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Local Field Effects in an Isolated Quantum Dot: Self-Consistent Microscopic Approach

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A self-consistent microscopic local-field theory for an isolated arbitrary shaped quantum dot (QD) has been presented. In our approach we introduce into the Schrödinger equation a depolarization field induced by an external electromagnetic field, and combine this idea with the second quantization technique for electron–hole pairs. As a result, we succeeded in deriving the general self-consistent nonlinear equations for the system "QD + classical electromagnetic field". In the linear approximation, our approach reproduces microscopically the depolarization shift of the QD gain band and, in anisotropically shaped QDs, polarization dependent splitting of this band.

Introduction Conventionally, local field effects are related to a difference between acting and mean fields in statistically large ensembles of particles like dense gases and artificial composite materials. Electromagnetic properties of such ensembles are usually modeled in the framework of the effective–medium approach. For this approach to hold, all inclusions must be electrically small, i.e., their maximal linear size must be small comparing with the wavelength. In quantum dots (QDs), besides collective effects inherent in ensembles, local field effects appear related to individual properties of isolated QDs. These effects are due to electromagnetic field discontinuity on the QD boundary and resonant nature of excitons in QDs. Local field effects in isolated QDs were considered in a number of papers, e.g., [1–3], on the basis of different phenomenological models. For the strong confinement regime, a phenomenological theory of linear electromagnetic response of regular 3D-ensembles of QDs has been elaborated in Refs. [4, 5]. This theory incorporates both collective and individual local field effects. In particular, polarization-dependent splitting of the gain band in anisotropically shaped QDs has been predicted. Collective local field effects in 2D arrays of QDs in both strong and weak confinement regimes are discussed in Ref. [6].

Phenomenological local-field theory developed in [4–6] contains a set of parameters which must be extracted from the microscopic consideration; moreover, extension of the approach to new problems, such as nonlinear interactions, interaction with nonclassical light, etc., meets significant difficulties. Thus, the microscopic self-consistent theory is a matter of interest for further progress of electromagnetic modeling of QDs. For an ensemble of two-level atoms in a dielectric continuum, such an approach has been de-

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veloped in Ref. [7]. The basic idea of the theory consist in that the relations between local and acting fields are included into equations of motion. In our approach we combine this idea with the second quantization technique for electron–hole pairs. This allows us to describe local fields in systems with varying number of particles.

Basic Relations Let an isolated QD interact with a given classical electromagnetic field. The QD is considered to be a strongly confined [1] two-level system (below indices e and g correspond to the excited and ground levels, respectively). In the strong confinement regime, quantization of the motion of electrons and holes in QD is determined by the interaction of the particles with the QD boundary. In that case, a QD exposed to an external electromagnetic field is described by the Hamiltonian $H = H_0 + H_{\mathrm{IL}}$, where $H_0 = \epsilon_{\mathrm{e}} a_{\mathrm{e}}^+ a_{\mathrm{e}} + \epsilon_{\mathrm{g}} a_{\mathrm{g}}^+ a_{\mathrm{g}}$ is the Hamiltonian of the particle motion, $\epsilon_{\mathrm{e,g}}$ are the energy eigenvalues, $a_{\mathrm{e,g}}$ and $a_{\mathrm{e,\,g}}^+$ stand for the creation and annihilation operators. The term H_{IL} describes interaction of particles with electromagnetic field. We use 3D Cartesian coordinate system with the unit vector $\mathbf{u}_x$ parallel to the electron–hole pair dipole moment: $\boldsymbol{\mu} = \mu \mathbf{u}_x$. In the chosen coordinates $H_{\mathrm{IL}} = -V\hat{P}_x E_{\mathrm{L}x}$, where $\hat{P}_x = V^{-1}(-\mu b^+ + \mu^* b)$ is the polarization operator, $b^+ = a_{\mathrm{g}} a_{\mathrm{e}}^+$ and $b = a_{\mathrm{g}}^+ a_{\mathrm{e}}$ are the creation and annihilation operators for electron–hole pairs, respectively, V is the QD volume.

