Quantum optics of a quantum dot: Local-field effects

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The role of local fields in quantum electrodynamics of isolated quantum dot (QD) has been analyzed. The system is modeled as a strongly confined in space two-level quantum oscillator illuminated by quantum light. Relation between local and acting fields in QD has been derived in the dipole approximation from the integral Maxwell equations for electromagnetic field operators. A formalism of the electromagnetic field quantization in electrically small scatterers has been developed. As a result, Hamiltonian of the system has been formulated in terms of the acting field with a separate term responsible for the effect of depolarization. Schrödinger equation with that Hamiltonian has been solved in linear approximation. Interaction of QD with different quantum states of light, such as Fock states, coherent states, Fock qubits, entangled states, has been analyzed. It has been shown that the local-fields induce a fine structure of the QD absorption (emission) spectrum: instead of a single line with the frequency corresponding to the exciton transition, a doublet appears with one component shifted to the blue (red). The value of the shift depends only on the geometrical and electronic properties of QD while the intensities of components are completely determined by the quantum light statistics. It has been demonstrated that in the limiting cases of classical light and single-photon state the doublet is reduced to a singlet shifted in the former case and unshifted in the latter one. A physical interpretation of the predicted effect has been proposed. Possible ways of experimental observation of the effect has been discussed together with the potentiality of its utilization in the quantum information processing.

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I. INTRODUCTION

Optical and electronic properties of quantum dots (QDs) is currently an area of active investigation owing to promising potential applications such as active media of double heterostructure laser [1]. Ultrahigh material and differential gain, orders of magnitude exceeding those in quantum well lasers, has been experimentally confirmed. Recently QDs have been proposed to serve as nodes of quantum networks that store and process quantum information being transmitted between nodes by entangled states of photons [2–5]. An experimental observation of a single-QD absorber has been reported in Ref. [6]. Application of semiconductor QDs in cavity quantum electrodynamics [7–9] and as potential quantumlight emitters [10–12] is now intensively discussed. Spontaneous emission in QDs [13] and electromagnetic fluctuations are also in the focus of interest. In connection with that problems, quantum electrodynamics (QED) of quantum dots and ensembles of QDs acquires a special significance. In this paper we analyze the role of local fields in interaction of QDs with nonclassical light.

QD is a structural inhomogeneity in a host semiconductor which confines charge carrier motion producing thus discrete energy spectrum, and scatters incident electromagnetic field inducing thus local fields. If scatterer is electrically small, that implies its linear extension to be small as compared with the wavelength inside the scatterer medium, the local-field effects can be accounted for as *depolarization* of the scatterer. Further QDs are assumed to be electrically small. Depolarization gives an especially significant impact on the electromagnetic response in the case when scatterer is a resonant system, (e.g., QD). There is no reason to assume the depolarization negligible in the interaction of nanoparticles with quantum light. In classical electrodynamics, the local-field effects in isolated QDs were considered in a number of papers, e.g., Refs. [14–16], on the basis of different macroscopic phenomenological models. For the strong confinement regime, where QD linear extension is much less then the Bohr radius of the bulk exciton, a phenomenological theory of linear electromagnetic response of regular 3D-ensembles of QDs has been elaborated in Refs. [17,18]. In particular, polarization-dependent splitting of the gain band in anisotropically shaped QDs has been predicted. Local-field effects in 2D arrays of QDs in both strong and weak confinement regimes were discussed in Ref. [19]. Microscopic models of the local-field effects in spherical QDs have been presented in Refs. [20,21]. In the framework of these models, spontaneous emission problem has been considered semiclassically on the basis of the self-field approach [21].

At the same time we have to state that a consistent consideration of local-field effects in QDs is still lacking; particular models are investigated instead. As a result, there exist qualitatively different predictions of the QD electromagnetic response. For instance, Refs. [14–18,20] predict depolarization shift of the resonant line while such a shift is absent in Ref. [21]; the sign of the shift turns out to be different for the absorption [14,15,20] and stimulated emission [15–18]. A lack of a consistent theory of local fields in QDs does not allow us to judge whether such dif-

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ferences reflect real properties of different optical processes or they are provided by particular approximations.

In general, QED 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. A general QED formalism for dissipative inhomogeneous media is now actively elaborated and is still far from completion. Different procedures, which are not always obviously identical and leads to identical results [22], of the electromagnetic field quantization in such media have been proposed (see, e.g., Refs. [23–35] and references therein). Peculiarities of the electromagnetic field quantization in dielectric media with inverse population were discussed in Ref. [36].

As in classical electrodynamics, both microscopic and macroscopic models are investigated in the QED of inhomogeneous media. The macroscopic approach [27–30] implies phenomenological description of the medium by means of a complex-valued Kramers-Kronig dielectric function. In order to fulfill the commutation relations for the electromagnetic field operators, auxiliary fields describing the medium must be introduced. One of the possible version of auxiliary field is the noise current used in Refs. [27,29–31]. Most general formalism for the noise current concept has been developed in Ref. [30], where QED has been formulated for arbitrary scattering system described by a casual and eventually nonlocal susceptibility tensor. Individualizing of the formalism for electrically small scatterers (point scatterers in the terminology of Savasta *et al.* [30]) has been carried out. However, resonant scatterers like QDs have remained beyond the consideration.

Microscopic approach [31–35] does not use *a priori* defined dielectric function. Instead, electrodynamics is supplemented with the charge carriers transport in the medium. In that case canonical quantization procedure is carried out for the system "electromagnetic field + medium"; there is no need to introduce noise current operators (auxiliary fields). Note that such an approach is more physically justified comparing with the macroscopic phenomenological description but loses generality: model of the charge-carrier transport must be specified before the electromagnetic field quantization.

The present paper introduces local-field effects into quantum optics of QDs. Consideration is based on the microscopic approach analogous to that utilized in Refs. [31,33,35] for plane semiconductor heterostructures. The paper is arranged as follows. In Sec. II, we combine a local-field theory for optically dense media [37,38], based on the relation between acting and local fields in the Liouville equations, with the secondary quantization technique usually being applied to the electron-hole pairs in QDs. Such an approach allows us to account for the local-field effects in systems with fluctuating number of particles. As a result, we find a Hamiltonian of the light interaction with electron-hole pairs in QD and we separate in this Hamiltonian a special term responsible for the local field effects. The Schrödinger representation is used. We reproduce the known results concerning the depolarization shift of the resonant frequency in QDs [14–16] and polarization-dependent splitting of the gain band [17,18], obtained earlier phenomenologically. In Sec. III we extend the Hamiltonian derived to nonclassical light and investigate interaction of QD with factorized states of quantum light. Sec. IV is devoted to local-field effects in QDs interacting with entangled states of light. Discussion of main results and their consequences is presented in Sec. V. Concluding remarks are given in Sec. VI.

