

Effective boundary conditions for planar quantum dot structures

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The effective-boundary condition method is extended to nanoscale mesoscopic systems. The (EBC's) appear as a result of the two-dimensional (2D)-homogenization procedure and have the form of two-side anisotropic impedance boundary conditions stated on the structure surface. It has been shown that, unlike to macroscopic electrodynamics, the surface impedance tensor exhibits sharp oscillations at frequencies of optical transitions. The EBC method supplemented with well-developed mathematical techniques of classical electrodynamics creates unified basis for solution of boundary-value problems in electrodynamics of nanostructures. We have shown that the radiative lifetime of 2D array of spherical quantum dots (QD's) drastically changes its dependence on QD radius in comparison with the case of a single QD in the range of radii smaller than Bohr radius.

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I. 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 has motivated permanent extension of their study. Among a variety of new results in this field, the recent progress in the synthesis of sheets of nanoscale three-dimensional (3D) confined narrow-gap insertions in a host semiconductor, quantum dots (QD's), is of a special interest. Indeed, QD-based structures provide practical realization of the idea proposed by Dingle and Henry¹ 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 characteristics as compared to conventional quantum-well (QW) lasers.^{2,3} For $\text{In}_x\text{Ga}_{1-x}\text{As}$ QD's on GaAs substrates, an exceptionally bright luminescence at $1.36\ \mu\text{m}$ was realized at room temperature⁴ in a spectral range far beyond those available for conventional strained $\text{In}_x\text{Ga}_{1-x}\text{As}$ -GaAs QW's. 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.

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 (QD's) can be treated as *electrically small* objects and electromagnetic response of such heterogeneous structures, *composites*, can be evaluated by means of effective-medium theory.⁶ Application of effective-medium approach to 3D arrays of QD's has been presented in Refs. 7 and 8. In many cases, however, a planar array of QD's with intrinsic 2D periodicity of characteristic period much less than the optical wavelength, can be treated as more adequate and realistic model.⁵

In this paper we present a general method for evaluation of electromagnetic response of planar arrays of QD's. This method, conventionally referred to as the effective-boundary-condition (EBC) method, has been originally developed for microwaves and antenna theory,⁹⁻¹¹ and has

found a wide application in these fields. Similar approaches have also been developed in acoustics, hydrodynamics, elasticity theory. Recently, the EBC method has been extended to low-dimensional nanostructures, such as QW's,^{12,13} carbon nanotubes,¹⁴⁻¹⁶ and semicontinuous metal films.¹⁷ General outlook of the EBC method applications in electrodynamics of nanostructures has recently been reported in Ref. 18.

The basic idea of the EBC method is that a smooth homogeneous surface is considered instead of the initial structure, and appropriate EBCs for the electromagnetic field are stated for this surface. These conditions are chosen in such a way that the spatial structures of the electromagnetic field, due to an effective current induced on the homogeneous surface, and the electromagnetic field of the real current in the initial structure turn out to be identical at some distance away from the surface. Material characteristics of the structure as well as its geometrical parameters are included in coefficients of the EBC's. Such an approach is applicable both to continuous thin films⁹ and 2D periodical structures (semitransparent grid screens, helical sheaths in traveling wave tubes, etc.). The effect of periodicity is taken into account by the field averaging over the period. Thus, in essence, the EBC method is modification of the effective-medium theory as applied to planar structures. The applicability of the EBC's is restricted by the requirement that the lattice period is small compared with the wavelength in the host medium. The effectiveness of the EBC method is determined by a possibility of its extension to more complicated situations like finite-sized and/or deformed structures, structures located in the vicinity of additional reflectors and scatterers, etc. Such an extension is only possible when the parameters involved in the EBC's do not depend on the spatial structure of the irradiating field, or, in another words, the EBC's must be *local*, i.e., they must couple field components and their spatial derivatives at a given position on the boundary surface.

In Ref. 19 the dielectric properties of thin films consisting of a few layers of molecules or particles are considered and, by calculation of the effective permittivity tensor, it has been shown that in such structures the local-field effects exhibit in

rather different way than in 3D bulk medium. However, the solution of the specific electrodynamic problems based on these results is more complicated than by application of EBC's, as the latter decreases the number of boundaries between areas with different material parameters that require the field matching.

This paper is arranged as follows: In Sec. II, we formulate effective boundary conditions for planar nanostructures and derive an expression for the 2×2 surface conductivity tensor. The contribution of dielectric function nonlocality inherent in QD's at weak confinement of carriers is discussed in Sec. III. The analysis presented in Secs. II and III is applied in Sec. IV to estimate the radiative decay rate in planar QD array. The paper concludes with a discussion in Sec. V.

