. The two-level system

In the absence of laser excitation, the coherencies decrease exponentially in time, with decay rates $\omega_\pm$ given by

$$\frac{\omega_\pm + \omega_{\text{rad}}/2}{\omega} = -1 \pm \left[1 - \left(\frac{\Delta}{2\omega}\right)^2 + i\left(\frac{\Delta}{\omega}\right)\left(\frac{\tau_\downarrow - \tau_\uparrow}{\tau_\downarrow + \tau_\uparrow}\right)\right]^{1/2} \tag{4}$$

where $1/\omega_{\text{rad}}$ is the radiative lifetime of the $|e_1\rangle \rightarrow |g\rangle$ transition, $\omega = \frac{1}{2}\left(\frac{1}{\tau_\uparrow} + \frac{1}{\tau_\downarrow}\right)$ is the average rate of change of the environment (trap population) and $\hbar\Delta = \langle e_1|V_\uparrow|e_1\rangle - \langle e_1|V_\downarrow|e_1\rangle$ is the characteristic energy correction on the dot state. The real and imaginary parts of $\omega_\pm$ give the broadenings and the energy positions of the two Lorentzians that determine the PL lineshape. In particular, for $\tau_\uparrow = \tau_\downarrow$ one easily obtains that the PL profile displays two distinct thin lines of the same amplitude for slowly varying random perturbation ($\omega < \Delta/2$), which merge to form a single line which is maximally broadened when $\omega = \Delta/2$ and narrow down continuously to the radiative term when the rate of variation of the environment increases (HWHM $\approx \hbar[\omega_{\text{rad}}/2 + \Delta^2/2\omega]$ when $\omega \gg \Delta/2$). Then, the model leads to the result that a thin (radiatively broadened) PL line may indistinctly arise from either a static or a rapidly fluctuating environment. Similar results were previously obtained by Uskov *et al* [2], within a different framework (the Kubo–Anderson process).

Our model allows us to treat the optically driven two-level system. Let us note that Ω_g is the Rabi frequency that characterizes the dipolar coupling of the ground transition. We recover the well known fluorescence triplet lineshape in the absence of perturbation, with peaks separated by $\hbar\Omega_g$ and with relative intensities 1:3:1 (dashed line in figure 2, see e.g. [5]). The triplet-like structure of the dot fluorescence spectrum is robust against dichotomic random fluctuations: the peak positions and linewidths are only slightly modified, while their relative intensities are very sensitive to the fluctuations. This can be understood when we recall (i) that a small detuning of the laser excitation from resonance affects principally the 1:3:1 ratio, by favouring the side band nearer to excitation and (ii) that the fluctuation essentially alters (in a symmetrical way for $\tau_\uparrow = \tau_\downarrow$) the resonance condition between the dot transition and the laser energy.

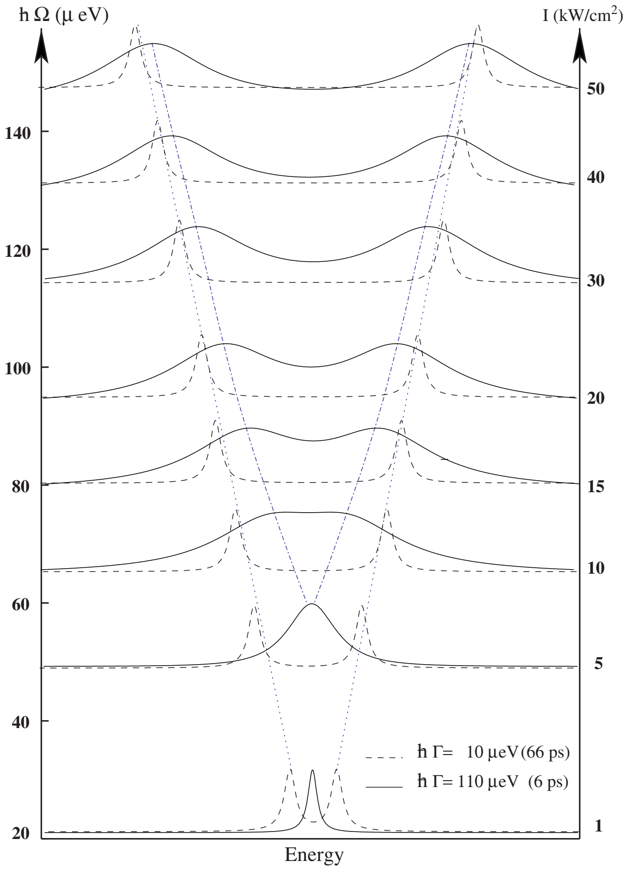

Figure 1. Evolution of the PL lineshape when increasing the excitation power for different relaxation rates.