II. LOCAL FIELDS AND SECONDARY QUANTIZATION OF ELECTRON-HOLE PAIRS

A. Model Hamiltonian for QD in classical electromagnetic field

Let an isolated QD be exposed to classical electromagnetic field. Further the QD is modeled as a strongly confined in space [14] two-level quantum oscillator. Obviously, QD is an essentially multilevel system. However, contribution all transitions lying far away from a given resonance can be approximated by a nonresonant dielectric function ε_h. We shall assume ε_h to be equal to the dielectric function of the 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 $\varepsilon=\varepsilon_h$. 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

$$c\rightarrow c/\sqrt{\epsilon_h} \quad \text{and} \quad \boldsymbol{\mu}\rightarrow\boldsymbol{\mu}/\sqrt{\epsilon_h} \tag{1}$$

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 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 developed in Refs. [33,35]. 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 for the system under analysis. However, in the practical use, the described approach turned out to be too complicated and was realized for the only simplest configuration: interaction of a material layer with the normally incident light [33,35] (see also Ref. [39]). 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 (see Appendix A). 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 $\mathbf{u}_\alpha$ $(\alpha=x,y,z)$ with the unite 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:

$$H_{IL}=-V\hat{P}_x E_{Lx}, \tag{2}$$

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 [40,41], 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 (see Refs. [42,31]). Here and below we mark operators by the label " ˆ " 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 the QD, $\mathbf{E}_L$, involved in Eq. (2), is different from the external acting field $\mathbf{E}_0$. 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 [43] (see also Fig. 1 for clarity):

$$\mathbf{E}_L=\mathbf{E}_0-4\pi\underline{\mathbf{N}}\,\mathbf{P}. \tag{3}$$

Here $\mathbf{P}=\langle\hat{\mathbf{P}}\rangle$ is the macroscopic polarization, $\underline{\mathbf{N}}$ is the depolarization tensor. This tensor is symmetrical [44] and depends only on the shape of the scattering object. Equation (3) is obtained from the integral equations of electrodynamics in the dipole approximation [44] (see Appendix A). Using Eq. (3) one can easily obtain

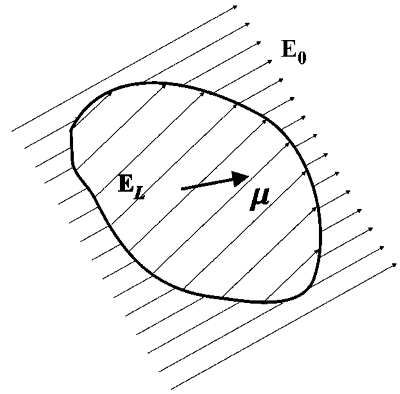

FIG. 1. Schematic picture of correlation between acting and local fields in electrically small scatterer. Local field is assumed to be homogeneous.

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

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

$$H=H_0+H_{I0}+\Delta H, \tag{5}$$

where

$$H_{I0}=-V\hat{P}_x E_{0x} \tag{6}$$

and

$$\Delta H=4\pi N_x P_x(-\mu b^\dagger+\mu^* b)=\frac{4\pi}{V}N_x(\mu^* b-\mu b^\dagger)(\mu^*\langle b\rangle-\mu\langle b^\dagger\rangle). \tag{7}$$

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 [45].

B. Equations of motion

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

$$i\hbar\frac{\partial|\psi\rangle}{\partial t}=H_{int}|\psi\rangle \tag{8}$$

with $|\psi(t)\rangle = \exp(iH_0t/\hbar)|\tilde{\psi}(t)\rangle$ and $H_{int} = \exp(iH_0t/\hbar)(H_{I0} + \Delta H)\exp(-iH_0t/\hbar)$. 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 unknown coefficients to be found, $|g\rangle$ and $|e\rangle$ are the wave functions 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. (8) we obtain the set of equations of motion

$$\begin{aligned} i\hbar\frac{\partial A}{\partial t} &= (4\pi N_x P_x - E_{0x})\mu B e^{i\omega_0 t}, \\ i\hbar\frac{\partial B}{\partial t} &= (4\pi N_x P_x - E_{0x})\mu^* A e^{-i\omega_0 t}, \end{aligned} \tag{9}$$

with macroscopic polarization determined by

$$P_x = \langle\tilde{\psi}|\hat{P}_x|\tilde{\psi}\rangle = \frac{1}{V}\mu^* A(t)B^*(t)e^{-i\omega_0 t} + \text{c.c.} \tag{10}$$

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 (10) into account and neglecting the fast-oscillating terms in Eq. (9), we derive final expressions for equations of motion [46]

$$\begin{aligned} i\hbar\frac{\partial A}{\partial t} &= \hbar\Delta\omega A|B|^2 - \frac{1}{2}\mathcal{E}(t)\mu B e^{i(\omega_0-\omega)t}, \\ i\hbar\frac{\partial B}{\partial t} &= \hbar\Delta\omega B|A|^2 - \frac{1}{2}\mathcal{E}^*(t)\mu^* A e^{-i(\omega_0-\omega)t}, \end{aligned} \tag{11}$$

where

$$\Delta\omega = \frac{4\pi}{\hbar V}N_x|\mu|^2. \tag{12}$$

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 (11) is analogous to the Bloch equations for optically dense media derived in Ref. [37]. The relaxation can easily be included into Eqs. (11) either by introduction of the phenomenological transverse and longitudinal relaxation times [37] or by corresponding modification of initial Hamiltonian (5).

C. Polarization of QD in classical electromagnetic field

The case of excited QD can be analyzed using Eqs. (11) 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. (11) are simplified and reduced to

$$i\hbar\frac{\partial B}{\partial t} = \hbar\Delta\omega B - \frac{1}{2}\mathcal{E}^*(t)\mu^* e^{i(\omega_0-\omega)t}. \tag{13}$$

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

$$B(t) \approx -\frac{\mathcal{E}^*\mu^*}{2\hbar(\omega_0 - \Delta\omega - \omega)}[e^{-i(\omega_0-\omega)t} - e^{-i\Delta\omega t}] \tag{14}$$

with $\Delta\omega$ determined by Eq. (12). Thus, one can see that the local-field (depolarization) leads to the shift $\Delta\omega$ of the resonant frequency. This shift was predicted in a number of papers [14–16] on the basis of different phenomenological models. In Refs. [17,18] 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.* [47] as a hypothesis explaining the experimentally observed polarization-dependent splitting of the PL spectrum of single anisotropically shaped QD.

Equation (12) is identical to that obtained in Refs. [17,18]. In order to demonstrate it we should make a substitution $|\mu|^2 \rightarrow |\mu_0|^2/3$ 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

$$P_x = \frac{1}{8\pi}\alpha_{xx}(\omega)\mathcal{E}[e^{-i\omega t} - e^{-i(-\Delta\omega+\omega_0)t}] + \text{c.c.}, \tag{15}$$

where

$$\alpha_{xx}(\omega) = \frac{4\pi|\mu|^2}{\hbar V(\omega + \Delta\omega - \omega_0)} \tag{16}$$

is the component of the QD polarizability tensor. Phenomenological consideration for QD modeled as single-resonance medium with the Lorentz dispersion $\varepsilon(\omega) = \varepsilon_h + g_0/(\omega - \omega_0)$ [17,18] gives the same result if we put $g_0 = 4\pi|\mu|^2/\hbar V$. This means that the Hamiltonian defined by Eqs. (5)–(7) 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

$$A(t) \approx \frac{\mathcal{E}\mu}{2\hbar(\omega_0 + \Delta\omega - \omega)}[e^{i(\omega_0-\omega)t} - e^{-i\Delta\omega t}]. \tag{17}$$

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.

