EXCITONIC COMPOSITES

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Abstract. A new class of composite materials, excitonic composite, has been considered by the example of a collection of identical semiconductor quantum dots (QDs). The QD is modeled by a spatially confined exciton interacting with external electromagnetic field. A self-consistent microscopic local-field theory for an isolated arbitrary shaped QD has been developed. A Hamiltonian of the system has been formulated in terms of the acting field with a separate term responsible for the effect of depolarization. Relations between local and acting fields in QD have been derived in the dipole approximation for both strong and weak confinement regimes with the account for pronounced spatial nonlocality of the electromagnetic response in the latter regime. Homogenezation procedures have been carried out for both cases and expressions for the effective dielectric function have been derived. It has been shown that the nonlocality changes components of the polarizability tensor but does not change the general representation of the scattering operators as compared to the strong confinement regime.

1. Introduction

Fascinating electronic and optical properties of spatially confined nanostructures irreducible to properties of bulk media, and great potentiality of such structures in engineering applications have motivated permanent extension of their study. Among a variety of new results in this field, the recent progress in the synthesis of sheets of nano-scale 3D confined narrow-gap insertions in a host semiconductor, quantum dots (QDs), is of a special interest. Indeed, QD-based structures provide practical realization of the idea proposed by Dingle and Henry [1] to use structures with size quantization of charge carriers in one or more directions as active media of double heterostructure laser. Such a laser will show radically changed

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characteristics as compared to conventional quantum well (QW) lasers [2, 3]. Kirstaedter *et al.* [4] demonstrated very low threshold current density and its practically complete temperature insensitivity in InGaAs/GaAs QD laser up to 180 K. Ultrahigh material and differential gain, orders of magnitude exceeding those in QW lasers in a spectral range far beyond those available for conventional strained InGaAs-GaAs QWs, has been experimentally confirmed [5]. The large body of recent results on physical properties of QDs and their utilization for the QD laser design has been accumulated in Ref. [5]. Recently QDs have been proposed to serve as nodes of a quantum network that store and process quantum information being transmitted between nodes by entangled states of photons (see, e.g., [6] and references therein).

The key peculiarities of QD heterostructures are related to spatial confinement of the charge carrier motion and intrinsic spatial inhomogeneity. Since the inhomogeneity scale is much less than the optical wavelength, inclusions (QDs) can be treated as *electrically small* objects and electromagnetic response of such heterogeneous structures, *composites*, can be evaluated by means of effective medium theory [7]. Application of effective medium approach to 3D arrays of QDs has been presented in Refs. [8, 9, 10, 11]. In many cases, however, a planar array of QDs with intrinsic 2D periodicity of characteristic period much less than the optical wavelength, can be treated as more adequate and realistic model [5]; a general method for evaluation of electromagnetic response of planar arrays of QDs has been presented in Ref. [12]. This method, conventionally referred to as the effective boundary condition method, has been originally developed for microwaves and antenna theory (see, e.g., [13] and references therein), and has found a wide application in these fields.

Effective medium theory must be modified to include specific properties of excitons, coupled electron-hole states, which define the QD response (by this reason, further we speak about excitonic composites). First, the excitonic composite is constituted by resonant particles and, consequently, is characterized by a resonant response; moreover, inverse population is possible owing to the discrete energy spectrum of excitons. Thus, *excitonic composite is a resonant active system.* Another specific property of excitonic composites is appeared owing to the quantum nature of excitons: the exciton Bohr radius can either exceed the QD radius ($a_B \gg R_0$, strong confinement regtime) or be much less ($a_B \ll R_0$, weak confinement regime). In the later case, often realized in experiments, the QD electromagnetic response becomes *nonlocal*: constitutive relations for the polarization of the QD medium takes the form of integral operators. Peculiarities of the excitonic composite are in the focus of the presented paper. Constitutive relations for a QD accounting for the nonlocality have been derived in

Refs. [14, 15, 16]. In our paper we solve the Maxwell equations imposed to constitutive relations of such a type.

2. Excitonic composite in the strong confinement regime

2.1. LIGHT-MATTER INTERACTION HAMILTONIAN FOR A SINGLE QD

Let an isolated QD be exposed to classical electromagnetic field. Further such a QD is modeled as a strongly confined in space two-level quantum oscillator. Obviously, QD is essentially multilevel system. However, contribution of transitions lying far away from a given resonance can be approximated by a nonresonant dielectric function ε_h. We shell assume ε_h to be equal to the dielectric function of host semiconductor. Thus, in our model interaction of quantum oscillator with external electromagnetic field occurs inside a homogeneous boundless medium characterized by the dielectric function ε_h. For our consideration it is essential that ϵ_h can be assumed to be frequency independent and real-valued. This allows us to put $\varepsilon_h = 1$ without loss of generality. Substitutions in final expressions $c \to c/\sqrt{\varepsilon_h}$ and $\boldsymbol{\mu} \to \boldsymbol{\mu}/\sqrt{\varepsilon_h}$ for the speed of light and the oscillator dipole moment, respectively, will restore the case $\epsilon_h \neq 1$.

In the strong confinement regime the Coulomb interaction is assumed to be negligible, so that electrons and holes in QD are moved independently and spatial quantization is entailed by the interaction of the particles with the QD boundary. In this section we aim at the development of the Hamilton formalism, which would describe the system "QD + electromagnetic field" taking into account the role of QD boundaries. Apparently, the most sequential and rigorous approach to the problem is based on the concept of spatially varying interaction coefficient [17]. However, utilization of the approach for systems with the stepwise interaction coefficient meets the problem that the Hamilton equations are inapplicable at the discontinuity. The same problem exists in macroscopic electrodynamics of stratified media. By analogy, introducing a transient layer and reducing its thickness, one can obtain boundary conditions complimentary to the Hamilton equations. However, in practical use, the approach described turned out to be too complicated and was realized for the only simplest case: interaction of a material layer with normally incident light [17]. Note that even in this simplest case the local field effects are left beyond the consideration.

