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Quantum fluctuations in semiconductor quantum dots and their contributions to the self-energy functions of exciton states

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Abstract. Influence of quantum fluctuations in a system consisting of a quantum dot and the reservoir of acoustic phonons on processes in which the quantum dot takes part is investigated. Under some conditions this influence is shown to be very strong. We find a contribution from the quantum fluctuations to the self-energy function of the exciton coupled to the quantum dot.

1. INTRODUCTION

Semiconductor quantum dots being nanometer-scale islands have unique optical properties [1] that make them attractive candidates for many optoelectronic applications such as new types of single-photon sources [2, 3] and lasers [4, 5], or for use as computational building blocks of a quantum computer [6–8]. However, quantum dots are embedded in a surrounding solid, and the carriers confined to the dot interact with their environment most notably with phonons. Because of a strong suppression of phonon induced transitions between electronic states localized in quantum dots, the carrier-phonon interaction results in the decoherence of the optical polarization i.e. in pure dephasing being a major source for the decoherence. For S-shell excitons, the dominant sources of the pure dephasing are the longitudinal acoustic phonons [9–17]. Pure dephasing is caused by real irreducible processes. However, a significant contribution to the dot-reservoir interaction comes also from quantum fluctuations in which the reservoir degrees of freedom manifest themselves in a virtual state (i.e. the energies of the intermediate states are not equal to the energy of the initial and final state). Quantum fluctuations give rise to the shift of the dot energy levels known as the polaron shift. They also give a dominant contribution to self-energy functions of the quantum dots (QD) states. The energy dependence of QD self-energy functions can have a significant effect on the emission spectra of a strongly coupled quantum-dot cavity systems [18–21]. The problem is that despite the self-energy functions are used for description of the evolution QD interacting with environment, usually they are derived from quantum-field Green functions. A consistent description of quantum dynamics is ensured only by making use of the Green operator. In atomic physics where the self-energy functions of atomic states determine the Lamb shifts and widths of spectral lines the Green-function method has turned out to be very effective. The reason for this is the weakness of the electromagnetic interaction. However, the self-interaction of QD excitons is much more significant than the interaction of an atom with its own radiation field. In this paper we investigate the effect of

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quantum fluctuations in a system consisting of a QD and the reservoir of acoustic phonons on processes in which the QD takes part. We find a contribution to the QD exciton self-energy function from the quantum fluctuations. In our investigation we make use of a generalized dynamical equation that has been derived by Gainutdinov [22] as the most general equation of motion consistent with the current concepts of quantum physics. We show that this equation that has turned out an important tool for solving many problems in nuclear physics [23–25], and quantum optics [26] provides a straightforward and effective way to describe the interaction of QDs with their environment.

2. Method

Being equivalent to the Schrödinger equation in the case of instantaneous interactions, the Gainutdinov equation of motion permits the generalization to the case where dynamics of a quantum system is governed by a nonlocal-in-time interaction. One of the energy representations [27–31] of this equation takes the form

$$\frac{dT(z)}{dz} = -T(z)(G_0(z))^2 T(z), \tag{1}$$

where $G_0(z) = (z - H_0)^{-1}$ with H_0 being the free Hamiltonian, and $T(z)$ is defined as

$$T(z) = i\int_0^\infty d\tau \exp(i(z - H_0)t_2)\tilde{S}(t_2, t_1)\exp(-i(z - H_0)t_1). \tag{2}$$

Here $\tilde{S}(t_2, t_1)$ is the contribution to the evolution operator from the process in which the interaction in the system begins at time t_1 and ends at time t_2. The boundary condition for equation (1) is,

$$T(z) \underset{|z|\to\infty}{\to} B(z) \tag{3}$$

where

$$B(z) = i\int_0^\infty d\tau \exp(iz\tau)H_{int}^{(s)}(\tau),$$

where

$$H_{int}^{(s)}(t_2 - t_1) = \exp(-i(z - H_0)t_2)H_I(t_2, t_1)\exp(-i(z - H_0)t_1),$$

with $H_I(t_2, t_1)$ being the generalized interaction operator.