III. INTERACTION OF QD WITH FACTORIZED STATES OF LIGHT

A. Model Hamiltonian for the case of nonclassical light

The light states which can be expressed by a superposition of fields with different linear polarizations shall further be referred to as factorized states. Linearly polarized quantum light is a particular realization of such states. In order to obtain Hamiltonian of a QD interacting with quantum electromagnetic field, one needs to supplement Eq. (5) by the term H_F corresponding to the free space field, and to change over in the term H_{I0} the electromagnetic field strength by the corresponding operator, $E_{0x}\to\hat{E}_{0x}$. In quantum optics of inhomogeneous media there is a problem of presentation of the electromagnetic field operator since the local fields are inhomogeneous. Unlike conventional approaches, the proposed scheme of the electromagnetic field quantization does not meet this problem since the interaction Hamiltonian is presented in terms of acting field but not the local one. As a result, usual plane-wave expansion is applicable to the operator $\hat{E}_{0x}$; the role of the QD boundary is taken into account by the term ΔH (7). Thus, the Hamiltonian for the case of quantum electromagnetic field is as follows:

$$H=H_0+\Delta H+H_{I0}+H_F, \tag{18}$$

where $H_{I0}=-V\hat{P}_x\hat{E}_{0x}$ and

$$\hat{E}_{0x}=i\sum_k \sqrt{\frac{2\pi\hbar\omega_k}{\Omega}}(c_k e^{i\mathbf{kr}}-c_k^\dagger e^{-i\mathbf{kr}}). \tag{19}$$

In this equation $\omega_k=c|\mathbf{k}|$ is the frequency of the photon mode $\mathbf{k}$, Ω is the normalization volume, $c_k^\dagger$ and c_k are the photon creation and annihilation operators, correspondingly. Taking into account Eq. (19) we obtain

$$H_F=\hbar\sum_k \omega_k\left(c_k^\dagger c_k+\frac{1}{2}\right), \tag{20}$$

$$H_{I0}=-\hbar\sum_k (g_k b^\dagger c_k-g_k^* b c_k^\dagger), \tag{21}$$

where $g_k=-i\mu\sqrt{2\pi\omega_k/\hbar\Omega}\exp(i\mathbf{kr}_c)$, and $\mathbf{r}_c$ is the radius vector of the QD geometrical center.

Hamiltonian (18) conforms to the use of relation (A15) for field operators instead relation (3) for classical fields. The term $\hat{\mathbf{E}}_0$ in Eq. (A15) presented by a superposition of photons [48] is an auxiliary field which can be interpreted as an incident field only in the classical limit. For the quantum light such a simple interpretation is inapplicable: operator $\hat{\mathbf{E}}_0$, in general, is not identical to the field inside or outside the QD; moreover, this term can arise even in the absence of any external sources (for example, in spontaneous transitions). Note that the field $\hat{\mathbf{E}}_0$ is transverse. As has been pointed out in Ref. [48], it is such a field that can be represented by a superposition of "genuine" photons. The total field inside QD is not transverse due to the second term in Eq. (A7) (see also Refs. [49,50]). Direct application of conventional quantization schemes to such fields leads to a crucial problem which is impossibility to fulfill commutative relations for electromagnetic field operators without introduction of auxiliary fields [27–30]. In our approach we reduce the problem of quantization of local field to that for acting field. Since the acting field is a superposition of plane waves, quantization procedure gets routined.

B. Equations of motion

In the interaction representation the system "QD + quantum electromagnetic field" with Hamiltonian (18) is described by Eq. (8) where the substitution $H_0\to H_0+H_F$ should be performed. In that case, wave function of the system can be presented by

$$|\psi(t)\rangle=\sum_{k,n_k=0}[A_k^{n_k}(t)|e\rangle+B_k^{n_k}(t)|g\rangle]|n_k\rangle, \tag{22}$$

where $A_k^{n_k}(t)$, and $B_k^{n_k}(t)$ are unknown functions of time to be found, $|n_k\rangle$ denotes the field states where there is n photons in mode $\mathbf{k}$ and no photons in all other modes, $|0\rangle$ is the wave function of the electromagnetic field in the vacuum state. In view of relation (22), formulas (10) for macroscopic polarization is transformed to

$$P_x=\frac{1}{V}\mu^*\theta(t)e^{-i\omega_0 t}+\text{c.c.}, \tag{23}$$

where

$$\theta(t)=\sum_{k,n_k=0}A_k^{n_k}(t)[B_k^{n_k}(t)]^*. \tag{24}$$

Then, after some standard manipulations with Schrödinger equation (8) we come to the infinite chain of coupled nonlinear differential equations for slowly varying amplitudes:

$$i\frac{dA_k^0}{dt}=\Delta\omega B_k^0\sum_{q,m_q}A_q^{m_q}(B_q^{m_q})^*+\sum_q g_q B_q^{1_q}e^{-i(\omega_q-\omega_0)t}, \tag{25}$$

$$i\frac{dB_k^0}{dt}=\Delta\omega A_k^0\sum_{q,m_q}(A_q^{m_q})^*B_q^{m_q}, \tag{26}$$

$$i\frac{dB_k^{1_k}}{dt}=\Delta\omega A_k^{1_k}\sum_{q,m_q}(A_q^{m_q})^*B_q^{m_q}+g_k^*A_k^0 e^{i(\omega_k-\omega_0)t}, \tag{27}$$

$$i\frac{dA_k^{n_k}}{dt}=\Delta\omega B_k^{n_k}\sum_{q,m_q}A_q^{m_q}(B_q^{m_q})^*+g_k\sqrt{n_k+1}B_k^{n_k+1}e^{-i(\omega_k-\omega_0)t}, \tag{28}$$

$$i\frac{dB_k^{n_k+1}}{dt}=\Delta\omega A_k^{n_k+1}\sum_{q,m_q}(A_q^{m_q})^*B_q^{m_q}+g_k^*\sqrt{n_k+1}A_k^{n_k}e^{i(\omega_k-\omega_0)t}, \tag{29}$$

for arbitrary n_k. This system will serve us as a basis for further analysis at different initial conditions. Note that namely accounting for the depolarization field is a specific property of this system which makes it nonlinear and couples all quantum states of electromagnetic field, distinguishing, thus, this system from conventional equations of quantum electrodynamics. In the limit $N_x\to 0$ the system (25)–(29) splits into recurrent sets of linear equations coupling only $|n_k\rangle$ and $|n_k+1\rangle$ states. In that limit the system becomes equivalent to the ordinary system of the equations of motion of a two-level atom exposed to quantum electromagnetic field [41]. It can easily be shown that system of equations (25)–(29) satisfies the following conservation law:

$$\frac{d}{dt}\sum_{k,n_k=0}(|A_k^{n_k}|^2+|B_k^{n_k}|^2)=0. \tag{30}$$

Thus, letting the wave function to be orthonormal in the initial point of time, we obtain the relation

$$\sum_{k,n_k=0}[|A_k^{n_k}(t)|^2+|B_k^{n_k}(t)|^2]=1 \tag{31}$$

for arbitrary point of time.

C. Interaction with single-photon states

1. Spontaneous emission

The process of spontaneous emission from a QD can be treated as interaction of an excited QD with two states of electromagnetic field, $|0\rangle$ and $|1_k\rangle$. Neglecting in Eqs. (25)–(29) all other states, we reduce the system to the following form:

$$i\frac{dA_k^0}{dt}=\Delta\omega B_k^0\sum_q A_q^{1_q}(B_q^{1_q})^*+\sum_q g_qB_q^{1_q}e^{-i(\omega_q-\omega_0)t},$$

$$i\frac{dB_k^0}{dt}=\Delta\omega A_k^0\sum_q(A_q^{1_q})^*B_q^{1_q}, \tag{32}$$

$$i\frac{dB_k^{1_k}}{dt}=\Delta\omega A_k^{1_k}\sum_q(A_q^{1_q})^*B_q^{1_q}+g_k^*A_k^0e^{i(\omega_k-\omega_0)t},$$

$$i\frac{dA_k^{1_k}}{dt}=\Delta\omega B_k^{1_k}\sum_q A_q^{1_q}(B_q^{1_q})^*,$$

with initial conditions given by

$$A_k^0(0)=1,\quad B_k^0(0)=B_k^{1_k}(0)=A_k^{1_k}(0)=0. \tag{33}$$

These conditions correspond to the excited state of electron-hole pair with zero number of photons in the initial time. In view of initial conditions (33), from system (32) follows that the terms $B_k^0(t)$ and $A_k^{1_k}(t)$ are of the higher-order infinitesimal and can be neglected. Then system (32) is reduced to

$$\frac{dA_k^0}{dt}=-i\sum_q g_qB_q^{1_q}e^{-i(\omega_q-\omega_0)t},$$

$$\frac{dB_k^{1_k}}{dt}=-ig_k^*A_k^0e^{i(\omega_k-\omega_0)t}. \tag{34}$$

In investigation of this system we should take into account natural width of the resonant transition. By this reason we cannot assume $A_k^0(t)\approx 1$ as we have done under derivation of Eq. (14). Let us integrate second equation in system (34):

$$B_k^{1_k}(t)=-ig_k^*\int_0^t A_k^0(t')e^{i(\omega_k-\omega_0)t'}dt'. \tag{35}$$