As applied to QDs, in our paper we develop a more constructive approach which utilizes the property of QDs to be electrically small. This property allows us to assume local and acting fields to be homogeneous inside the QD. In fact, this implies that we introduce a spatial averaging of the electric field over the QD volume. The approach enables us to solve the problem considering fields only inside the QD. Moreover, it proves to

be possible to examine separately, to a certain extent, the electromagnetic field and the particles. On the other hand, the simplification restricts the analysis to the strong confinement regime; the theory should be drastically modified to include inhomogeneity and nonlocality into consideration.

In the framework of the above stated approximation, the system "QD + electromagnetic field" is described by the Hamiltonian $H = H_0 + H_{IL}$, where $H_0 = \epsilon_e a_e^\dagger a_e + \epsilon_g a_g^\dagger a_g$ is the Hamiltonian of the carriers motion, $\epsilon_{g,e}$ are the energy eigenvalues, $a_{g,e}^\dagger$ and $a_{g,e}$ stand for the creation and annihilation operators (here and below indices e and g correspond to the excited and ground states of electron, respectively). These operators satisfy the anticommutative relations usual for fermions. The term H_{IL} describes interaction with the electromagnetic field. In this paper we use a 3D Cartesian coordinate system $\mathbf{u}_i$ $(i = x, y, z)$ 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 the term H_{IL} takes the form as follows:

$$H_{IL} = -V \hat{P}_x E_{Lx}\,, \tag{1}$$

where $\hat{P}_x = V^{-1}(-\mu b^\dagger + \mu^* b)$ is the polarization operator, the operators $b^\dagger = a_g a_e^\dagger$ and $b = a_g^\dagger a_e$ are the creation and annihilation operators for electron-hole pairs, V is the QD volume. Thus, we define the light-matter interaction Hamiltonian in the dipole approximation [18], i.e., we reject a negligibly small term proportional to $\mathbf{A}^2$. Such an approximation is valid, at least, in the vicinity of the exciton resonance [19]). Here and below we mark operators by the label "$\hat{}$" if it is necessary to distinguish them from their macroscopically averaged values denoted by the same letters. We use underlined letters to mark tensors. Note that our model also describes higher excitonic modes; in that case operators $b^\dagger$ and b move up the exciton into the next energy level and return it back.

The field inside QD, $\mathbf{E}_L$, involved in Eq. (1), is different from the external acting field $\mathbf{E}_0$(see Fig. 1). Since we postulate the QD to be electrically small, and, as consequence, the field inside QD to be homogeneous, this difference is determined by the depolarization field [20]. To evaluate the depolarization field, we start with the frequency–domain integral relation defined inside the QD [21],

$$\mathbf{E}_L(\mathbf{r}) = \mathbf{E}_0(\mathbf{r}) + (\nabla\nabla\cdot + k^2) \int\limits_V \mathbf{P}(\mathbf{r}') G(\mathbf{r} - \mathbf{r}')\, d^3\mathbf{r}' \tag{2}$$

which follows from the Maxwell equations. Here $G(\mathbf{r}) = \exp(ik|\mathbf{r}|)/4\pi|\mathbf{r}|$ is the vacuum Green function, $\mathbf{P} = \langle \hat{\mathbf{P}} \rangle$ is the macroscopic polarization. This relation couples the local $\mathbf{E}_L(\mathbf{r})$ and the acting $\mathbf{E}_0(\mathbf{r})$ fields inside the QD. Letting the QD to be electrically small, we can neglect retardation in this

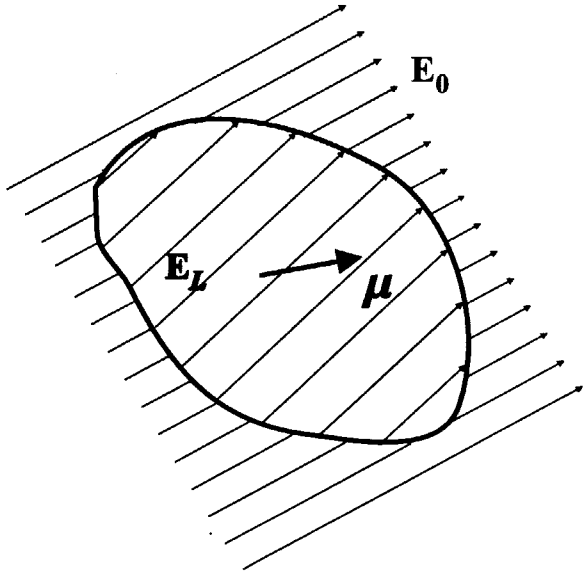

Figure 1. Correlation between acting and local fields in electrically small scatterer. Local field is assumed to be homogeneous.

equation and reduce it to

$$\mathbf{E}_L(\mathbf{r}) = \mathbf{E}_0(\mathbf{r}) + \nabla\nabla \cdot \int_V \mathbf{P}(\mathbf{r}') \frac{d^3\mathbf{r}'}{|\mathbf{r} - \mathbf{r}'|} \,. \tag{3}$$

Also, the above made approximation allows us to suppose the acting field and, consequently, the polarization $\mathbf{P}$ to be constant over the QD volume. As a result, Eq. (3) is transformed to

$$\mathbf{E}_L = \mathbf{E}_0 - 4\pi \underline{\mathbf{N}} \mathbf{P} \,. \tag{4}$$