The operator $T(z)$ determines the Green operator $G(z)$ being the evolution operator in the energy representation

$$G(z) = G_0(z) + G_0(z)T(z)G_0(z). \tag{4}$$

Equation of motion (1) with the boundary condition determines the dynamics of the system. The contribution to the Green operator $G(z)$, which comes from the processes associated with the self-interaction of the system, has the same structure as the free Green operator $G_0(z)$. For this reason it is natural to replace $G_0(z)$ by the operator $\tilde{G}_0(z)$, which describes the evolution of the system in the case where the interaction in the system is reduced to the self-interaction and hence has the structure

$$\tilde{G}_0(z) = (z - H_0 - C(z))^{-1}, \tag{5}$$

where the operator $C(z)$ has the same eigenvectors as H_0 ($C(z)|n\rangle = C_n(z)|n\rangle, H_0|n\rangle = E_n|n\rangle$). Correspondingly, the operator $T(z)$ is the operator describing the whole interaction in the system except the self-interaction. These operators are related as follows

$$G(z) = G_0(z) + G_0(z)T(z)G_0(z) = \tilde{G}_0(z) + \tilde{G}_0(z)M(z)\tilde{G}_0(z). \tag{6}$$

By making use of this equation the equation of motion (1) can be represented in the form of two equations for $M(z)$ and $C(z)$. Equation for the self-energy function takes the form

$$\frac{dC_n(z)}{dz} = -\sum_m \frac{\langle n|M(z)|m\rangle\langle m|M(z)|n\rangle}{(z - E_m - C_m(z))^2}. \tag{7}$$

3. Quantum fluctuations in phonons and the exciton self-energy function.
Let us consider the self-energy of a QD exciton within independent boson model (IBM) [18]. The IBM Hamiltonian describing phonons and exciton-phonon coupling reads [19]

$$\widehat{H}_{ph} + \widehat{H}_{ep} = \sum_q \omega_q b_q^* b_q + \sum_q g_x^q \left(b_q + b_q^+\right)|x\rangle\langle x|, \tag{8}$$

where $|x\rangle$ is the vector of the excitonic state, q denote the different phonon modes with energy ω_q, the creation (b_q^+) and annihilation (b_q) operators of phonons with momentum $\mathbf{q}$ and frequency ω_a obey the usual commutation relations for bosons, and G_x^q is the deformation potential coupling, which depends on the material parameters of the host semiconductor and the exciton wave function.

The self-energy function not only determines the energy shift of the exciton caused by the vacuum fluctuations in the QD exciton-phonon reservoir system, but also its energy dependence has a significant effect on the exciton spectrum. It has been shown [20] that a good account of the exciton spectrum can be obtained already at the second-Born perturbation level. At this order the solution of equation (7) takes the form

$$\frac{d\langle x,\nu|C^{(2)}(z)|x,\nu\rangle}{dz} = -\sum_\mu \frac{\langle x,\nu|H_I(z)|x,\mu><x,\mu|H_I(z)|x,\nu\rangle}{(z - E_\mu)^2}. \tag{9}$$

By taking the thermal average of (9) we get

$$\frac{d\langle x,|C^{(2)}(E)|x\rangle}{dz} = -\sum_q \left\{ \frac{|g(q)|^2(1+n(q))}{(E - E_x - w(q) + i0)^2} + \frac{|g(q)|^2 n(q)}{(E - E_x + w(q) - i0)^2} \right\}, \tag{10}$$

with $n(q) = [e^{\omega(q)/k_B T} - 1]^{-1}$ being the mean phonon occupation number at a bath temperature T. Solving this equation yields

$$C_x^{(2)}(E) = \sum_q \left\{ \frac{|g(q)|^2(1+n(q))}{E - E_x - w(q) + i0} + \frac{|g(q)|^2 n(q)}{E - E_x - w(q) - i0} \right\}. \tag{11}$$

This equation can be rewritten as

$$C_i(E) = \frac{S_{HR}}{\omega_b^2} \int \omega^3 \exp\left(-\frac{\omega^2}{2\omega_b}\right) \left(\frac{1+n(q)}{E - E_i - \omega + i0} + \frac{n(q)}{E - E_i + \omega - i0}\right) d\omega, \tag{12}$$

where S_{HR} is dimensionless Huang-Rhys parameter characterizing the phonon-exciton coupling.