Substitution of this expression into first equation of the system leads us to the integro-differential equation

$$\frac{dA_k^0}{dt}=\int_0^t K(t-t')A_k^0(t')dt' \tag{36}$$

with operator of the Volterra type and kernel

$$K(t)=-\sum_k|g_k|^2e^{-i(\omega_k-\omega_0)t}. \tag{37}$$

By means of the substitution

$$\sum_k[\cdot]\to\frac{\Omega}{(2\pi)^3}\int_0^{2\pi}d\varphi\int_0^{\pi}\sin\theta d\theta\int_0^{\infty}k^2[\cdot]dk, \tag{38}$$

which correspond to the limit transition $\Omega\to\infty$, and subsequent integration [51], we reduce Eq. (37) to the simple notation $K(t)=-\Gamma_{sp}\delta(t)/2$, where

$$\Gamma_{sp}=\frac{4|\mu|^2\omega_0^3}{\hbar c^3}=\frac{4|\boldsymbol{\mu}_0|^2\omega_0^3}{3\hbar c^3} \tag{39}$$

is the radiative linewidth. Note that the account for nonresonant transitions by means of substitutions (1) gives the result identical to that obtained in Ref. [13]. Analogous result has been obtained in Ref. [34] under consideration of the spontaneous emission of an excited atom imbedded in a lossy dispersive dielectric medium. Equation (36) leads to the elementary relation

$$A_k^0(t)=\exp(-\Gamma_{sp}t/2). \tag{40}$$

Although relations (35) and (40) have been obtained as a result of approximate integration of Eqs. (32), it can easily be shown that these relations together with the conditions

$B_k^0(t)=A_k^{1_k}(t)=0$ generate an exact solution of system (32) independently on whether or not the role of depolarization fields is small.

Solution (40) defines the spontaneous emission process characterized by the resonant line

$$\frac{1}{\omega-\omega_0+i\Gamma_{sp}/2}.$$

It should be emphasized that the spontaneous emission line, unlike absorption and stimulated emission, does not experience depolarization shift (this line shows only a small Lamb shift neglected in our analysis). The depolarization does not also influence the resonance linewidth. Analogous situation appears in interaction of QD with any pure state of electromagnetic field. To make clear physical sense of the result obtained, let us consider mean value of the electric field for the operator $\hat{E}_{0x}$ (19) with wave function defined by Eq. (22):

$$\langle \hat{E}_{0x}\rangle=\langle\tilde{\psi}|\hat{E}_{0x}|\tilde{\psi}\rangle$$
$$=-2\,\mathrm{Im}\Bigg\{\sum_{q,n_q}\sqrt{\frac{2\pi\hbar\omega_q}{\Omega}}e^{i(\mathbf{q}\mathbf{r}_c-\omega_q t)}$$
$$\times\sqrt{n_q+1}\big[(A_q^{n_q})^*A_q^{n_q+1}+(B_q^{n_q})^*B_q^{n_q+1}\big]\Bigg\}. \tag{41}$$

It follows from this expression that $\langle\hat{E}_{0x}\rangle=0$ for any state of electromagnetic field with a fixed number of photons. Thus, if initial state of electromagnetic field is a pure state (as it take place in the case of spontaneous emission), its mean value is equal zero and it does not induce depolarization field. The situation is drastically changed in the case of field states with fluctuating number of photons. This problem is considered below in Sec. III D.

2. Absorption of a single photon

Absorption of a single photon with the wave number $q=|\mathbf{q}|$ is described by system of Eqs. (32) imposed to the initial conditions

$$B_k^{1_k}(0)=\delta_{kq},\quad A_k^0(0)=B_k^0(0)=A_k^{1_k}(0)=0. \tag{42}$$

In accordance with the procedure presented in the previous section, we can put $B_k^0(t)=A_k^{1_k}(t)=0$ in Eqs. (32) and thus reduce them to set (34). Solution of this set with the above stated initial conditions leads us to

$$A_q^0(t)=\frac{g_q}{\omega-\omega_0+i\Gamma_{sp}/2}\big[e^{i(\omega_0-\omega)t}-e^{-\Gamma_{sp}t/2}\big], \tag{43}$$

$$B_k^{1_k}(t)=\delta_{kq}-ig_k^*\int_0^t A_q^0(t')e^{i(\omega_k-\omega_0)t'}dt'. \tag{44}$$

The absorption cross section for a single photon is determined by the formulas $\sigma_{ph}=w(\infty)\Omega/c$, where $w(t)=\partial|A_q^0|^2/\partial t$ is the probability of the transition in a given point of time. After some standard manipulations with Eqs. (43) and (44), we come to

$$\sigma_{ph}=\frac{|g_q|^2\Omega\Gamma_{sp}/c}{(\omega-\omega_0)^2+\Gamma_{sp}^2/4}. \tag{45}$$

Thus, as different from the emission and absorption of classical electromagnetic waves, defined by Eqs. (14) and (17), spontaneous emission and absorption of a single photon are characterized by the same resonant frequency and the same radiative linewidth. From that we conclude that the single-photon processes are insensitive to the depolarization field. This can easily be understood from that the mean electric field of a single photon, in accordance with Eq. (41), is equal zero.

D. Polarization of QD by coherent state of light

Now, let us consider interaction of an isolated QD with an elementary coherent state of light $|s_q\rangle$ which is determined as eigenfunction of the photon annihilation operator [40] of a given photonic mode $q=|\mathbf{q}|$: $c_q|s_q\rangle=s_q|s_q\rangle$. This coherent state can be expanded into a series in the energy states $|n_q\rangle$:

$$|s_q\rangle=\sum_{n_q=0}^{\infty}F_{s_q}(n_q)|n_q\rangle. \tag{46}$$

Here the coefficients $F_{s_q}(n_q)$ are given by Ref. [40]

$$F_{s_q}(n_q)=\exp(-|s_q|^2/2)\frac{s_q^{n_q}}{\sqrt{n_q!}},$$

and $|s_q|^2=\langle n_q\rangle_s$ stands for the mean value of number of photons. The coefficients $F_{s_q}(n_q)$ satisfy the orthonormalization condition

$$\sum_{n_q=0}^{\infty}F_{s_q}^2(n_q)=1.$$

Mean value of the complex-valued electric field amplitude for the incident coherent state is given, in line with Eq. (41), by the expression

$$\langle\hat{\mathcal{E}}\rangle=-2\frac{\hbar g_q}{\mu}\sum_{n_q=0}^{\infty}\sqrt{n_q+1}\;F_{s_q}(n_q)F_{s_q}(n_q+1). \tag{47}$$