Here $\underline{\mathbf{N}}$ is the depolarization tensor with its components defined by

$$N_{\alpha\beta} = -\frac{1}{4\pi} \frac{\partial^2}{\partial r_\alpha \partial r_\beta} \int_V \frac{d^3\mathbf{r}'}{|\mathbf{r} - \mathbf{r}'|} \,, \tag{5}$$

where $r_\alpha = x, y, z$. This tensor is symmetrical [21] and depends only on the shape of the scattering object, i.e., QD. For instance, for a sphere $\underline{\mathbf{N}} = \underline{\mathbf{I}}/3$. For an spheroid the tensor $\underline{\mathbf{N}}$ is diagonal in a basis related to the spheroid's axes [20]:

$$N_{ZZ} = \frac{e^2 + 1}{e^3} (e - \arctan e) \,, \quad N_{XX} = N_{YY} = \frac{1}{2} (1 - N_{ZZ}) \,, \tag{6}$$

where $e = \sqrt{a_{el}^2/b_{el}^2 - 1}$ is the spheroid eccentricity, a_{el} and b_{el} are the spheroid semiaxes in the $X0Y$ plane and the Z-direction, respectively. These formulae hold true for both disklike ($a_{el} > b_{el}$) and cigarlike spheroids ($a_{el} < b_{el}$). Infinite stretching of the spheroid ($a_{el}/b_{el} \to 0$) results in $N_{ZZ} \to 0$, $N_{XX} \to 1/2$ and Eq. (6) reproduce the polarizabilities of the cylinders (see, e.g., [8]). It should be noted that for an arbitrary three-axis ellipsoid, the tensor $\underline{\mathbf{N}}$ does not depend on the coordinates. Consequently, the local field $\mathbf{E}_L(\mathbf{r})$ is also constant over the QD volume. For non-ellipsoidal QDs, the tensor $\underline{\mathbf{N}}$ and, thus, the local field become spatially inhomogeneous what contradicts to the basic assumption used under formulation of the Hamiltonian. To eliminate the contradiction, we should average relation (4) over the QD volume. This leads us again to Eq. (4) with $\mathbf{E}_L$, $\mathbf{E}_0$, $\mathbf{P} = \text{const}$ and

$$N_{\alpha\beta} = -\frac{1}{4\pi V} \iint\limits_{V\,V} \frac{\partial^2}{\partial r_\alpha \partial r_\beta} \frac{d^3\mathbf{r} d^3\mathbf{r}'}{|\mathbf{r} - \mathbf{r}'|} \,. \tag{7}$$

Returning to the co-ordinate system related to the QD dipole momentum, Eq. (4) can be rewritten by

$$E_{Lx} = E_{0x} - 4\pi N_x P_x \,, \tag{8}$$

where the depolarization coefficient N_x is as follows: $N_x = (\boldsymbol{\mu} \cdot \underline{\mathbf{N}}\boldsymbol{\mu})/|\mu|^2 \equiv (\mathbf{u}_x \cdot \underline{\mathbf{N}}\mathbf{u}_x)$. In view of the above consideration, the total Hamiltonian is represented by

$$H = H_0 + H_{I0} + \Delta H \,, \tag{9}$$

where

$$H_{I0} = -V\widehat{P}_x E_{0x} \tag{10}$$

and

$$\begin{aligned} \Delta H &= 4\pi N_x P_x(-\mu b^\dagger + \mu^* b) \\ &= \frac{4\pi}{V} N_x(\mu^* b - \mu b^\dagger)(\mu^* \langle b\rangle - \mu\langle b^\dagger\rangle) \,. \end{aligned} \tag{11}$$

Thus, in the total Hamiltonian we have separated contribution of the interaction of electron-hole pairs with acting field, H_{I0}, from contribution of depolarization, ΔH. Such a separation allows us to include the local field effects into consideration without explicit solution of the electrodynamical boundary-value problem. This is of special importance when we come to the quantization of the electromagnetic field. Note that the quantity ΔH is expressed in terms of dynamic variables of the particle motion. Thus, coefficient N_x contains complete information about electromagnetic interaction.

2.2. EQUATIONS OF MOTION

Let $|\tilde{\psi}(t)\rangle$ be the wavefunction of the system "QD + electromagnetic field". In the interaction representation this system is described by the Schrödinger equation

$$i\hbar\frac{\partial|\psi\rangle}{\partial t} = H_{int}|\psi\rangle \tag{12}$$

with $H_{int} = \exp(iH_0t/\hbar)(H_{I0} + \Delta H)\exp(-iH_0t/\hbar)$ and $|\psi(t)\rangle = \exp(iH_0t/\hbar)|\tilde{\psi}(t)\rangle$. We represent then $|\psi(t)\rangle$ by the sum as follows

$$|\psi(t)\rangle = A(t)|e\rangle + B(t)|g\rangle\,,$$

where $A(t)$ and $B(t)$ are coefficients to be found, $|g\rangle$ and $|e\rangle$ are the wavefunctions of QD in ground and excited states, respectively. Taking into account the well-known identities $b^\dagger|e\rangle = b|g\rangle = 0$ and $b|e\rangle = |g\rangle$, $b^\dagger|g\rangle = -|e\rangle$, from Eq. (12) we obtain the set of equations of motion

$$\begin{aligned} i\hbar\frac{\partial A}{\partial t} &= (4\pi N_x P_x - E_{0x})\mu B e^{i\omega_0 t}\,,\\ i\hbar\frac{\partial B}{\partial t} &= (4\pi N_x P_x - E_{0x})\mu^* A e^{-i\omega_0 t}\,, \end{aligned} \tag{13}$$

with macroscopic polarization determined by

$$P_x = \langle\tilde{\psi}|\hat{P}_x|\tilde{\psi}\rangle = \frac{1}{V}\mu^* A(t)B^*(t)e^{-i\omega_0 t} + \text{c.c.}\,. \tag{14}$$

Further we restrict ourselves to the slow-varying amplitude approximation. For that aim, we present the acting field by $E_{0x} = \mathrm{Re}[\mathcal{E}(t)\exp(-i\omega t)]$ with $\mathcal{E}(t)$ as a slow-varying amplitude. Then, taking relation (14) into account and neglecting the fast-oscillating terms in Eq. (13), we derive final expressions for equations of motion:

$$\begin{aligned} i\hbar\frac{\partial A}{\partial t} &= \hbar\Delta\omega A|B|^2 - \frac{1}{2}\mathcal{E}(t)\mu B e^{i(\omega_0-\omega)t}\,,\\ i\hbar\frac{\partial B}{\partial t} &= \hbar\Delta\omega B|A|^2 - \frac{1}{2}\mathcal{E}^*(t)\mu^* A e^{-i(\omega_0-\omega)t}\,, \end{aligned} \tag{15}$$

where

$$\Delta\omega = \frac{4\pi}{\hbar V}N_x|\mu|^2\,. \tag{16}$$

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 (15) is analogous to the Bloch equations

for optically dense media derived in Ref. [22]. The relaxation can easily be included into Eqs. (15) either by introduction of the phenomenological transverse and longitudinal relaxation times [22] or by corresponding modification of initial Hamiltonian (9).