The field inside the QD, $\mathbf{E}_{\mathrm{L}}$, is different from the external acting field $\mathbf{E}_0$. Since we postulate the QD to be electrically small, this difference is determined by [8]

$$\mathbf{E}_{\mathrm{L}} = \mathbf{E}_0 - 4\pi \hat{\mathbf{N}} \mathbf{P} \,. \tag{1}$$

Here $\mathbf{P} = \langle \hat{\mathbf{P}} \rangle$ is the macroscopic polarization, $\hat{\mathbf{N}}$ is the depolarization tensor. This tensor is symmetrical and depends only on the QD shape. Using Eq. (1) one can easily obtain $E_{\mathrm{L}x} = E_{0x} - 4\pi N_x P_x$, where the depolarization coefficient N_x is as follows: $N_x = (\boldsymbol{\mu} \cdot \hat{\mathbf{N}} \boldsymbol{\mu})/|\mu|^2 \equiv (\mathbf{u}_x \cdot \hat{\mathbf{N}} \mathbf{u}_x)$. Taking into account the above expression for $E_{\mathrm{L}x}$, the total Hamiltonian can be represented by

$$H = H_0 + H_{\mathrm{I0}} + \Delta H, \tag{2}$$

where

$$H_{\mathrm{I0}} = -V\hat{P}_x E_{0x} \,, \tag{3}$$

$$\Delta H = 4\pi N_x P_x(-\mu b^+ + \mu^* b) = \frac{4\pi}{V} N_x(\mu^* b - \mu b^+)\,(\mu^* \langle b \rangle - \mu \langle b^+ \rangle) \,. \tag{4}$$

Thus, in the total Hamiltonian we have separated the contribution of the interaction of electron–hole pairs with acting field, H_{I0}, and the contribution of the depolarization, ΔH. Such a separation allows us to describe local field effects without explicit solution of the electrodynamical boundary-value problem.

Let $|\psi(t)\rangle$ be the wavefunction of the system “QD + classical electromagnetic field”. In the interaction representation this system is described by

$$i\hbar \frac{\partial |\tilde{\psi}\rangle}{\partial t} = H_{\mathrm{int}} |\tilde{\psi}\rangle \,, \tag{5}$$

where $|\tilde{\psi}\rangle = \exp(iH_0 t)\,|\psi(t)\rangle$ and $H_{\mathrm{int}} = \exp(iH_0 t)\,(H_{\mathrm{I0}} + \Delta H)\exp(-iH_0 t)$. Let $|\tilde{\psi}(t)\rangle = D(t)\,|c\rangle + M(t)\,|v\rangle$ with $D(t)$ and $M(t)$ as functions to be found, and $|c\rangle = |0_{\mathrm{g}}, 1_{\mathrm{e}}\rangle$ and $|v\rangle = |1_{\mathrm{g}}, 0_{\mathrm{e}}\rangle$. Taking into account the well known identities $b^+ |c\rangle = b\,|v\rangle = 0$ and

$b\,|c\rangle = |v\rangle$, $b^+\,|v\rangle = -|c\rangle$, from Eq. (5) we obtain the following set of equations of motion:

$$i\hbar\,\frac{\partial D}{\partial t} = (4\pi N_x P_x - E_{0x})\,\mu M\,\mathrm{e}^{i\omega_0 t}, \qquad i\hbar\,\frac{\partial M}{\partial t} = (4\pi N_x P_x - E_{0x})\,\mu^* D\,\mathrm{e}^{-i\omega_0 t}\,. \tag{6}$$

Here $P_x = \langle\psi|\,\hat{P}_x\,|\psi\rangle = V^{-1}[\mu D^*(t)\,M(t)\,\mathrm{e}^{i\omega_0 t} + \text{c.c.}]$ is the macroscopic polarization. In the slow-varying amplitude approximation the acting field is given by $E_{0x} = [\mathcal{E}(t)\exp(-i\omega t) + \text{c.c.}]/2$. Using the above expression for P_x and neglecting the fast-oscillating terms in Eqs. (6), we derive the final expressions for the equations of motion

$$i\hbar\,\frac{\partial D}{\partial t} = \frac{4\pi N_x}{V}\,|\mu|^2\,D\,|M|^2 - \frac{1}{2}\,\mathcal{E}(t)\,\mu M\,\mathrm{e}^{i(\omega_0-\omega)\,t}\,, \tag{7}$$

$$i\hbar\,\frac{\partial M}{\partial t} = \frac{4\pi N_x}{V}\,|\mu|^2\,M\,|D|^2 - \frac{1}{2}\,\mathcal{E}^*(t)\,\mu^* D\,\mathrm{e}^{-i(\omega_0-\omega)\,t}\,. \tag{8}$$

These equations constitute a basic self-consistent system describing the interaction of QD with electromagnetic field. The consistency is provided by the depolarization-induced first terms in the right-hand parts of the equations. Physically, system (7)–(8) is analogous to the Bloch equations for optically dense media derived in Ref. [7]. The relaxation can easily be included into Eqs. (7)–(8) either by the introduction of the phenomenological transverse and longitudinal relaxation times [7] or by a corresponding modification of initial Hamiltonian (2).