The initial conditions for a ground-state QD exposed to the coherent light state are as follows:

$$B_k^{n_k}(0)=\delta_{kq}F_{s_q}(n_q),\;\;A_k^{n_k}(0)=0. \tag{48}$$

In such a case, only terms with $\mathbf{k}=\mathbf{q}$ can be retained in Eqs. (25)–(29). Further we restrict ourselves to the temporal intervals as small as compared to the radiation lifetime: $t\ll\tau\sim1/\Gamma_{sp}$. Then the approximate relations $B_q^{n_q}(t)\approx F_{s_q}(n_q)$

hold true for arbitrary $n_q \geqslant 0$. Multiplying Eq. (28) by $F_{s_q}(n_q)$ and Eq. (25) by $F_{s_q}(0)$, after summation we obtain the equation

$$i\hbar \frac{\partial \theta_q}{\partial t} = \hbar \Delta\omega \theta_q - \frac{1}{2}\langle \hat{\mathcal{E}}(t)\rangle \mu e^{i(\omega_0-\omega)t}, \tag{49}$$

where

$$\theta_q(t) = \sum_{n_q=0}^{\infty} A_q^{n_q}(t) F_{s_q}(n_q). \tag{50}$$

Equation (49) is identical to Eq. (13) correct to the change $\mathcal{E} \to \langle \hat{\mathcal{E}} \rangle$. Thus, we state the correspondence $\theta_q(t) \to A(t)$. As a result, solution of Eq. (49) gives us the polarization

$$P_x = -\frac{1}{8\pi}\alpha_{xx}(\omega)\langle \hat{\mathcal{E}}\rangle [e^{-i\omega t} - e^{-i(\Delta\omega+\omega_0)t}] + \text{c.c.}, \tag{51}$$

where $\alpha_{xx}(\omega)$ is determined by Eq. (16) with the change $\Delta\omega \to -\Delta\omega$. Thus, by analogy with the prediction of classical electrodynamics, absorption line for the coherent light is shifted by the value $\Delta\omega$.

Now, let us dwell on the problem of an excited QD exposed to the coherent light. In that case, the initial conditions take the form

$$A_k^{n_k}(0) = \delta_{kq} F_{s_q}(n_q), \quad B_k^{n_k}(0) = 0; \tag{52}$$

they can be reduced to the approximate relations $A_q^{n_q}(t) \approx F_{s_q}(n_q)$. Further manipulations lead us to the equation

$$i\hbar \frac{\partial \theta_q^*}{\partial t} = \hbar \Delta\omega \theta_q^* - \frac{1}{2}\langle \hat{\mathcal{E}}(t)\rangle^* \mu^* e^{-i(\omega_0-\omega)t} \tag{53}$$

for the quantity

$$\theta_q^*(t) = \sum_{n_q=0}^{\infty} B_q^{n_q}(t) F_{s_q}(n_q).$$

This equation states the correspondence $\theta_q^*(t) \to B(t)$. Thus, an excited QD exposed to the coherent light shows the shift $\Delta\omega$ of resonant line in the direction opposite to that for a ground-state QD.

To conclude this section, note that the expressions for the macroscopic polarization obtained here in the framework of quantum electrodynamics are identical to the expressions following from the classical electrodynamics correct to the change $\mathcal{E} \to \langle \hat{\mathcal{E}} \rangle$. However, in quantum electrodynamics, unlike classical one, electromagnetic response of a system is irreducible to macroscopic polarization. This statement is illustrated in the following section.

E. Scattering of electromagnetic Fock qubits

A superposition of two arbitrary quantum field states are referred to as qubit. Accordingly, Fock qubit is a superposition of two arbitrary Fock states which are eigenfunctions of the Hamiltonian H_F (20); Fock states are the states with a fixed number of photons. Let a ground-state QD interacts with electromagnetic field in the Fock qubit state of the mode q: $\beta_{N_q}|N_q\rangle + \beta_{N_q+1}|N_q+1\rangle$. Here β_{N_q} and β_{N_q+1} are the complex-valued quantities, for which the normalization relation $|\beta_{N_q}|^2 + |\beta_{N_q+1}|^2 = 1$ is fulfilled. Physical principles of generation of arbitrary quantum states of light and, particularly, electromagnetic qubits, were considered in Refs. [52–57].

In the case being considered, explicit expressions for wave functions can easily be found; this allows analytical treatment of the scattering problem. We start with the case $N_q \geqslant 2$. The other cases, $N_q = 0$ and $N_q = 1$, can be considered by analogy but lead to mathematically differing results. More detail consideration of the case $N_q = 0$ is given at the end of this section. Dynamical properties of the system are described by Eqs. (25)–(29) imposed to the initial conditions:

$$B_k^{n_k}(0) = (\delta_{N_k,n_k}\beta_{N_k} + \delta_{N_k+1,n_k}\beta_{N_k+1})\delta_{kq},$$
$$A_k^{n_k}(0) = A_k^0(0) = B_k^0(0) = 0, \tag{54}$$

with $n_k \geqslant 1$. Since we deal with the given photon mode, index q in N_q is further omitted. As in previous section, we restrict the analysis to temporal intervals small as compared to the radiation lifetime. Then the approximate relations

$$B_q^N(t) \approx \beta_N = \text{const}, \quad B_q^{N+1}(t) \approx \beta_{N+1} = \text{const} \tag{55}$$

hold true. As a result, amplitudes A_q^N and A_q^{N+1} satisfy the coupled differential equations

$$\frac{d}{dt}\begin{pmatrix} A_q^N \\ A_q^{N+1}\end{pmatrix} = -i\Delta\omega \begin{pmatrix} |\beta_N|^2 & \beta_N\beta_{N+1}^* \\ \beta_N^*\beta_{N+1} & |\beta_{N+1}|^2 \end{pmatrix}\begin{pmatrix} A_q^N \\ A_q^{N+1}\end{pmatrix} + \begin{pmatrix} f_q(t) \\ 0 \end{pmatrix}, \tag{56}$$

while amplitude A_q^{N-1} satisfies the single differential equation

$$\frac{dA_q^{N-1}}{dt} = -ig_q\sqrt{N}\beta_N e^{-i(\omega_q-\omega_0)t}. \tag{57}$$

Here $f_q(t) = -ig_q\sqrt{N+1}\beta_{N+1}\exp[-i(\omega_q-\omega_0)t]$.

First, let us analyze system of Eqs. (56). If we let $f_q(t) = 0$, the partial solutions of the type $A_q^{N+1,N} \sim \exp(-i\lambda t)$ satisfy the characteristic equation $\lambda^2 - \lambda\Delta\omega = 0$, which has two roots, $\lambda_1 = 0$ and $\lambda_2 = \Delta\omega$, with $\Delta\omega$ defined by Eq. (12). Thus, the eigenstate spectrum of system (56) contains states with resonant frequency both unshifted and shifted due to the depolarization; these eigenstates degenerate at $N_x \to 0$. Note that the gap between resonances significantly exceeds the linewidth: $\Delta\omega \gg \Gamma_{sp}/2$, where coefficient Γ_{sp} is given by expression (39).