2.3. POLARIZABILITY OF A SINGLE QD IN THE STRONG CONFINEMENT REGIME

The case of excited QD can be analyzed using equations (15) with the initial conditions $A(0) = 1$ and $B(0) = 0$ imposed. In linear approximation with respect to electromagnetic field we can put $A(t) \approx 1$. Physically, this means that we restrict the analysis to temporal intervals essentially less than the relaxation time of the given resonant state. In such a situation, Eqs. (15) are simplified and reduced to

$$i\hbar\frac{\partial B}{\partial t} = \hbar\Delta\omega B - \frac{1}{2}\mathcal{E}^*(t)\mu^* e^{i(\omega_0-\omega)t}\,. \tag{17}$$

For time-harmonic excitation, i.e., for $\mathcal{E}(t) = \mathcal{E}$ =const, this equation is exactly integrable:

$$B(t) \approx -\frac{\mathcal{E}^*\mu^*}{2\hbar(\omega_0 - \Delta\omega - \omega)}\left[e^{-i(\omega_0-\omega)t} - e^{-i\Delta\omega t}\right] \tag{18}$$

with $\Delta\omega$ determined by Eq. (16). Thus, one can see that the local fields (depolarization) leads to the shift $\Delta\omega$ of the resonant frequency. This shift was predicted in a number of papers [23, 24, 10] on the basis of different phenomenological models. In Refs. [8, 9] it has been predicted and experimentally verified that this shift in anisotropically shaped QDs provides polarization splitting of the gain band. Note also that the depolarization effect has been proposed by Gammon *et al.* [25] as a hypothesis explaining the experimentally observed polarization-dependent splitting of the PL spectrum of single anisotropically shaped QD.

Eq. (16) is identical to that obtained in Refs. [8, 9]. In order to demonstrate it we should make a substitution $|\mu|^2 \to |\mu_0|^2/3$ where μ_0 is the matrix element of the dipole moment of a corresponding bulk sample (coefficient $1/3$ is appeared as a result of orientational averaging in bulk samples). We should also take into account the spin degeneracy of electron-hole pairs which results in duplication of $\Delta\omega$. This is because the total polarization of the system is provided by superposition of two partial polarizations corresponding to two spin components. Then, expressing macroscopic polarization in terms of $B(t)$, we find

$$P_x = \frac{1}{8\pi}\alpha_{xx}(\omega)\mathcal{E}\left[e^{-i\omega t} - e^{-i(-\Delta\omega+\omega_0)t}\right] + \text{c.c.}\,, \tag{19}$$

where

$$\alpha_{xx}(\omega) = \frac{4\pi|\mu|^2}{\hbar V(\omega + \Delta\omega - \omega_0)} \tag{20}$$

is the component of the QD polarizability tensor. Phenomenological consideration for QD modeled as single-resonance medium with the Lorentz dispersion $\varepsilon(\omega) = \varepsilon_h + g_0/(\omega - \omega_0)$ [8, 9] gives the same result if we put $g_0 = 4\pi|\mu|^2/\hbar V$. This means that the Hamiltonian defined by Eqs. (9)–(11) comprises that phenomenological model as a particular case.

For a ground-state QD, the initial conditions has the form as follows: $A(0) = 0$, $B(0) = 1$. Applying to this case the above presented procedure, we obtain

$$A(t) \approx \frac{\mathcal{E}\mu}{2\hbar(\omega_0 + \Delta\omega - \omega)} \left[e^{i(\omega_0 - \omega)t} - e^{-i\Delta\omega t}\right] . \tag{21}$$

Thus, for the ground state the local field effects manifest themselves in the same shift $\Delta\omega$ of the resonance but with the opposite sign. If we introduce now into consideration a finite radiation linewidth, interaction of a ground-state QD with the electromagnetic field corresponds to the 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 latter one.

2.4. HOMOGENIZATION PROCEDURE

In the strong confinement regime the homogenization procedure for excitonic composites is carried out in ordinary way with the account for the depolarization. Electromagnetic properties of composites are usually modeled in the framework of the effective-medium approach, which implies electromagnetic field averaging over material inhomogeneities. Thus, a homogeneous medium with effective constitutive parameters, such as conductivity, susceptibility, permittivity, instead a composite is being considered. The effective parameters are expressed in terms of the generic and the geometrical parameters of the inclusions and the host medium. The general approach for estimating the effective constitutive parameters of a composite material is as follows: First, the field scattered by a single inclusion in the host medium is found; then, the scattering contributions from all inclusions are summed and averaged over a vanishingly small region. For this approach to hold, all inclusions must be electrically small, i.e., their linear size must be small comparing with the wavelength. Having linear extension of the spatial inhomogeneity of the order of several nanometers, QD–based structures completely satisfy the above condition in the visible

range. This justifies applicability of the effective medium approach to the description of the electromagnetic response properties of QD arrays.