II. FORMULATION OF EFFECTIVE BOUNDARY CONDITIONS

In order to derive the EBC's, a kernel problem must be solved in each particular case. For example, for grid screens this problem is formulated as the problem of plane-wave scattering by the infinite plane screen.¹¹ In QD's, apart from the charge-carrier confinement,⁵ there exists a class of electrodynamic effects related to light diffraction by QD's and QD ensembles that strongly influences the electromagnetic response of such systems.^{7,8,21} Here we consider 2D arrays of QD's to establish correlation between properties of such systems and homogeneous 2D structures like quantum wells. The kernel problem here is the diffraction by infinite planar quadratic lattice constituted by identical QD's embedded in a host medium. The host medium is assumed to be dispersionless and transparent. As for the dispersion in QD, two regimes are differentiated by the carrier confinement. One of them, the strong confinement regime, is realized when the exciton Bohr radius a_B exceeds significantly the QD linear extension and, thus, Coloumb interaction between electron and hole is negligible. In opposite case (weak confinement regime), namely, Coloumb interaction forms exciton in QD providing strong nonlocality of the QD electromagnetic response. In the strong confinement regime, the well-known Lorentz dispersion law

$$\varepsilon(\omega) = \varepsilon_h + \frac{g_0}{\omega - \omega_0 + i/\tau}. \quad (1)$$

can be used as the simplest phenomenological model of dispersion in a single QD in the vicinity of the exciton resonance.^{22,23} In the above equation ω_0 is the resonant frequency of the transition; τ is the relaxation time (effective exciton dephasing time) in the QD; ε_h is the host medium permittivity; phenomenological parameter g_0 is given by $g_0 = -4\pi|\boldsymbol{\mu}|^2 W/3\hbar V$ (see, e.g., Ref. 22) with $\boldsymbol{\mu}$ as the matrix element of the dipole moment, W as the level population difference ($W < 0$ in an inverted medium), and V as the QD volume.

Further, we restrict ourselves to the dipole approximation of the diffraction theory assuming the lattice period d and QD size to be small as compared to the wavelength in the host medium and inside the QD; thus, QD's are assumed to

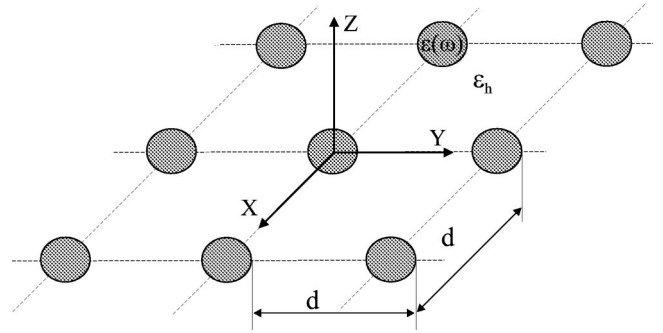


FIG. 1. Schematic 2D array of QD's.

be electrically small. In that case, electromagnetic field scattered from an isolated QD can be expressed in terms of Hertz potentials by

$$\begin{aligned} \mathbf{E}(\mathbf{r}) &= \mathbf{E}_0(\mathbf{r}) + \sum_{l,m=-\infty}^{\infty} [\nabla\nabla \cdot \boldsymbol{\Pi}_{lm}^e(\mathbf{r}) + k_1^2 \boldsymbol{\Pi}_{lm}^e(\mathbf{r})], \\ \mathbf{H}(\mathbf{r}) &= \mathbf{H}_0(\mathbf{r}) - ik\varepsilon_h \sum_{l,m=-\infty}^{\infty} \nabla \times \boldsymbol{\Pi}_{lm}^e(\mathbf{r}), \end{aligned} \quad (2)$$

where $k = \omega/c$, $k_1 = k\sqrt{\varepsilon_h}$, c is the speed of light, $\mathbf{E}_0(\mathbf{r})$, $\mathbf{H}_0(\mathbf{r})$ stand for the incident field; an $\exp(-i\omega t)$ time dependence is supposed. The incident field is assumed to be \mathbf{e}_x -polarized plane wave propagating at angle θ with respect to the z axis (see Fig. 1). Note that the polarization of the structure in the direction \mathbf{n} normal to the surface S , α_{zz} , should be included into consideration for QD's like spheres with comparable extensions in all directions. In that case,

$$\boldsymbol{\Pi}_{lm}^e(\mathbf{r}) = [\mathbf{e}_x \alpha_{xx} \mathcal{E}_x(\mathbf{R}_{lm}) + \mathbf{e}_z \alpha_{zz} \mathcal{E}_z(\mathbf{R}_{lm})] \frac{\exp(ik_1 \rho_{lm})}{\rho_{lm}}. \quad (3)$$

For QD's with planar configuration in the xy plane, e.g., discs, islands, flattened pyramids, etc., the QD polarizability in the z direction α_{zz} can be neglected to simplify significantly the further analysis. Here $\mathbf{R}_{lm} = \{ld, md, 0\}$ is the radius vector of QD in the lattice, $\rho_{lm} = |\mathbf{R}_{lm} - \mathbf{r}| = [(ld-x)^2 + (md-y)^2 + z^2]^{1/2}$, α_{ii} are the components of the QD polarizability tensor $\hat{\boldsymbol{\alpha}}$, and \mathcal{E} is the electric field inside QD. This field is related to the Hertz potentials by the equation analogous to Eqs. (2):

$$\mathcal{E}(\mathbf{r}) = \mathbf{E}_0(\mathbf{r}) + \lim_{\mathbf{r} \rightarrow 0} \sum_{l,m=-\infty}^{\infty'} [\nabla\nabla \cdot \boldsymbol{\Pi}_{lm}^e(\mathbf{r}) + k_1^2 \boldsymbol{\Pi}_{lm}^e(\mathbf{r})]. \quad (4)$$

Prime in this equation excludes the term with $l = m = 0$.