In general the self-energy function $C_x(E)$ can be represented as a sum of the contribution $\Sigma_x(E)$ from the off-shell quantum-fluctuation processes and the contribution $C_x^{on}(E)$ from on-shell processes leading to the system damping

$$C_x(E) = \Sigma_x(E) + C_x^{(on)}(E).$$

The interaction of the QD exciton with acoustic phonon reservoir generates pure quantum fluctuations that do not lead to decay processes, and, as a result, $C_x^{(2)}(E)$ gives contribution only to $\Sigma_x(E)$. Up to now we did not account for the self-energy processes caused by the interaction of the exciton with its own radiation field. These processes also give contributions to the quantum fluctuation function $\Sigma_x(E)$. However, this contribution is small compared with that from the quantum fluctuations in the QD exciton-phonon reservoir system and can be neglected. At the same time, the interaction of the exciton with the radiation field leads to its radiative recombination processes that being on-shell ones give contribution to the function $C^{(on)}(E) = -i\Gamma_r/2$ with Γ_r being the radiative decay rate.

For the effect of the quantum fluctuation to be strong the Huang-Rhys parameter S_{HR} must be large enough. In a field of exciton-acoustic phonon coupling (EPC) in QDs experimental studies generally show little agreement with theory or each other even among studies on the same material [32–34]. And it is hard to predict exciton-phonon coupling strength dependence on the nanocrystal radius or temperature [33, 34]. There are several experimental techniques that can be used to obtain EPC. Each method suffers from its own set of technical and/or interpretive complexities. The most straightforward is vibrationally resolved absorption or emission spectroscopy. In the low-temperature limit, the ratio of the one-phonon to zero-phonon intensity is equal to the parameter S_{HR} for that phonon mode. In Ref. [35] by using photoluminescence (PL) spectroscopy authors measured the Huang-Rhys constant for the system with InAs self-assembled quantum dots embedded in GaAs, and find the value $S_{HR} = 0.5$. They consider the Frohlich interaction of the electron-hole pair with bulk GaAs phonons in the system and interpret the phonon-assisted photoluminescence as being indicative of an enhanced electron-phonon interaction. The second type of experiments, resonance Raman scattering (RRS), provides the same type of information. Using this method it is much easier to measure quantities, depending on the Huang-Rhys parameter, but the interpretation of the data is less straightforward. In Ref. [33] authors obtain the Huang-Rhys parameter $S_{HR} = 0.7$ for PbS nanocrystals, 1.5 nm radius using RRS. Also, there are some domain measurements: femtosecond pump-probe, photon echo, etc. Short light pulses in these methods can impulsively excite those phonons that are coupled to the electronic excitation, producing a modulation in the signal as a function of probe delay. Here the Huang-Rhys factor can be obtained by Fourier transformation, followed by modelling of the time-dependent signal [34]. Experimental measurements, made in Ref. [32] show Huang-Rhys factor in range 0.15-0.35.

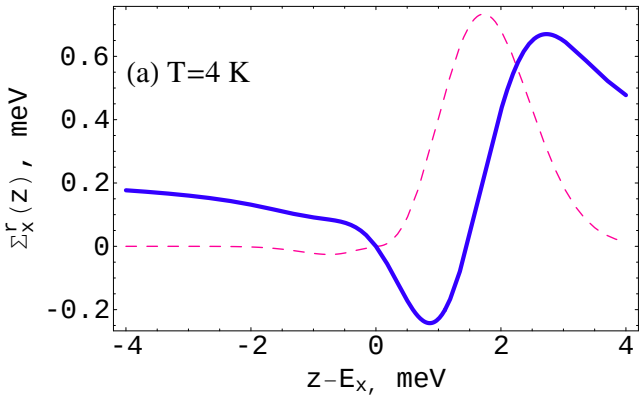


Figure 1. Quantum fluctuation contribution to the excitonic self-energy function ($T = 4$ K and $S_{HR} = 0.2$), where the solid curve represents the real contribution and the dashed curve the imaginary contribution.