There is a large number of different modifications of the effective medium theory [7]. Among them, we choose the Maxwell Garnett approach as it is based on rigorous solution of the integral equations of macroscopic electrodynamics for composites with small volume fraction of inclusions ($f_V < 0.4-0.5$, [7]); i.e., a weak modification of the electronic spectrum and the gain of QD ensemble is assumed as compared to that of individual QD. In the framework of the Maxwell Garnett approach a composite medium comprising a regular ensemble of uniform in size, electrically small dielectric inclusions dispersed in a host dielectric material is characterized by the effective permittivity tensor as follows [26]:

$$\underline{\varepsilon}_{eff}(\omega) = \underline{\mathbf{I}} + f_V \underline{\boldsymbol{\alpha}}(\omega) \left[\underline{\mathbf{I}} - f_V \underline{\boldsymbol{\delta}} \underline{\boldsymbol{\alpha}}(\omega)\right]^{-1} , \tag{22}$$

where $\underline{\mathbf{I}}$ is the unit tensor, $\underline{\boldsymbol{\alpha}}(\omega)$ is the polarizability tensor of single QD with components defined by Eq. (20), and $\underline{\boldsymbol{\delta}}$ is the lattice tensor completely determined by geometry of the array. The notation $\underline{\boldsymbol{\delta}}\,\underline{\boldsymbol{\alpha}}$ stands for the inner tensor product, $(\underline{\boldsymbol{\delta}}\,\underline{\boldsymbol{\alpha}})^j_k = \delta^j_l \alpha^l_k$.

Eq. (22) states that the QD ensemble comprises an optically anisotropic medium even if both QD and host materials are isotropic. Thus, we predict *electromagnetic* anisotropy of the electromagnetic response in QD arrays due to diffraction of the electromagnetic field by inclusions. In Ref. [26] it has been shown that, assuming linear size of the lattice elementary cell to be much less than the wavelength, components of the lattice tensor are given by the integrals over the elementary cell volume Ω as follows:

$$\delta_{ij} = -\frac{3}{4\pi} \int\limits_{\Omega} \frac{r_i r_j}{r^5} d^3\mathbf{r} \quad \text{if} \quad i \neq j\,, \quad \text{and} \quad \delta_{ii} = -\frac{1}{4\pi} \int\limits_{\Omega} \frac{3r_i^2 - r^2}{r^5} d^3\mathbf{r}\,. \tag{23}$$

Here $\mathbf{r}$ is the radius vector of a point inside the elementary cell and $r = |\mathbf{r}|$; indices i and j stand for the Cartesian components x, y, z. Note that the lattice tensor (23) is found without any reference to specific properties of QDs as quantum-mechanical objects.

3. Excitonic composite in the weak confinement regime

3.1. POLARIZABILITY OF A SINGLE QD

Let us now study the role of nonlocality in electromagnetic response of an isolated QD. Again, we decompose the total Hamiltonian of the system "QD+electromagnetic field" into two parts, $H = H_0 + H_{IL}$, with H_{IL} responsible for the light-matter interaction. As different from the previous

case, in the weak confinement regime Coloumb interaction contributes to the Hamiltonian H_0. The Hamiltonian H_{IL} contains an external field averaged over the crystalline lattice of the QD material. Here we apply the Coloumb calibration for the the vector potential $\nabla \cdot \mathbf{A} = 0$ which implies electromagnetic field to be transverse. In that case, $H_{IL} = -i\mathbf{A}\mathbf{p}/mc$, where e, m are the electron charge and mass, respectively, $\mathbf{p} = -i\hbar\nabla$ is the momentum operator. The charge carrier motion in QD is described by the Liuwille equation $i\hbar\partial\rho/\partial t = [H, \rho]$ for the density matrix ρ, which we will solve to the linear approximation with respect to electromagnetic field. For complex–valued amplitudes one finds:

$$P_\alpha(\mathbf{r}) = \int_V \chi_{\alpha\beta}(\mathbf{r}, \mathbf{r}', \omega) E_\beta(\mathbf{r}') d^3\mathbf{r}', \tag{24}$$

where

$$\chi_{\alpha\beta}(\mathbf{r}, \mathbf{r}', \omega) = \left(\frac{e\hbar}{m\omega}\right)^2 \sum_{p,n} (\rho_{0n} - \rho_{0p}) \left[\frac{1}{\hbar\omega + \epsilon_n - \epsilon_p} - \frac{1}{\hbar\omega - \epsilon_n + \epsilon_p}\right]$$
$$\times \Psi_p^*(\mathbf{r}) \nabla_\alpha \Psi_n(\mathbf{r}) \Psi_n^*(\mathbf{r}') \nabla'_\beta \Psi_p(\mathbf{r}'), \tag{25}$$

$\epsilon_{n,p}$ and $\Psi_{n,p}(\mathbf{r})$ stand for the eigenvalues and eigenfunctions of the Hamiltonian H_0, respectively, while quantities $\rho_{0n,p}(\mathbf{r})$ are the eigenvalues of the unperturbed density matrix.

Let us estimate contribution of excitons to the QD polarizability. For that aim, we put $\rho_{0n} = 0$ and $\rho_{0p} = 1$ into Eq. (25) assuming thus that n–th state is an exciton while p–th one is the ground state. Exciton and ground–state wavefunctions are as follows [27]

$$\Psi_n = \sum_{j',j} F_j(\bar{\mathbf{j}}) \Phi(\mathbf{j}_r) W_j^c(\mathbf{r}_j) \prod_{i \neq j} W_i^v(\mathbf{r}_i), \qquad \Psi_p = \Psi_g = \prod_i W_i^v(\mathbf{r}_i), \tag{26}$$

with $W_j^{c,v}(\mathbf{r}_j)$ as the Wannier functions of the conduction and valence bands, respectively. The function $\Phi(\mathbf{j}_r)$ describes the electron–hole relative motion with $\mathbf{j}_r \equiv j - j'$ while $F_j(\bar{\mathbf{j}})$ characterizes the center–of–mass motion of the exciton with $\bar{\mathbf{j}} \equiv (m_e j + m_h j')/(m_e + m_h)$; $m_{e,h}$ are the electron and hole effective masses, respectively.