The next step in derivation of EBC's is the 2D averaging of the electromagnetic field in the $z=0$ plane. As it was mentioned above, such a procedure implies replacement of the discrete 2D elementary scatterer by a homogeneous $d \times d$ element of surface. By analogy with effective-medium theory for bulk composites,^{6,24} mathematically this procedure reduces summation in Eqs. (2) and (4) to integration, i.e.,

$$\sum_{l,m=-\infty}^{\infty} \{\dots\} \rightarrow \frac{1}{d^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \{\dots\} dx dy, \quad (5)$$

$$\sum_{l,m=-\infty}^{\infty'} \{\dots\} \rightarrow \frac{1}{d^2} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \{\dots\} dx dy - \frac{1}{d^2} \int_{-d/2}^{d/2} \int_{-d/2}^{d/2} \{\dots\} dx dy. \quad (6)$$

In view of rules (5) and the condition $k_1 d \ll 1$, combination of Eqs. (2) and (4) leads to the relation for electric field in an arbitrary lattice site \mathbf{R}_{lm} :

$$E_i^I(\mathbf{R}_{lm}) + E_i^{II}(\mathbf{R}_{lm}) = 2 \left(1 + \alpha_{ii} \frac{\delta_i}{d^2} \right) \mathcal{E}_i(\mathbf{R}_{lm}). \quad (7)$$

Although this equation has been derived for lattice sites, averaging procedure described above allows us to extend it over the whole plane $z=0$. The coefficients δ_i are given by the equations as follows:

$$\delta_x = \int_{-d/2}^{d/2} \int_{-d/2}^{d/2} \frac{2x^2 - y^2}{(x^2 + y^2)^{5/2}} dx dy \approx -\frac{8}{\sqrt{2}d}, \quad (8)$$

$$\delta_z = \lim_{z \rightarrow 0} \int_{-d/2}^{d/2} \int_{-d/2}^{d/2} \frac{3z^2 - r^2}{r^5} dx dy \approx \frac{2\pi\sqrt{\pi}}{d}, \quad (9)$$

where $r = (x^2 + y^2 + z^2)^{1/2}$. Equation for δ_y is obtained from Eq. (8) by the substitution $x \leftrightarrow y$; it is easy to show that $\delta_y = \delta_x$. It should be noted that the above integrals converge conditionally. For instance, in Eq. (8) result of integration depends on the order of integration (first, integration over x should be performed). In Eq. (8), interchanging integration and limiting transition is impermissible.

The different signs of $\delta_{x,y}$ and δ_z reveals the difference in the local field effects for the structures considered here and 3D medium. The same was found in Ref. 19, where the interaction diadic \hat{F}_{MM} was introduced with elements F_{MM}^{ii} analogous in the physical meaning to the coefficients δ_i . Herewith, the relation $\delta_{x,y}/\delta_z = F_{MM}^{xx,yy}/F_{MM}^{zz}$ takes place and, as one can see, this is valid with high accuracy for values $F_{MM}^{xx,yy} = 0.359$ and $F_{MM}^{zz} = -0.718$ obtained in Ref. 19.

Now one should find discontinuities of the mean field tangential components at $z=0$. Application of procedure (5) to Eq. (4) leads us to

$$\begin{aligned} H_y(\mathbf{r}) &= H_{0y}(\mathbf{r}) \\ &- \frac{i k \varepsilon_h}{d^2} \alpha_{xx} \frac{\partial}{\partial z} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{1}{\rho} \exp(ik_1 \rho) \mathcal{E}_x(\mathbf{R}) d^2 \mathbf{R} \\ &+ \frac{i k \varepsilon_h}{d^2} \alpha_{zz} \frac{\partial}{\partial x} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{1}{\rho} \exp(ik_1 \rho) \mathcal{E}_z(\mathbf{R}) d^2 \mathbf{R}, \end{aligned} \quad (10)$$

$$\begin{aligned} E_x(\mathbf{r}) &= E_{0x}(\mathbf{r}) + \frac{1}{d^2} \alpha_{xx} \left(\frac{\partial^2}{\partial x^2} + k_1^2 \right) \\ &\times \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{1}{\rho} \exp(ik_1 \rho) \mathcal{E}_x(\mathbf{R}) d^2 \mathbf{R} \\ &+ \frac{1}{d^2} \alpha_{zz} \frac{\partial^2}{\partial x \partial z} \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} \frac{1}{\rho} \exp(ik_1 \rho) \mathcal{E}_z(\mathbf{R}) d^2 \mathbf{R}, \end{aligned} \quad (11)$$

where \mathbf{R} is the radius vector in the xy plane and $\rho = |\mathbf{R} - \mathbf{r}|$. Note that the integrals involved into Eqs. (10) and (11) are the single-layer potentials for scalar Helmholtz equation.²⁰ For further consideration we take into account continuity of the incident field and single-layer potentials through the xy plane and discontinuity of the normal derivatives of these potentials on the surface.²⁰ Then, carrying out the limiting transition $z \rightarrow \pm 0$, we obtain:

$$E_x^I - E_x^{II} = -\frac{4\pi}{d^2} \alpha_{zz} \frac{\partial \mathcal{E}_z}{\partial x}, \quad (12)$$

$$H_y^I - H_y^{II} = \frac{4\pi i k \varepsilon_h}{d^2} \alpha_{xx} \mathcal{E}_x. \quad (13)$$