Polarization of QD in Linear Approximation The case of the ground-state QD can be analyzed using Eqs. (7) and (8) with the initial conditions $D(0) = 0$, $M(0) = 1$ imposed. In the linear approximation we can put $M(t) \approx 1$. Physically, this means that we restrict the analysis to temporal intervals essentially small in comparison to the relaxation time of the given resonant state. In such a situation, Eqs. (7) and (8) are reduced to

$$i\hbar\,\frac{\partial D}{\partial t} = \frac{4\pi}{V}\,N_x\,|\mu|^2\,D - \frac{1}{2}\,\mathcal{E}(t)\,\mu\,\mathrm{e}^{i(\omega_0-\omega)\,t}\,. \tag{9}$$

For the time-harmonic excitation, when $\mathcal{E}(t) = \mathcal{E} = \text{const}$, this equation gives

$$D(t) \approx \frac{\mathcal{E}\mu}{2\hbar(\omega_0 + \Delta\omega - \omega)}\,[\mathrm{e}^{i(\omega_0-\omega)\,t} - \mathrm{e}^{-i\,\Delta\omega\,t}]\,, \tag{10}$$

where

$$\Delta\omega = \frac{4\pi}{\hbar V}\,N_x\,|\mu|^2\,. \tag{11}$$

Thus, depolarization leads to the shift $\Delta\omega$ of the resonant frequency [1–3].

Equation (11) is identical to that obtained in [4, 5]. In order to demonstrate it, we should substitute $|\mu|^2 \to |\mu_0|^2/3$, where μ_0 is the matrix element of the dipole moment of a corresponding bulk sample (the coefficient 1/3 is due to the orientational averaging in bulk samples). We should also take into account the spin degeneracy of electron–hole pairs. Then, expressing the macroscopic polarization in terms of $D(t)$, we find

$$P_x = -\frac{1}{8\pi}\,\alpha_{xx}(\omega)\,\mathcal{E}[\mathrm{e}^{-i\omega t} - \mathrm{e}^{-i(\Delta\omega+\omega_0)\,t}]\,, \tag{12}$$

where

$$\alpha_{xx}(\omega) = \frac{4\pi\,|\mu|^2}{\hbar V(\omega - \Delta\omega - \omega_0)} \tag{13}$$

is the component of the QD polarizability tensor. This quantity has been evaluated in Refs. [4, 5] phenomenologically with the Lorentz dispersion law for the permittivity: $\varepsilon(\omega) = \varepsilon_h + g_0/(\omega - \omega_0)$. Correlation made between expressions obtained in Refs. [4, 5] and the above analysis allows us to express the phenomenological coefficient g_0 in terms of the microscopic parameters of QD: $g_0 = 4\pi|\mu|^2/V$.

For an excited QD, the initial conditions have the following form: $D(0) = 1$, $M(0) = 0$. Applying to this case the above presented procedure, we obtain

$$M(t) \approx \frac{\mathcal{E}^* \mu^*}{2\hbar(\omega_0 + \Delta\omega' - \omega)}\,[\mathrm{e}^{-i(\omega_0 - \omega)\,t} - \mathrm{e}^{-i\,\Delta\omega\,t}] \tag{14}$$

with $\Delta\omega' = -\Delta\omega$. Thus, for an excited state the local field effects lead to the same shift $\Delta\omega$ (11) of the resonance frequency but with the opposite sign. Interaction of the ground state with the electromagnetic field corresponds to the optical absorption, while interaction with an excited QD corresponds to the case of stimulated emission. In other words, the optical absorption and gain of an isolated QD could be distinguished owing to the depolarization shift, blue in the former case and red in the later one.

Conclusion Taking into account the local fields, we have formulated *general self-consistent microscopic nonlinear equations* describing the interaction of an isolated QD with a classical electromagnetic field. In the linear approximation, our approach reproduces the depolarization shifts of the absorption and the gain bands, blue in the former case and red in the later one, and gives microscopic expressions for the input parameters of the phenomenological theory presented in Refs. [4, 5]. The derived general system can be applied to investigation of different nonlinear processes in QDs which are expected to be strongly influenced by the local fields; phenomenological model of the depolarization-induced bistability has been presented in Ref. [1]. A new class of problems, interaction of QDs with nonclassical light (spontaneous emission, quantum fluctuations, etc.), also promises nontrivial manifestations of the local field effects. For this class of phenomena, a special procedure of quantization of the inhomogeneous electromagnetic fields should be elaborated. The Hamiltonian derived in the present paper can be used for the extension of system (7)–(8) to the case of nonclassical light.

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