General solution of Eqs. (56) is represented by a superposition of two eigenstates considered and can be found by the variation of constants

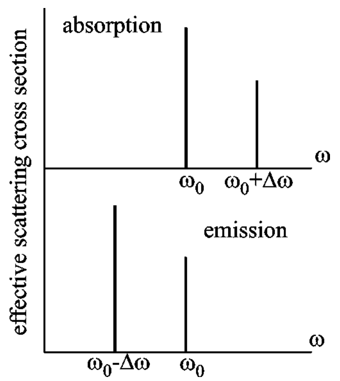

FIG. 2. Fine structure of the electromagnetic response of QD illuminated by quantum light. For the depicted cases of absorption and emission, the weighting coefficients β_N and β_{N+1} have been chosen different.

$$A_q^N(t)=c_{1q}(t)+c_{2q}(t)e^{-i\Delta\omega t},$$
$$A_q^{N+1}(t)=-\frac{\beta_N^*}{\beta_{N+1}^*}c_{1q}(t)+\frac{\beta_{N+1}}{\beta_N}c_{2q}(t)e^{-i\Delta\omega t}, \tag{58}$$

where

$$c_{1q}(t)=-g_q\sqrt{N+1}\beta_{N+1}|\beta_{N+1}|^2\ \frac{e^{i(\omega_0-\omega)t}-1}{\omega_0-\omega},$$
$$c_{2q}(t)=-g_q\sqrt{N+1}\beta_{N+1}|\beta_N|^2\ \frac{e^{i(\omega_0-\omega+\Delta\omega)t}-1}{\omega_0-\omega+\Delta\omega}.$$

Solving Eq. (57) we obtain

$$A_q^{N-1}(t)=-g_q\sqrt{N}\beta_N\frac{[e^{i(\omega_0-\omega)t}-1]}{\omega_0-\omega}. \tag{59}$$

The wave functions derived allows us to express the transition probability in the system by

$$w(t)=\frac{d}{dt}[|A_q^{N-1}(t)|^2+|A_q^N(t)|^2+|A_q^{N+1}(t)|^2]$$
$$=\frac{d}{dt}\left[|A_q^{N-1}(t)|^2+\left|\frac{c_{1q}(t)}{\beta_{N+1}}\right|^2+\left|\frac{c_{2q}(t)}{\beta_N}\right|^2\right]. \tag{60}$$

After some elementary manipulations and substitution $\sin(\alpha t)/\pi\alpha\rightarrow\delta(\alpha)$ in the limit $t\rightarrow\infty$, we find

$$w(\infty)=2\pi|g_q|^2\{[N|\beta_N|^2+(N+1)|\beta_{N+1}|^4]\delta(\omega_0-\omega)+(N+1)|\beta_N|^2|\beta_{N+1}|^2\delta(\omega_0+\Delta\omega-\omega)\}. \tag{61}$$

This quantity defines the QD effective scattering cross section: $\sigma=w(\infty)/I$, where $I=[N+|\beta_{N+1}|^2]c/\Omega$ is the flux density of incident photons. Figure 2 schematically represents the QD optical response defined by Eq. (61) for absorption of Fock qubit. In the case of stimulated emission the substitution $\Delta\omega\rightarrow-\Delta\omega$ should be performed in Eq. (61).

Let us analyze the limiting cases of Eq. (61). Neglecting the local-field effects, i.e., in the limit $\Delta\omega\rightarrow 0$, Eq. (61) reduces to

$$w(\infty)=2\pi|g_q|^2[N+|\beta_{N+1}|^2]\delta(\omega_0-\omega), \tag{62}$$

giving unshifted resonance. In the case when incident-field contains the only photon state, we must substitute in Eq. (61) $\beta_{N+1}\rightarrow 0$ and $\beta_N\rightarrow 1$, or vice versa, $\beta_{N+1}\rightarrow 1$ and $\beta_N\rightarrow 0$. In the former case we obtain

$$w(\infty)=2\pi|g_q|^2N\delta(\omega_0-\omega), \tag{63}$$

while the latter one leads to the identical expression with $N\rightarrow N+1$ substituted. Thus, single-photon states are characterized by unshifted resonances like it take place under neglecting depolarization. However, amplitudes of resonances are quite different.

The above analysis demonstrates that two spectral lines are presented in the effective scattering cross section. One of these lines has the frequency of the exciton transition and another one is shifted owing to the induced depolarization of QD. The shifted line is due to macroscopic polarization of QD. This can easily be shown explicitly by evaluation of macroscopic polarization from Eq. (23) taking into account relations (55) and (58). As a result, we obtain

$$P_x=-\frac{\mu^*}{V}\beta_N^*\beta_{N+1}g_q\sqrt{N+1}\frac{[e^{-i\omega t}-e^{-i(\omega_0+\Delta\omega)t}]}{\omega_0-\omega+\Delta\omega}+\text{c.c.} \tag{64}$$

Since, in accordance with Eq. (41), the complex-valued amplitude of the mean incident field is given by $\langle\hat{\mathcal{E}}\rangle=-2\hbar\beta_N^*\beta_{N+1}g_q\sqrt{N+1}/\mu$, polarization (64) can be presented in the form (51). Thus, we can conclude that the shifted line is related only with the classical polarization. Unlike that, presence of the unshifted line is conditioned by the quantum nature of electromagnetic field. This line does not exist in the framework of classical electrodynamics. Indeed, the classical approach implies that the scattering cross section is completely determined by the QD macroscopic polarization. We can illustrate it using the results of Sec. II C. Since for the classic light $w(t)=d|A|^2/dt$, Eq. (17) yields us $w(\infty)=\pi|\mathcal{E}|^2|\mu|^2\delta(\omega_0+\Delta\omega-\omega)/2\hbar^2$. Quantum nature of the electromagnetic field gives rise to electromagnetic response, which is not related to the medium polarization and is conditioned by the field eigenstates with a fixed number of photons. Spontaneous emission is an example of such kind of response. The key result of our paper is that we have shown that the electromagnetic field states with fixed and fluctuating numbers of photons differently react to the local fields. The states of the first type do not feel the local fields while states of the second type demonstrate a shift of resonant frequency.

Consider now the case $N=0$, which corresponds to interaction with QD of a single-photon related to the vacuum state of the electromagnetic field. This process is described by the system of Eq. (56) at $N=0$ with initial conditions

$$A_q^0(0)=A_q^1(0)=0,\quad B_q^0(0)=\beta_0,\quad B_q^1(0)=\beta_1. \tag{65}$$

The only difference that in the previous case system (56) was supplemented by independent Eq. (57) whose analog is absent under consideration of interaction of QD with virtual photons. This circumstance leads to an essential modification of the solution. Indeed, in the given case the transition probability $w(t)$ is expressed by the formulas

$$w(t)=\frac{d}{dt}[|A_q^0(t)|^2+|A_q^{1q}(t)|^2]=\frac{d}{dt}\left[\left|\frac{c_{1q}(t)}{\beta_1}\right|^2+\left|\frac{c_{2q}(t)}{\beta_0}\right|^2\right], \tag{66}$$

which in the limit $t\to\infty$ is reduced to

$$w(\infty)=2\pi|g_q|^2|\beta_1|^2[|\beta_1|^2\delta(\omega_0-\omega)+|\beta_0|^2\delta(\omega_0+\Delta\omega-\omega)]. \tag{67}$$

This relation, the same as Eq. (61), presents the transition probability by superposition of two lines separated by the frequency gap $\Delta\omega$. However, because of absence in Eq. (66) the third term defined by Eq. (57), the ratio of amplitudes differs from that given by Eq. (61). In the limiting case $\beta_0\to 0$ (single photon), Eq. (67) is reduced to Eq. (63) with $N=1$. In the opposite case $\beta_0\to 1$ (virtual photon), from Eq. (66) follows the result $w(\infty)=0$ which is not the case in Eq. (61). Physically this result reflects disability of the vacuum state to change an equilibrium state of quantum system.