Superscripts I and II mark limiting values of corresponding quantities for $z \rightarrow +0$ and $z \rightarrow -0$, respectively. Corresponding equation for E_y is obtained from Eq. (12) by the changes $E_x \rightarrow E_y$ and $\partial/\partial x \rightarrow \partial/\partial y$. In order to derive equation for H_x , the changes $H_y \rightarrow H_x$, $E_x \rightarrow -E_y$ and $\alpha_{xx} \rightarrow \alpha_{yy}$ should be performed in Eq. (13). Substitution of \mathcal{E}_i determined by Eq. (7) into all four equations leads us to the EBC's in scalar representation. To present them in more convenient, covariant, notation we introduce the tangential vectors $-E_y \mathbf{e}_x + E_x \mathbf{e}_y \equiv \mathbf{n} \times \mathbf{E}$ and $H_x \mathbf{e}_x + H_y \mathbf{e}_y \equiv -\mathbf{n} \times \mathbf{n} \times \mathbf{H}$; then, combining the scalar EBC's by pairs, we come to the following covariant notation of the EBC's:

$$\mathbf{n} \times \mathbf{n} \times (\mathbf{H}^I - \mathbf{H}^{II}) = -\frac{2\pi}{c} \mathbf{n} \times \hat{\boldsymbol{\sigma}} (\mathbf{E}^I + \mathbf{E}^{II}), \quad (14)$$

$$\mathbf{n} \times (\mathbf{E}^I - \mathbf{E}^{II}) = -\xi \mathbf{n} \times \nabla [\mathbf{n} \cdot (\mathbf{E}^I + \mathbf{E}^{II})] \quad (15)$$

where

$$\hat{\boldsymbol{\sigma}} = \begin{pmatrix} \hat{\boldsymbol{\sigma}}_{\parallel} & 0 \\ 0 & 0 \end{pmatrix}.$$

The 2×2 surface conductivity tensor $\hat{\boldsymbol{\sigma}}_{\parallel}$ and the coefficient ξ are defined by

$$\hat{\boldsymbol{\sigma}}_{\parallel} = i \frac{\omega \varepsilon_h}{d^2} \hat{\boldsymbol{\alpha}}_{\parallel} \left(\hat{\mathbf{I}}_{\parallel} + \frac{\delta_x}{d^2} \hat{\boldsymbol{\alpha}}_{\parallel} \right)^{-1}, \quad \xi = \frac{2\pi \alpha_{zz}}{d^2 + \delta_z \alpha_{zz}}. \quad (16)$$

Here $\hat{\mathbf{I}}_{\parallel}$ is the 2×2 unit tensor and $\hat{\boldsymbol{\alpha}}_{\parallel}$ is given by the in-plane components α_{ij} ($i, j = x, y$) of the QD polarizability

tensor. Second term in the brackets in Eq. (16) is due to the depolarization related to the difference between mean and acting fields.

The equations (14)–(16) constitute the complete system of EBC's for electromagnetic field in low-dimensional nanostructures. They have been obtained in the ordinary way, by the averaging of a microscopic field over a physically infinitesimal volume. The technique of macroscopic averaging is similar to one that introduces the constitutive parameters for bulk media, but differs in that the averaging occurs in boundary conditions rather than in field equations. Correspondingly, the averaging was carried out over the 2D surface but not over the 3D spatial element. Thus, in electrodynamics of low-dimensional structures the EBC's play the same role as constitutive relations in electrodynamics of bulk media. Although the EBC's have been derived for 2D periodical structure with quadratic lattice, they keep validity for arbitrary configuration of elementary cell and for planar layers with random distribution of QD's. The difference will manifest itself in the modified coefficients δ_i . Since we did not concretize the incident field structure under derivation of EBC's (14) and (15), these boundary conditions hold true at arbitrary excitation (plane waves and wave beams, moving external charges, etc.). The only restriction is absence of external sources of electromagnetic field in the xy plane. This restriction allows us to assume \mathbf{E}_0 , \mathbf{H}_0 to be continuous through the plane. Note also that the EBC's (14) and (15) turn out to be analogous to corresponding EBC's for QW's Refs. 12 and 13 if spatial dispersion in the latter can be neglected, i.e., in the limit of infinitely large exciton mass. Thus, a planar layer comprising a 2D array of QD's can be treated as an effective QW. As a result, well-developed formalism of investigation of QW's Refs. 12,25–28 can be extended to QD arrays by introducing of effective integral parameters of the array defined by Eqs. (16).

For practical utilization of the derived EBC's the polarizability tensor requires to be known. For simplest configuration of QD's (sphere, disc) this tensor can be found analytically⁷ whereas direct numerical simulation is required for more complicated configurations like cubic or pyramidal. In particular, the minimal autonomic block method can be successfully applied for this purposes (see Ref. 8).