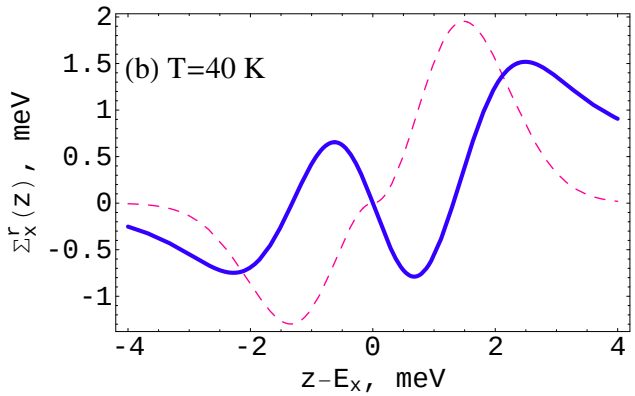


Figure 2. Quantum fluctuation contribution to the excitonic self-energy function ($T = 40$ K and $S_{HR} = 0.2$), where the solid curve represents the real contribution and the dashed curve the imaginary contribution.

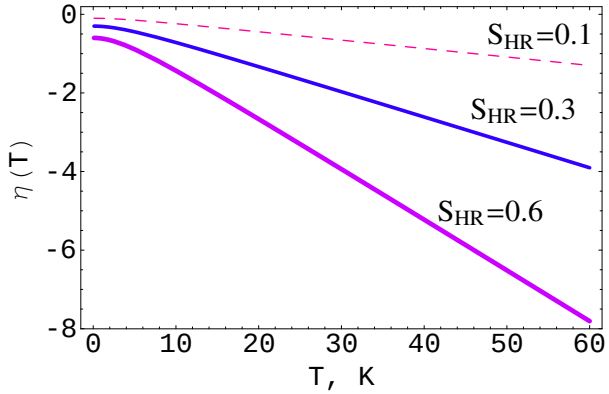


Figure 3. The temperature dependence of the parameter η.

The results of the calculations of the pure self-energy function presented in figures 1–3 show that the second-Born approximation we have made in deriving equation (11) actually is not well-defined for $S_{HR} \geq 0.1$ even at very low temperatures. In fact, in this case the self-energy function in the denominator of equation (7) cannot be neglected. Substituting $C_i(E)$ represented in the form

$$C_x(E) = (E - E_x)\eta + \tilde{C}_x(E), \tag{13}$$

(E_x is assumed to include the self-energy shift $C_x(E_x)$) with

$$\eta = \frac{dC_x(E)}{dE}|_{E=E_x} \tag{14}$$

into equation (11), and neglecting $\tilde{C}_x(E)$ that is small for the relevant energies we get

$$C_i^{(2)}(E) = \sum_q \left\{ \frac{|g(q)|^2 Z_1^2 (1 + n(q))}{(E - E_x - w(q) + i0)Z_2^2} + \frac{|g(q)|^2 Z_1^2 n(q)}{(E - E_x + w(q) - i0)Z_2^2} \right\}, \tag{15}$$

where $Z_2 = 1 - \eta$. The factor Z_2 can be regarded as a constant renormalizing the exciton propagator. Appearance of the factor Z_1 manifests the fact that the renormalization of the

exciton propagator must be accompanied by a renormalization of the exciton-phonon coupling. An additional factor Z_2 appears because of the renormalization of the propagators associated with external lines. Since as usual $Z_1 = Z_2$, these constants in equation (15) compensate each other and we come back to equation (11), in which the parameter Z_2 does not manifest itself. However, there are physical situations where this parameter and hence the self-energy function $\Sigma_x(E)$ come into play. This, for example, takes place in the case when the quantum dot is strongly coupled to a cavity. In this case the cavity-emitted spectrum depends on the exciton self-energy function [18–21].

4. Conclusion

We have shown that the exciton self-interaction associated with quantum fluctuations in the combined system consisting of a semiconductor quantum dot and a reservoir of acoustic phonons can be strong. Our investigations show that this takes place in the case when Huang-Rhys parameter S_{HR} is more than 0.1. In this case the parameter Z_2 that determines strength of self interactions becomes more than one even at low temperatures. This parameter that in the renormalization theory renormalizes propagators of particles, in the case of semiconductor QDs can manifests itself, for example, in the cavity-emitted spectrum when a dot is embedded in a resonant cavity. The quantum fluctuation processes can manifest themselves not only in the self-energy function. In the case when the QD is coupled to resonance laser field they can give rise to the nonradiative transitions between the dressed states. A deeper understanding of these processes can provide a novel insight into the problem of the pure dephasing.

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