Carrying out averaging over the infinitesimal volume in the atomic scale, from Eq. (24) one can obtain

$$\langle \mathbf{P}^{ex}(\mathbf{r}) \rangle = \sum_n (\boldsymbol{\mu} \otimes \boldsymbol{\mu}) \Phi^2(0) \left[\frac{1}{\hbar\omega + \epsilon_n - \epsilon_g} - \frac{1}{\hbar\omega - \epsilon_n + \epsilon_g}\right]$$
$$\times F_n(\mathbf{r}) \int_V F_n(\mathbf{r}') \mathbf{E}(\mathbf{r}') d^3\mathbf{r}', \tag{27}$$

where the symbol $\otimes$ marks a diadic product of vectors. If QD comprises an isotropic material, orientational averaging in it leads to the change $(\boldsymbol{\mu} \otimes \boldsymbol{\mu}) \rightarrow |\boldsymbol{\mu}_0|^2 \delta_{ij}/3$, where δ_{ij} is the Kronecker symbol. In that case, anisotropy of the QD electromagnetic response arises as a result of anisotropy of the QD shape (or, in composites, as result of anisotropy of array).

Consider scattering of an initial electromagnetic field $\mathbf{E}_0(\mathbf{r})$ by an isolated QD assuming the background permittivity of the QD to be equal to the permittivity of the host medium ε_h. Thus, we neglect the image-potential effects. For the analysis we make use again Eqs. (2)–(3) with the substitution $k \rightarrow k_1 = k\sqrt{\varepsilon_h}$. However, unlike the strong confinement regime, now polarization of QD is related to the local electric field $\mathbf{E}_L(\mathbf{r})$ by means of a linear integral operator. Restricting ourselves to the exciton component of polarization, one come to the following relation [16]

$$\mathbf{P}(\mathbf{r}) \approx DF(\mathbf{r})\boldsymbol{\Lambda}\,, \tag{28}$$

where

$$D \approx \frac{2}{3}|\boldsymbol{\mu}|^2 \frac{\Phi^2(0)}{\hbar\omega + \epsilon - \epsilon_g}\,, \qquad \boldsymbol{\Lambda} = \int\limits_V F(\mathbf{r})\mathbf{E}_L(\mathbf{r})d^3\mathbf{r}\,. \tag{29}$$

Index n of the excitonic mode in the coefficient D is omitted for simplicity. Multiplier 2 in the expression for D takes into account spin degenerating of excitonic modes. Note that Eq. (28) defines very special type of nonlocality: the integral operator in it has degenerated kernel with degeneration order equal to unit. In view of that, integral differential equations (2) and (3) turn out to be equivalent to the integral Fredholm equations with degenerated kernels. For arbitrary degenerating order, such equations are reduced to systems of algebraic equations [28]; in our case, presence of a degenerated kernel makes possible analytical consideration of the nonlocality problem. First, Eq. (3) allows us to find vector $\boldsymbol{\Lambda}$ omitting the procedure of evaluation of the local electromagnetic field $\mathbf{E}_L(\mathbf{r})$ inside QD; to do this, let us multiply Eq. (3) by the function $F(\mathbf{r})$ and integrate it over the QD volume. Then, solving Eq. (2) by the method presented in Ref. [12], we derive the expression for the Hertz potential in the far zone:

$$\boldsymbol{\Pi}^e = \frac{V}{4\pi r} e^{ik_1 r} \underline{\boldsymbol{\alpha}} \mathbf{E}_0\,, \tag{30}$$

where polarizability tensor of an isolated QD $\underline{\boldsymbol{\alpha}}$ is expressed by

$$\underline{\boldsymbol{\alpha}} = \frac{4\pi}{V} DM^2 \left(\underline{\mathbf{I}} - D\underline{\mathfrak{F}}\right)^{-1}\,, \tag{31}$$

with $M = \int_V F(\mathbf{r})d^3\mathbf{r}$. The tensor $\underline{\mathfrak{F}}$, which describes the role of the QD depolarization, is given by its components as

$$\mathfrak{F}_{\alpha\beta} = 4\pi \int_V \int_V F(\mathbf{r}')F(\mathbf{r})\frac{\partial^2}{\partial r_\alpha \partial r_\beta} G(\mathbf{r}-\mathbf{r}')d^3\mathbf{r}'d^3\mathbf{r}\,. \tag{32}$$

Using the Puasson equation for the Green function $G(\mathbf{r})$, we obtain the important normalization condition

$$\mathfrak{F}_{xx} + \mathfrak{F}_{yy} + \mathfrak{F}_{zz} = 4\pi \int_V F^2(\mathbf{r})d^3\mathbf{r}\,. \tag{33}$$

This equality can be used to control the accuracy of numerical integration of complicated expressions (32).

Thus, we have shown that the special law of the nonlocality (28) inherent to an isolated QD admits description of the electromagnetic field scattering by the QD using the polarizability tensor independent on the incident field structure. In other words, the nonlocality changes values of the polarizability tensor components but does not change the general representation of the scattering operators as compared to the strong confinement regime.This result allows extension of the Maxwell Garnett approach to 3D composites constituted by QDs in the weak confinement regime.