III. THE ROLE OF NONLOCALITY

EBC's (14) and (15) are valid for QD with strong confinement of carriers. Namely in this case the local model of the QD permittivity presented by Eq. (1) holds true; otherwise, in the case of weak confinement of carriers, the QD electromagnetic response becomes nonlocal: the constitutive relation for the QD medium polarization takes the form of the integral operator as follows:²⁹

$$\mathbf{P}(\mathbf{r}) = A \Phi(\mathbf{r}) \int_V \Phi(\mathbf{r}') \mathcal{E}(\mathbf{r}') d^3 \mathbf{r}', \quad (17)$$

where $A = \pi \varepsilon_h \omega_{LT} a_B^3 / (\omega_0 - \omega - i/\tau)$, the function $\Phi(\mathbf{r})$ is related to the envelope function of the exciton ground state,

ω_{LT} is the exciton longitudinal-transverse splitting in the bulk material. Integration in this equation is carried out over the QD volume V .

Let us now study the role of nonlocality presented by Eq. (17) in electromagnetic response of an isolated QD. We start with the integrodifferential wave equation that describes electromagnetic field both inside and outside the QD:

$$\mathbf{E}(\mathbf{r}) = \mathbf{E}_0(\mathbf{r}) + 4\pi(\nabla\nabla \cdot + k_1^2) \int_V \mathbf{P}(\mathbf{r}') G(\mathbf{r} - \mathbf{r}') d^3 \mathbf{r}'. \quad (18)$$

Here $G(\mathbf{r}) = \exp(ik_1|\mathbf{r}|)/4\pi|\mathbf{r}|$ is the Green function. In the far zone, electromagnetic field scattered by QD is characterized by the Hertz potential

$$\mathbf{\Pi}^e = \frac{e^{ik_1 r}}{r} \int_V \mathbf{P}(\mathbf{r}') d^3 \mathbf{r}' = \frac{e^{ik_1 r}}{r} A N \mathbf{\Lambda}, \quad (19)$$

where $N = \int_V \Phi(\mathbf{r}) d^3 \mathbf{r}$, $\mathbf{\Lambda} = \int_V \Phi(\mathbf{r}) \mathcal{E}(\mathbf{r}) d^3 \mathbf{r}$. Inside QD, the retardation can be neglected and, thus, Eq. (18) reduces to more simple form:

$$\mathcal{E}(\mathbf{r}) = \mathbf{E}_0(\mathbf{r}) + \nabla\nabla \cdot \int_V \frac{\mathbf{P}(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} d^3 \mathbf{r}'. \quad (20)$$

Note that Eq. (17) defines very special type of nonlocality: the integral operator in it has degenerated kernel with $M = 1$ degeneration order. In view of that, integral differential equations (18) and (20) turn out to be equivalent to the integral Fredholm equations with degenerated kernels. For arbitrary degeneration order, such equations reduce to systems of algebraic equations;³⁰ in our case, presence of a degenerated kernel makes possible analytical consideration of the nonlocality problem. First, Eq. (20) allows us to find vector $\mathbf{\Lambda}$ omitting the procedure of evaluation of the electromagnetic field $\mathcal{E}(\mathbf{r})$ inside QD; to do this, let us multiply Eq. (20) by the function $\Phi(\mathbf{r})$ and integrate it over the QD volume. As a result, we obtain

$$\mathbf{\Lambda} \approx N \mathbf{E}_0 + 4\pi A \hat{\mathbf{Y}} \mathbf{\Lambda}, \quad (21)$$

where the 3D-tensor $\hat{\mathbf{Y}}$ is given by its components by

$$Y_{\alpha\beta} = \frac{1}{4\pi} \int_V \int_V \Phi(\mathbf{r}) \Phi(\mathbf{r}') \frac{\partial^2}{\partial x_\alpha \partial x_\beta} \frac{1}{|\mathbf{r} - \mathbf{r}'|} d^3 \mathbf{r} d^3 \mathbf{r}' \quad (22)$$

with $x_{\alpha,\beta} = x, y, z$. Substitution of explicit expression for $\mathbf{\Lambda}$ obtained from Eq. (22) into Eq. (19) leads to the Hertz potential $\mathbf{\Pi}^e = \exp(ik_1 r) \hat{\boldsymbol{\alpha}} \mathbf{E}_0 / r$ with the polarizability tensor of an isolated QD defined by

$$\hat{\boldsymbol{\alpha}} = A N^2 (\hat{\mathbf{I}} - 4\pi A \hat{\mathbf{Y}})^{-1}. \quad (23)$$

Thus, we have shown that the special law of the nonlocality (17) 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 another 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 allows one to conclude that the above introduced EBC's remain valid in the weak confinement regime as well. Note that the above result admits extension of the Maxwell Garnett approach⁶ to 3D composites constituted by QD's in weak confinement regime.