IV. INTERACTION OF QDS WITH ENTANGLED STATES OF LIGHT

Problem of interaction of a QD with entangled states of light require to be carefully considered from the standpoint of the results obtained in previous sections. Since the entangled states are generated by photons with different polarization, the items H_F, H_{I0}, and ΔH in Hamiltonian (18) should be modified to take polarization into account:

$$H_F=\hbar\sum_{\sigma=1,2}\sum_k \omega_k\left(c_{k\sigma}^\dagger c_{k\sigma}+\frac{1}{2}\right), \tag{68}$$

$$H_{I0}=-\hbar\sum_{\sigma=1,2}\sum_k (g_{k\sigma}b^\dagger c_{k\sigma}-g_{k\sigma}^* b c_{k\sigma}^\dagger), \tag{69}$$

$$\Delta H=4\pi\sum_{\sigma,\sigma'=1,2}N_{\sigma\sigma'}P_{\sigma'}(-\mu_\sigma b^\dagger+\mu_\sigma^* b), \tag{70}$$

where the index σ enumerates the photon polarization states, $g_{k\sigma}=-i\mu_\sigma\sqrt{2\pi\omega_k/\hbar\Omega}\exp(i\mathbf{k}\mathbf{r}_c)$, $P_\sigma=V^{-1}(\mu_\sigma^*\langle b\rangle-\mu_\sigma\langle b^\dagger\rangle)$. The term H_0 in Eq. (18) remains unchanged.

Let the ground-state QD be exposed to an arbitrary entangled state $C_{00}|00\rangle+C_{01}|01\rangle+C_{10}|10\rangle+C_{11}|11\rangle$ being characterized by a given wavevector $\mathbf{q}$. The notation $|ij\rangle$ denotes the product $|i\rangle_{\sigma=1}|j\rangle_{\sigma'=2}$ of single-photon wave functions with different polarizations. The arbitrary complex-valued coefficients C_{ij} satisfy the normalization condition $\Sigma_{ij}|C_{ij}|^2=1$. In the case of a fixed wave vector, expressions (68)–(69) as well as an expression for the wave function of the system "QD+electromagnetic field," are drastically simplified; the latter takes the form as follows:

$$\psi=\sum_{i,j=0,1}(A_{ij}|e\rangle+B_{ij}|g\rangle)|ij\rangle, \tag{71}$$

where A_{ij} and B_{ij} are the coefficients to be found. Using the Schrödinger equation in the interaction representation (8), one can derive a system of differential equations for these amplitudes. In matrix notation this system is given by

$$\begin{aligned}\frac{d}{dt}\mathbf{A}&=-i\Delta\omega\mathbf{B}\mathbf{B}^\dagger\mathbf{A}-i\underline{\mathcal{G}}\mathbf{B}\exp\{i(\omega_0-\omega_q)t\},\\ \frac{d}{dt}\mathbf{B}&=-i\Delta\omega\mathbf{A}\mathbf{A}^\dagger\mathbf{B}-i\underline{\mathcal{G}}^\dagger\mathbf{A}\exp\{-i(\omega_0-\omega_q)t\},\end{aligned} \tag{72}$$

where

$$\underline{\mathcal{G}}=\begin{pmatrix}0 & g_{q2} & g_{q1} & 0\\ 0 & 0 & 0 & g_{q1}\\ 0 & 0 & 0 & g_{q2}\\ 0 & 0 & 0 & 0\end{pmatrix}.$$

Initial conditions for this system are as follows:

$$\mathbf{A}(0)=0,\ \ \mathbf{B}(0)=\mathbf{C}. \tag{73}$$

In the above equations $\mathbf{A}$, $\mathbf{B}$, and $\mathbf{C}$ are the column matrices with A_{ij}, B_{ij}, and C_{ij} as components, respectively. In the linear approximation with respect to electromagnetic field, and by analogy with Sec. II we can assume the approximate relations $\mathbf{B}(t)\approx\mathbf{C}$ to be held. Taking this into account we transform system (72) into the set of linear equations:

$$\frac{d}{dt}\mathbf{A}=-i\Delta\omega\mathbf{C}\mathbf{C}^\dagger\mathbf{A}-i\underline{\mathcal{G}}\mathbf{C}\exp\{i(\omega_0-\omega_q)t\}. \tag{74}$$

Initial conditions for this system are stated by the first equality in relations (73).

Evaluation from system (74) the QD response to an arbitrary entangled state is a complicated and intricate problem which is beyond the scope of our consideration. Nevertheless, a significant conclusion follows from set (74) without its explicit integration. Indeed, characteristic equation of the homogeneous system is given by $\mathrm{Det}(\lambda\text{-}\Delta\omega\ \mathbf{CC}^+)\equiv\lambda^3(\lambda-\Delta\omega)=0$. Thus, we conclude that the response contains two spectral components divided by the gap $\Delta\omega$. The ratio of intensities of the components depends on the exciting entangled state, i.e., on the coefficients C_{ij}. Let us demonstrate this for particular case of the state with $C_{10}=C_{01}=0$ which is a superposition of independent qubit and virtual photon: $C_{00}|00\rangle+C_{10}|10\rangle=(C_{00}|0\rangle_1+C_{10}|1\rangle_1)|0\rangle_2$; consequently, this state is not "genuine" entangled state. From the last formula follows that the transition probability $w(\infty)$ for the considered state is given by relation (67) with $\beta_0\to C_{00}$, $\beta_1\to C_{10}$, and $g_q\to g_{q1}$. Immediately, we conclude that in the case under consideration the QD response exhibits two spectral lines with ratio of intensities determined by the ratio C_{00}/C_{10}.

Consider now interaction of a QD with the Bell state of light, $\Phi^{\pm}=(|00\rangle \pm |11\rangle)/\sqrt{2}$ and $\Psi^{\pm}=(|01\rangle \pm |10\rangle)/\sqrt{2}$, which play a fundamental role in electrodynamics of entangled states. The Bell states are characterized by maximal entanglement and form a complete system, i.e., an arbitrary entangled state can be presented as a superposition of the Bell states.

So far $C_{00}=1/\sqrt{2}$, $C_{10}=C_{01}=0$, and $C_{11}=\pm 1/\sqrt{2}$ for the states $\Phi^{\pm}$, system (74) is significantly simplified for these states and is reduced to

$$\frac{d}{dt}A_{01}=\mp i\frac{g_{q1}}{\sqrt{2}}\exp\{i(\omega_0-\omega_q)t\},$$
$$\frac{d}{dt}A_{10}=\mp i\frac{g_{q2}}{\sqrt{2}}\exp\{i(\omega_0-\omega_q)t\}, \tag{75}$$

with trivial solutions $A_{00}(t)=A_{11}(t)=0$. The transition probability for both states $\Phi^{\pm}$ is given by the expression $w(t)=d(|A_{10}(t)|^2+|A_{01}(t)|^2)/dt$. After some elementary manipulations, in the limit $t\to\infty$ we obtain

$$w(\infty)=\pi(|g_{q1}|^2+|g_{q2}|^2)\delta(\omega-\omega_0). \tag{76}$$

Analogous consideration for the states $\Psi^{\pm}$, for those $C_{00}=C_{11}=0$, $C_{01}=1/\sqrt{2}$, and $C_{10}=\pm 1/\sqrt{2}$, leads to the equations

$$\frac{d}{dt}A_{00}=-i\frac{1}{\sqrt{2}}(g_{q1}\pm g_{q2})\exp\{i(\omega_0-\omega_q)t\}, \tag{77}$$

and $A_{10}(t)=A_{01}(t)=A_{11}(t)=0$. Consequently, the transition probability for $\Psi^{\pm}$ is as follows:

$$w(\infty)=\pi|g_{q1}\pm g_{q2}|^2\delta(\omega-\omega_0). \tag{78}$$

Thus, Eqs. (76) and (78) show that the QD response to the Bell states of light contains only one spectral component with the unshifted frequency ω_0. To interpret this fact physically let us evaluate mean value of the electric field operator. For wave function defined by expression (71) this mean value has the form analogous to that given by Eq. (41):

$$\langle\hat{\mathbf{E}}\rangle=-2\sqrt{\frac{2\pi\hbar\omega_q}{\Omega}}\,\mathrm{Im}\sum_{i=0,1}\{[\mathbf{e}_1(A_{1i}A_{0i}^*+B_{1i}B_{0i}^*)$$
$$+\mathbf{e}_2(A_{i1}A_{i0}^*+B_{i1}B_{i0}^*)]e^{i(\mathbf{q}\mathbf{r}_c-\omega_q t)}\}. \tag{79}$$