3.2. EFFECTIVE CONSTITUTIVE RELATIONS FOR EXCITONIC COMPOSITES

Let the composite comprises a regular 3D lattice of identical QDs with $\mathbf{h}_\tau$ as the lattice vector. As before, the starting point of the analysis is the integral equations for the electric field inside the QD:

$$\mathbf{E}_L(\mathbf{r}+\mathbf{h}_\tau) = \mathbf{E}_0^{(\tau)}(\mathbf{r}+\mathbf{h}_\tau) + (\nabla\nabla\cdot + k_1^2)\mathbf{\Pi}_\tau^e(\mathbf{r})\,, \tag{34}$$

where

$$\mathbf{\Pi}_\tau^e(\mathbf{r}) = D\mathbf{\Lambda}_\tau \int_V \frac{e^{ik_1|\mathbf{r}-\mathbf{h}_\tau-\mathbf{r}'|}}{|\mathbf{r}-\mathbf{h}_\tau-\mathbf{r}'|} F(\mathbf{r}')d^3\mathbf{r}'\,, \tag{35}$$

$$\mathbf{E}_0^{(\tau)}(\mathbf{r}+\mathbf{h}_\tau) = \mathbf{E}_0(\mathbf{r}+\mathbf{h}_\tau) + \sum_\nu{}' (\nabla\nabla\cdot + k_1^2)\mathbf{\Pi}_\nu^e(\mathbf{r})\,, \tag{36}$$

prime in the summation excludes the term with $\nu = \tau$, $\mathbf{E}_0(\mathbf{r})$ is the incident field. The quantity $\mathbf{\Lambda}_\tau$ is given by

$$\mathbf{\Lambda}_\tau = \int_V F(\mathbf{r}+\mathbf{h}_\tau)\mathbf{E}_L(\mathbf{r}+\mathbf{h}_\tau)d^3\mathbf{r} = \int_V F(\mathbf{r})\mathbf{E}_L(\mathbf{r}+\mathbf{h}_\tau)d^3\mathbf{r}\,.$$

Under derivation of this equation we made use the property of the excitonic wavefunction of the composite to be periodical $F(\mathbf{r}+\mathbf{h}_\tau)=F(\mathbf{r})$. We also neglect overlapping of excitonic wavefunctions. Then we come to the infinite system of integral equations for infinite set of lattice vectors $\mathbf{h}_\tau$.

Electromagnetic field outside the QD, in the region $\mathbf{r}\notin V$, is presented by:

$$\begin{aligned}\mathbf{E}(\mathbf{r}) &= \mathbf{E}_0+\sum_\nu(\nabla\nabla\cdot+k_1^2)\mathbf{\Pi}_\nu^e(\mathbf{r})\,,\\ \mathbf{H}(\mathbf{r}) &= \mathbf{H}_0-ik_1\varepsilon_h\sum_\nu\nabla\times\mathbf{\Pi}_\nu^e(\mathbf{r})\,,\end{aligned}\tag{37}$$

where

$$\mathbf{\Pi}_\nu^e(\mathbf{r})=DM\frac{e^{ik_1|\mathbf{r}-\mathbf{h}_\nu|}}{|\mathbf{r}-\mathbf{h}_\nu|}(\underline{\mathbf{I}}-D\underline{\mathfrak{F}})^{-1}\mathbf{\Lambda}_\nu=\frac{V}{4\pi M}\frac{e^{ik_1|\mathbf{r}-\mathbf{h}_\nu|}}{|\mathbf{r}-\mathbf{h}_\nu|}\underline{\boldsymbol{\alpha}}\mathbf{\Lambda}_\nu\,.$$

No we can apply the Maxwell Garnett procedure to the excitonic composite. For that aim, we introduce mean fields by averaging of microscopic fields over the elementary cell volume Ω. For mean fields we obtain expressions analogous to expressions (37) with

$$\mathbf{\Pi}_\nu^e(\mathbf{r})\to\widetilde{\mathbf{\Pi}}_\nu^e(\mathbf{r})=\frac{f_V}{4\pi M}\underline{\boldsymbol{\alpha}}\mathbf{\Lambda}_\nu\int\limits_\Omega\frac{\exp\{ik_1|\mathbf{r}-\mathbf{h}_\nu-\boldsymbol{\eta}|\}}{|\mathbf{r}-\mathbf{h}_\nu-\boldsymbol{\eta}|}d^3\boldsymbol{\eta}\,.\tag{38}$$

To complete the procedure we should couple the mean field with the local one acting inside the QD. We found

$$\mathbf{E}_L(\mathbf{r}+\mathbf{h}_\nu)=\langle\mathbf{E}(\mathbf{r}+\mathbf{h}_\nu)\rangle-\frac{f_V}{4\pi M}\left(\nabla\nabla\cdot\int\limits_\Omega\frac{1}{|\mathbf{r}-\boldsymbol{\eta}|}d^3\boldsymbol{\eta}\right)\underline{\boldsymbol{\alpha}}\,\mathbf{\Lambda}_\nu\,.\tag{39}$$

Multiplying this equation by $F(\mathbf{r})$, after integration over the QD volume we derive the equality

$$\mathbf{\Lambda}_\nu=M\,(\underline{\mathbf{I}}-f_V\underline{\boldsymbol{\delta}}\,\underline{\boldsymbol{\alpha}})^{-1}\,\langle\mathbf{E}(\mathbf{h}_\nu)\rangle\,,\tag{40}$$

where $\underline{\boldsymbol{\delta}}$ is the tensor given by its components:

$$(\underline{\boldsymbol{\delta}})_{ij}=-\frac{1}{4\pi M}\int\limits_V\int\limits_\Omega F(\mathbf{r})\frac{\partial^2}{\partial r_i\partial r_j}\frac{1}{|\mathbf{r}-\boldsymbol{\eta}|}d^3\boldsymbol{\eta}d^3\mathbf{r}\,.\tag{41}$$

Further we follow to the conventional procedure of the electrodynamics of composite media: Eq. (40) obtained for a discrete set of points $\mathbf{r}=\mathbf{h}_\nu$ is extended to all space. After that, this equation presents a generalization of the Mossotti–Clausius factor to the excitonic composite. Peculiar property

of such composites is that the relation between mean and acting fields is nonlocal. Note that the condition $k\Omega^{1/3} \ll 1$ allows us the change

$$\tilde{\Pi}^e_N(\mathbf{r}) \to \frac{1}{4\pi M} f_V \underline{\alpha} \int\limits_{\Omega} \mathbf{\Lambda}(\mathbf{h}_\nu + \boldsymbol{\eta}) \frac{\exp\{ik_1|\mathbf{r} - \mathbf{h}_\nu - \boldsymbol{\eta}|\}}{|\mathbf{r} - \mathbf{h}_\nu - \boldsymbol{\eta}|} d^3\boldsymbol{\eta}\,.$$

In this case, macroscopic polarization of the excitonic composite is expressed by $\mathbf{P} = (\underline{\varepsilon}_{eff}/\varepsilon_h - 1)\langle \mathbf{E} \rangle / 4\pi$ with $\underline{\varepsilon}_{eff}$ determined by equation (22) and tensors $\underline{\alpha}$ and $\underline{\delta}$ given by Eqs. (31) and (41), respectively. This allows us to conclude that the excitonic composite in the weak confinement regime is equivalent to a homogeneous anisotropic medium with the tensorial dielectric function $\underline{\varepsilon}_{eff}$ and unit permeability.