IV. RADIATIVE DECAY RATE IN PLANAR ARRAY OF QD's

Let us apply the EBC method for the investigation of exciton radiative time in 2D array of QD's that are assumed to be spherical inclusions of the radius R . Corresponding problem for QW's was considered in detail in a number of papers (see, e.g., Refs. 12,13,25–28). It can easily be shown that EBC's (14) and (15) describe a QW with the tensorial dielectric function

$$\hat{\epsilon}(\omega) = \epsilon_{\parallel}(\omega)(\mathbf{e}_x\mathbf{e}_x + \mathbf{e}_y\mathbf{e}_y) + \epsilon_{\perp}(\omega)\mathbf{e}_z\mathbf{e}_z, \quad (24)$$

where $\epsilon_{\parallel}(\omega) = \epsilon_h - 4\pi i\sigma_{xx}/ckL_{QW}$, $\epsilon_{\perp}(\omega) = \epsilon_h/(1 + 2\xi/L_{QW})$ and L_{QW} is the QW thickness. Plane-wave reflection coefficients for this QW are given by¹²

$$r_s = \frac{ik\eta_{\parallel}}{2\sqrt{\epsilon_h}\cos\theta - ik\eta_{\parallel}}, \quad (25)$$

$$r_p = \frac{ik[\eta_{\parallel}\cos^2\theta + \epsilon_h^2\eta_{\perp}\sin^2\theta]}{2\sqrt{\epsilon_h}\cos\theta - ik[\eta_{\parallel}\cos^2\theta - \epsilon_h^2\eta_{\perp}\sin^2\theta]} \quad (26)$$

for s polarizations and p polarizations, correspondingly. Here θ is the angle of incidence, $\eta_{\parallel} = L_{QW}[\epsilon_{\parallel}(\omega) - \epsilon_h]$ and $\eta_{\perp} = L_{QW}[\epsilon_{\perp}^{-1}(\omega) - \epsilon_h^{-1}]$. The quantities σ_{xx} and ξ are given by Eqs. (16). For spherical particles, the polarizability tensor takes the form as follows:

$$\hat{\alpha} = R^3 \frac{\epsilon(\omega) - \epsilon_h}{\epsilon(\omega) + 2\epsilon_h} \hat{\mathbf{I}}. \quad (27)$$

Thus, in view of Eq. (1), the reflection coefficients for planar array of spherical QD's are given by Eqs. (25) and (26) after substitutions $L_{QW} \rightarrow 2R$. By analogy with Ref. 12 we conclude that three types of polaritons, T polaritons, L polaritons and Z polaritons, can propagate in the planar array of QD's considered. Frequency poles of function $r_s(\omega)$ correspond to polaritons of T type while poles of $r_p(\omega)$ correspond to polaritons of T type and Z type. Real parts of these poles determine resonant frequencies of corresponding modes. It can easily be found that

$$\omega_T = \omega_L = \omega_0 - \frac{g_0}{3\epsilon_h} \left(1 + \frac{R^3\delta_x}{d^2} \right), \quad (28)$$

for T mode and L mode and corresponding expression for the Z mode is obtained from the above equation by the substitution $\delta_x \rightarrow \delta_z$. Second term in brackets in the right-hand part of this equation is a local-field effect due to electromagnetic

interaction of QD's in array. Namely, the electromagnetic interaction is responsible for the frequency gap between L (T) modes and Z mode.

Imaginary parts of frequency poles of the reflection coefficients (25) and (26) determine the radiative decay rate. Then, using results of Ref. 12 and above-defined substitutions, we obtain

$$\Gamma_T = \frac{\Gamma_0}{\cos\theta}, \quad \Gamma_L = \Gamma_0 \cos\theta, \quad \Gamma_Z = \Gamma_0 \frac{\omega_Z \sin^2\theta}{\omega_L \cos\theta}, \quad (29)$$

for T polaritons, L polaritons, and Z polaritons, correspondingly, where

$$\Gamma_0 = -\frac{2\pi\tilde{\omega}_0 R^3}{3d^2 c \sqrt{\epsilon_h}} g_0 \left(1 - \frac{\delta_x R^3}{3\epsilon_h \tilde{\omega}_0 d^2} g_0 \right), \quad (30)$$

is the radiative decay rate across the plane of QD structure; $\tilde{\omega}_0 = \omega_0 - g_0/3\epsilon_h$. Under derivation of Eqs. (29) we have assumed all three modes to be independent. Indeed, in a wide range of not too big θ the inequality $(g_0 R^3/3\epsilon_h d^2) |\delta_x - \delta_z| \gg \Gamma_{L,Z}$ holds true and, consequently, the LZ splitting (frequency splitting between l polaritons and z polaritons) exceeds linewidths of corresponding modes allowing thus the above assumption.

To compare the radiative decay rates of 2D array of QD's and of a single spherical QD $\gamma = -2(kR)^3 g_0 \sqrt{\epsilon_h}/9$ (see Ref. 31) we rewrite Eq. (30) as

$$\Gamma_0/\gamma \approx B, \quad (31)$$

where $B = 3\pi/(k_1 d)^2$ is the superradiance factor. The latter enhances substantially the radiative lifetime in dense arrays. Note that the quantity B coincides with that derived in Ref. 32 [Eq. (19)] using an approximate analysis of the light diffraction by a 2D array of QD's. Analogous super-radiance factor with the Bohr radius a_B instead of d was introduced also for quantum well.³³ One can interpret the coefficient B in Eq. (31) in close analogy with Ref. 33: it results from the coherent excitation of QD's located at distance d from each other.