4. The three-level system

In order to simulate different interband optical experiments, we have considered a three-level system that incorporates a relaxation channel between the two dot excited levels, in addition to diagonal energy corrections due to electrostatic fluctuations (non-diagonal couplings $\langle e_2|V_\sigma|e_1\rangle$ are neglected, since usually they are much smaller than $E(|e_2\rangle) - E(|e_1\rangle)$). The coupling of $|e_2\rangle$ with the excitation laser is characterized by the Rabi frequency Ω, and the energy relaxation from $|e_2\rangle$ to $|e_1\rangle$ by the rate Γ (for example, due to polaron-related relaxation [6]). Our model allows us to tackle many situations, such as the strong coupling to light, and also to study the effects of the relaxation process on the spectral characteristics.

4.1. PL simulation

At low relaxation and fluctuation rates, the calculated PL profile displays the strong coupling features recently observed [3]. By exciting at exact resonance with the excited transition, the PL profile displays two peaks of equal intensity separated by $\hbar\Omega$ as one could have expected within a 'dressed atom' framework. When the laser energy is detuned, the lineshape appears asymmetrical.

Actually, our model predicts that the spectral profile of the ground state luminescence is strongly dependent on the interplay between the dipolar interaction and the relaxation process. Figure 1 shows the evolution of the PL lineshape when increasing the excitation power for two different values of the relaxation rate.

If the rate is low (for instance, if $\hbar\Gamma = 10\ \mu$eV, dashed line in figure 1), the evolution of the PL lineshape shows a 'Rabi-like' splitting which is not the case when the rate is higher (for the solid line $\hbar\Gamma = 110\ \mu$eV, which corresponds to a relaxation time of the order of 6 ps, a reasonable value for a polaron interlevel relaxation in a single dot [6]). Following the calculations of [7], we show that the splitting occurs when $\Omega > \frac{\Gamma+\Gamma_{rad}^{|e_2\rangle}}{2}$ and that the broadening of the two lines is constant and equal to $\Gamma_{rad}^{|e_1\rangle} + \frac{\Gamma+\Gamma_{rad}^{|e_2\rangle}}{2}$ where $\Gamma_{rad}^{|e_1\rangle,|e_2\rangle}$ are the radiative rates of the two excited dot levels.

Finally, in the presence of fluctuations, the PL spectrum may evolve into a more intricate PL profile, with up to four peaks for a dichotomous process under suitable conditions: low relaxation rate and large energy corrections on the dot excited levels.

4.2. PLE simulation

The photoluminescence excitation (PLE) spectrum for excitation around the high energy transition may differ significantly from the one we would expect for a level broadened only by the radiative coupling and relaxation process.

For instance, when the detection is integrated within a small energy window (typically 1 meV wide, as in many experiments) around the $|e_1\rangle \rightarrow |g\rangle$ transition, the calculated PLE signal has a resonant-like (Lorentzian) profile which can be much larger than $\hbar\Gamma$ or $\hbar\Omega$. In fact, we can easily show that when $\Gamma \gg \omega_{rad}$ the width of the PLE spectrum is approximated by FWHM $\approx \hbar[\Gamma^2 + \Gamma\Omega^2/\omega_{rad}]^{1/2}$. It increases slowly with Ω at low excitations and becomes proportional to the Rabi splitting under high excitations, with a proportionality constant dependent on the relaxation rate.

5. Conclusion

In conclusion, we have presented a model to evaluate the PL and PLE of a quantum dot, which is non-perturbative in the dipolar coupling to a laser field and allows us to account for the influence of dichotomic random fluctuations due, for example, to charging/discharging of a trap level situated near the dot. Note that most of the results presented above do not rely on a precise description of the origin of the fluctuating process $V_{per}(t)$. We can, as an example, think about a carrier trapped by the presence of residual impurities during the growth process of the dots or about an exciton trapped by an interface defect of the wetting layer.

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