4. Cubic lattice of spherical QDs: comparison of the weak and strong confinement regimes

Let a collection ofv uniform spherical QDs of the radius R_0 constitutes a cubic lattice. Exciton wavefunction for an isolated spherical QD is as follows (see, e.g., [23]):

$$F(\mathbf{r}) \equiv F_{nlm}(\mathbf{r}) = C_{nl} Y_{lm}(\theta, \varphi) J_{l+1/2}\left(\kappa_{nl} \frac{\rho}{R_0}\right) \frac{1}{\sqrt{\rho}},$$

where $Y_{lm}(\theta, \varphi)$ is the spherical harmonics, $J_\nu(x)$ is the Bessel function, ρ, θ, φ are the spherical co-ordinates, κ_{nl} is the n-th root of the Bessel function $J_{l+1/2}(x)$, indices n and l define the working mode in the oscillator spectrum. Coefficient $C_{nl} = \sqrt{2}[R_0 J_{l+3/2}(\kappa_{nl})]^{-1}$ provides orthonormalization of functions F_{nlm}. Further we restrict ourselves to the case $l = m = 0$; in this case $Y_{00}(\theta, \varphi) = 1/2\sqrt{\pi}$. Let the dipole moment of the QD is oriented along the x-axis; in this case polarizability tensor is characterized by the xx-component. By integration in Eqs. (32) we obtain $M^2 = 32\pi R_0^3/\kappa_{n0}^2$, $\Im_{xx} = 4\pi/3$. Since for a sphere all directions are equivalent, identical results are obtained for zz - and yy-components. It can easily be shown that the components satisfy the equality (33). Choosing excitonic mode with another set of numbers n, l, m we obviously obtain another magnitude of components $\Im_{\alpha\alpha}$ and, consequently, another magnitude of the polarizability tensor $\underline{\alpha}$ (31). Thus, we come to peculiar property of the weak confinement regime: depolarization depends on the excitonic mode number. This property follows from the nonlocality of the exciton polarization. Unlike that, in the strong confinement regime depolarization tensor (5) is completely determined by the QD geometry.

For the particular case of the cubic lattice, a special approach based on the the trace theorem $(\underline{\delta})_{xx} + (\underline{\delta})_{yy} + (\underline{\delta})_{zz} = 1$ can be applied. Taking into

account isotropy of cubic lattice we find $(\underline{\delta})_{xx} = (\underline{\delta})_{yy} = (\underline{\delta})_{zz} = \delta = 1/3$. This result, together with the previous ones, allows us to evaluate depolarization shift of the exciton resonant frequency in the weak confinement regime:

$$\Delta\omega = -\frac{4}{9}\left(\frac{R_0}{a_B}\right)^3 g_0 \,. \tag{42}$$

The formula $\Phi(r) = (1/\pi a_B^3)^{1/2}\exp(-r/a_B)$ [27] has been utilized under derivation of the shift; a_B is the exciton Bohr radius. This shift is different from that given by Eq. (16) for spherical QDs in the strong confinement regime: $\Delta\omega = -g_0/3\varepsilon_h$. Thus we state another peculiar property of the weak confinement regime: radial dependence of the depolarization shift.

5. Conclusion

In our paper we have developed effective medium theory of 3D inhomogeneous semiconductor heterostructure — excitonic composite, constituted by QDs imbedded in a transparent host medium. We have started with the self-consistent microscopic local-field theory for an isolated arbitrary shaped QD. In our approach we introduce into Schrödinger equation 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 of 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.

Both types of the exciton localization, strong and weak confinement regimes have been considered. Pronounced nonlocality is characteristic of the latter regimes. The basic result is that in both cases excitonic composite is described by spatially local constitutive relations although electromagnetic response of an isolated QD in the weak confinement regime is spatially nonlocal. The nonlocality manifests itself in specific dependence of the effective constitutive parameters on the geometry and electronic properties of single QD.

To conclude, let us dwell on possible ways of the further development of the theory presented. In our consideration we have restricted ourselves to a simplest model of the excitonic composite: perfect lattice of identical QDs, whereas experimentally available structures show intrinsic size dispersion and periodicity violation. These effects may be characterized by a distribution function and taken into account by a theory of irregular

excitonic composites. Elaboration of such a theory is the topical question. Another essential restriction of the analysis presented is that nonclassical light-matter interaction has been left beyond the analysis. Quantum nature of the light interacting with an excitonic composite is expected to be of importance for many problems where collections of excitons are involved: quantum computing, electromagnetic fluctuations, *etc.* In general, quantum electrodynamics provides necessary formalism for investigation of the problem. However, since QDs are electrically small inhomogeneities with inherent energy dissipation (absorption or gain) and dispersion, canonical quantization scheme of the electromagnetic field becomes invalid: dissipation results in that the operators corresponding to the Maxwell equations turn out to be non-Hermitian. To avoid this difficulty, a quantization scheme which involves auxiliary fields has been proposed in Ref. [18]. The theory elaborated in our paper allows synthesis with that quantization scheme and, thus, creates necessary basis for investigation of quantum optics of excitonic composites.

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