Let us analyze dependence of the radiative rates (30) on the QD radius. Since $g_0 \sim 1/R^3$ in the strong confinement regime,²² the explicit dependence of Γ_0 on the QD radius vanishes; this dependence manifests itself only as a weak radial dependence of the renormalized resonant frequency $\tilde{\omega}_0$. The last effect is a combination of two mechanisms one of which originates from the radial dependence of the exciton transition energy $\hbar\omega_0$, and the second one is provided by the local-field effects responsible for the renormalization of the transition energy $\omega_0 \rightarrow \tilde{\omega}_0$. Analogous situation occurs in QW's, where $\Gamma_0^{QW} = -\omega_0 L_{QW} g_0^{QW}/2c\sqrt{\epsilon_h}$ with $g_0^{QW} \sim 1/L_{QW}$. Thus, the QW thickness L_{QW} appears in Γ_0^{QW} as the thickness dependence of the oscillator strength proportional to the transition frequency. This dependence has been study in Ref. 34 over a wide range of thickness including the transition region from strong to weak confinement regime.

In order to study carefully the dependence of radiative lifetime $\tau_0 = 1/\Gamma_0$ in 2D layer of QD's on size of QD's and lateral lattice spacing between QD's, which is of the most

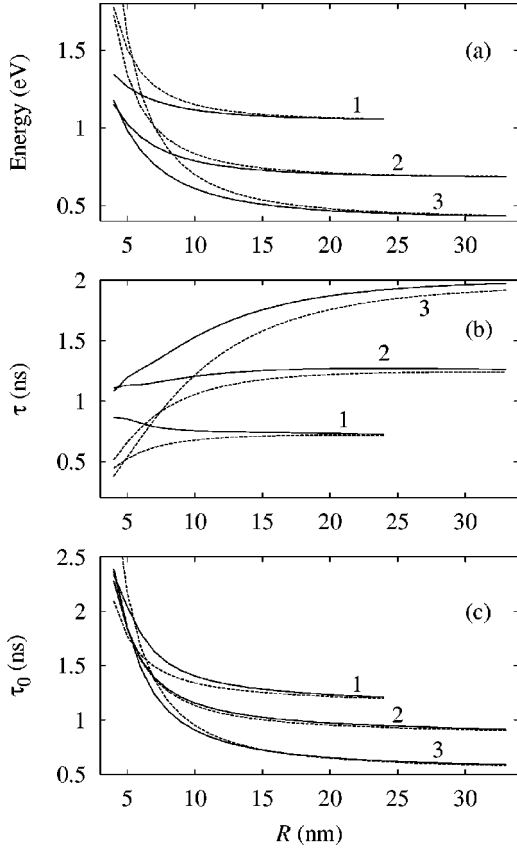


FIG. 2. (a) The ground-state energy of the exciton, (b) the radiative lifetime of the single QD and (c) the radiative lifetime of the planar array of QDs versus the QD radius for spherical $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ QD's with the potential confinement by finite (solid line) and infinitely high (dashed line) barriers and In content $x=0.33$ (1), 0.67 (2), 1 (3); the lattice period $d=200$ nm.

interest especially in the region of small QD sizes less than Bohr radius, we have to find precisely g_0 as a function of QD size. For that, we have performed calculations of the dipole moment $|\mu|$ or oscillator strength of exciton ground state³⁵ by solving Schrödinger equation for the Coulomb-correlated electron-hole pair in $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ QD's. The spherical shape of QD's was supposed and we have restricted ourselves by s -like ground state of the exciton. Two-particle Schrödinger equation was solved by the discrete variable representation³⁶ that was shown to be very effective also for the spherical coordinates³⁷ and can be easily generalized for two-particle problem. Coulomb interaction potential of the electron and hole as charged spherical surfaces was used taking into account different dielectric constants of the materials formed QD and, therefore, surface charge at the $\text{In}_x\text{Ga}_{1-x}\text{As}/\text{GaAs}$ boundary. For confinement potential we have used step-wise function with finite-potential barriers between potentials of QD material $\text{In}_x\text{Ga}_{1-x}\text{As}$ and matrix material GaAs, which are different for conduction and valence bands and dependent on content of In. To reveal in more obvious way the role of the confinement we considered also the infinite high potential barriers in QD's. All material parameters of $\text{In}_x\text{Ga}_{1-x}\text{As}$ as a function of x were taken from Ref. 38.

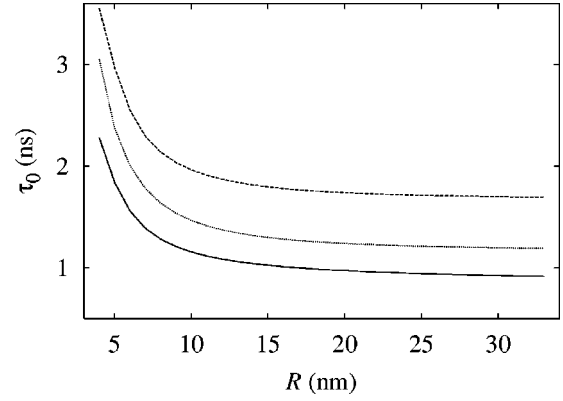


FIG. 3. The radiative lifetime of the planar array of QD's for the ground-state exciton state in $\text{In}_{0.67}\text{Ga}_{0.33}\text{As}/\text{GaAs}$ QD's (solid line) and exciton state without Coulomb correlation and the potential confinement by finite (dashed line) and infinitely high (dotted line) barriers.

In Fig. 2(a) the dependence of the ground-state energy on the QD's radius for the Coulomb-correlated exciton is shown in the case of finite and infinitely high potential barriers at the spherical QD boundary. For both cases of the potential, the photon energy has the pronounced radial dependence in the range of small radii considered. At the same time, overlap integral of the exciton wave function $\Psi(r_e, r_h)$ over equal electron and hole radial coordinate $r_e=r_h$, which defines the oscillator strength,³⁵ grows with radius in a lesser degree. This is a reason for a drastic alteration of the radial dependence of the radiative lifetimes for single QD $\tau=1/\gamma$ and for QD array $\tau_0=1/\Gamma_0$ shown in Fig. 2(b) and 2(c), respectively, which follows from Eqs. (30) and (31). For growing QD radii the ground-state energy as well as calculated radiative lifetimes of the single QD and QD array tend to the radius-independent limits dealt with the relative movement of the electron and hole in the Coulomb potential. However, for increasing radii the movement of the center of mass comes into play;^{39,40} this makes the wave function of the exciton state of nonspherical symmetry that is not considered here. Therefore, in Fig. 2 we restricted the radius rise by the Bohr radius (for $\text{In}_{0.33}\text{Ga}_{0.67}\text{As}$, case 1, $a_B=24.1$ nm).

To show the important role of the Coulomb interaction for the small-sized QD's, in Fig. 3 the radiative lifetime τ_0 of the array of $\text{In}_{0.67}\text{Ga}_{0.33}\text{As}/\text{GaAs}$ QD's is depicted together with the radiative lifetime calculated with the help of the wave functions of the Coulomb-uncorrelated electron and hole confined in the finite and infinite-potential barriers. As one can see, the Coulomb interaction enhances essentially the value of τ_0 even in the range of small radii shown, which is typically considered as a strong confinement regime with negligible contribution of the Coulomb interaction.⁴¹

Finally, it should be noted that besides of the polaritons considered here, in 2D layer of QDs there exist surface polaritons with spatial dependence of the fields as $\exp(-k_z|z|+ik_{\parallel}x)$, where $k_{\parallel}=\sqrt{k_1^2+k_z^2}$. As follows from the EBC's (14) and (15), the quantity k_z is given by

$$k_z = -i \frac{2\pi}{c} k \sigma_{xx}. \quad (32)$$

For the quantum wells these polaritons were considered in Ref. 12 with dispersive equations followed from Eqs. (25) and (26) under substitution $k\sqrt{\epsilon_h}\cos\theta \rightarrow -ik_z$. For QW's they are nonradiative with $\Gamma_{L,T,Z}=0$. It is not a case for 2D layer of QD's if one takes into account radiative effect of a single QD. For that, according to Refs. 21 and 42, we substitute $\hat{\alpha} \rightarrow \hat{\alpha}(\hat{I} - 2ik_1^3\hat{\alpha}/3)^{-1}$ in Eq. (27), equate the denominator in Eq. (25) with zero and find

$$\Gamma_T \approx \frac{6\pi k_1}{k_z} \gamma. \quad (33)$$

The similar but more cumbersome result can be obtained for the case of L polaritons and Z polariton. The physical interpretation of such kind of radiation is followed from exciton interaction with the boundaries of a single QD.

Note that dispersion equation for surface polaritons (32) can also be obtained by a standard but more complicated procedure¹⁹ on the basis of the conventional 3D effective-medium approach. Indeed, planar array of electrically small spherical QD's can be treated as a homogeneous anisotropic layer with the dielectric function given by Eq. (24). Solution of the boundary-value problem for the layer leads us to the dispersion equation for the symmetrical TE mode;⁴³

$$k_z = \sqrt{\frac{2\pi k}{icR} \sigma_{xx} - k_z^2} \tan\left(R \sqrt{\frac{2\pi k}{icR} \sigma_{xx} - k_z^2}\right).$$

In the thin layer limit, when $k_z R \ll 1$, this equation is reduced to Eq. (32). The above example demonstrates us the effectiveness of the EBC method as compared to the conventional 3D effective-medium approach, but also illustrates the above statement that the EBC method is applicable only to electrically thin layers.

V. CONCLUSION

EBC's given by Eqs. (14) and (15) state mathematical equivalence of optical properties of a 2D periodical layer of QD's and an isolated quantum well. It should be stressed that the mechanisms of transport processes and oscillator strengths in each case are essentially different. Nevertheless, the equivalence makes it possible to extend to QD-based planar structures with more complicated configurations (finite-sized QD layer, QD layer in microcavity, several QD layers, etc.) the well-developed mathematical formalism of investigation of quantum wells. Namely, this equivalence provides promising potentiality of the derived EBC's for particular electro-dynamical problems in QD-based structures. In particular, threshold current for QD-based lasers can be evaluated by analogy with solution of corresponding problem for the QW lasers;²⁸ the EBC method allows us to analyze electromagnetic response of a QD layer (or a multilayer structure) placed in microcavity: this is very important for the design of QD-based semiconductor lasers.⁵

It should be emphasized that the extension of the EBC method to deformed or other type of complicated structures (finite-sized QD layer, QD layer in microcavity, several QD layers, etc.) is only possible when the modification of geometrical parameters of the structure does not change the electron-transport properties in it; otherwise, modification of EBC is required. For example, too close location of two planar layers with QD's will change the energy spectrum because of overlapping of exciton wave functions and tunneling. Thus, justification of applicability of EBC's must be given in each particular case.

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