From this relation follows that $\langle\hat{\mathbf{E}}\rangle=0$ for all Bell states. In other words, Bell states behave like single photons or any states with a fixed number of photons: their interaction with QDs does not induce macroscopic polarization and, consequently, does not produce depolarization field. Continuing this analogy, we can state that superposition of Bell states can induce in QD two lines separated by the gap $\Delta\omega$.

V. DISCUSSION

1. Observability of the depolarization effect

The basic physical result of the analysis presented in the given paper is the prediction of a fine structure of the absorption (emission) line in a QD interacting with quantum light. Instead of a single line with a frequency corresponding to the exciton transition ω_0, a doublet is appeared with one component shifted to the blue (red) by the value $\Delta\omega$ (12). We have revealed that the fine structure is due to depolarization of QD and has no analogs in classical electrodynamics. The value of the shift depends only on the geometrical properties of QD while the intensities of components are completely determined by statistics of the quantum light. It has been shown that in the limiting cases of classical light and single-photon states the doublet is reduced to a singlet shifted in the former case and unshifted in the latter one. Let us estimate the shift using well-known data for QD characteristics. For that aim, we rewrite Eq. (12) using Eq. (39), corrected to the host medium influence by means of substitutions (1):

$$\Delta\omega=\frac{\pi N_x}{V\tau}\left(\frac{c}{\sqrt{\varepsilon_h}\omega_0}\right)^3. \tag{80}$$

For a GaAs spherical QD ($N_x=1/3$) with the radius $R\simeq 3$ nm, dielectric constant $\varepsilon_h=12$ and radiation lifetime $\tau=1/\Gamma_{sp}\simeq 1$ ns [1], at the wavelength $\lambda=1.3$ μm formula Eq. (80) gives $\hbar\Delta\omega\simeq 1$ meV. This value correlates well with the theoretical estimate given in Ref. [21] and is of the same order of magnitude as polarization-dependent splitting described in Refs. [17,18]. Note that the Bohr radius for such QDs is about 10 nm [58], so that the strong confinement approximation used in our paper is valid. The frequency gap of the order of 1 meV has been observed in Refs. [47]. Recent low-temperature measurements of the QD dipole moment [6] give $\tau\simeq 0.05-0.15$ ns. However, QDs studied in Ref. [6] have lateral extension much larger then their thickness and the Bohr radius. Since $N_x\to 0$ in this case, we do not predict an observable depolarization shift for such QDs.

For experimental detection of the predicted fine structure, the value $\hbar\Delta\omega$ must exceed the linewidths of the doublet components: $\Delta\omega\gg\Gamma_{sp}/2$ and $\Delta\omega\gg\Gamma_{hom}/2$, where Γ_{hom} is the homogeneous broadening of the spectral line due to dephasing. As follows from Eq. (80), the first inequality is fulfilled at $N_x\gg(2\pi)^2V/\lambda^3$, i.e., for any realistic arbitrary shaped QDs. Analysis shows that the dominant contribution to the magnitude of Γ_{hom} gives exciton-phonon interaction. Recent low-temperature ($T=20$–40 K) measurements [11,59,60] give $\hbar\Gamma_{hom}\sim 1-20$ μeV. Analogous estimate follows from calculations presented in Ref. [61] at $T=77$ K. Thus, at low temperatures the predicted value of the shift turns out to be sufficiently large to be measured. At room temperatures the quantity $\hbar\Gamma_{hom}$ growths up to $0.2-1$ meV [1,60,61]. In such a situation line broadening may result in overlapping of the doublet components. However, even in that case local-field effects are of importance for adequate prediction of the spectral line shape of QD illuminated by quantum light.

2. Physical interpretation

Physical interpretation of the depolarization effect can be given by analogy with the $\mathbf{k}\cdot\mathbf{p}$–theory of bulk crystals [62] utilizing the concept of the electron-hole effective mass. For spherical QD, using the expressions for ω_0 [14] and for $\Delta\omega$ (12), we obtain

$$\hbar\omega_0+\hbar\Delta\omega=\epsilon_g+\frac{\hbar^2\kappa_{nl}^2}{2R^2M}+\frac{|\mu|^2}{R^3}, \tag{81}$$

where M is the mass of electron-hole pair in QD, ϵ_g is the width of the forbidden band gap, κ_{nl} is the nth root of the Bessel function $J_{l+1/2}(x)$, indices n and l defines the working mode in the quantum oscillator spectrum. The third term in the right-hand part of the equation describes contribution of the depolarization field. The right-hand part of the equation can be rewritten as $\epsilon_g+\hbar^2\kappa_{nl}^2/2R^2M_{eff}$ with M_{eff} given by

$$M_{eff}=\frac{M}{1+\dfrac{2|\mu|^2M}{\hbar^2\kappa_{nl}^2R}}. \tag{82}$$

The quantity M_{eff} can be interpreted as effective mass of the electron-hole pair in the QD. Thus, electromagnetic edge effects at the QD boundary responsible for the QD depolarization, change the exciton effective mass. Analogous consideration for the case of asymmetrically shaped QDs leads to the tensorial effective mass that gives rise to the polarization-dependent splitting of the gain band predicted and detected in Ref. [17].

In our paper we have only taken into account the local fields due to the QD boundary. In Ref. [63] is stated that the dipole-dipole interaction entailed by the electromagnetic field inhomogeneity on the interatomic scale [37,38], excites the local fields that compensate completely contribution from the boundary. Accordingly to Ref. [63], electromagnetic field acting on exciton in QD differs from the mean (on the interatomic scale) field which is considered in our theory as acting field inside QD, see Eq. (3). However, we cannot agree with the statement of Ref. [63]. Indeed, typical exciton Bohr radius exceeds significantly the interatomic distance and, thus, exciton wave function is disposed over a volume large on the interatomic scale (QD volume in the strong confinement regime and sphere with the Bohr radius in the weak confinement regime). This means that exciton feels the electromagnetic field averaged over the QD (or exciton) volume; consequently, the use of Eq. (3), the key relation in our theory, is physically justified.

3. Energy balance

Another key result of the paper is that the depolarization shift (if it exists) has opposite signs for absorptive and inverted exciton levels. This property of QDs exposed to classical light has been elucidated in Refs. [15,17] on the basis of classical electrodynamics. As a rule, results obtained for the classical light are extended to the quantum light using the concept of the Einstein coefficients [48]. In the case under consideration, such a transformation being applied to single-photon states leads to the energetic paradox: energies of absorbed and emitted photons differ by the quantity $2\hbar\Delta\omega$. At the first glance, there is no physically correct interpretation to this energy defect. However, the presented theory withdraws this paradox. Indeed, in accordance with Sec. III C, the single-photon processes are insensitive to the depolarization field, so that the spontaneous emission and absorption of a single-photon occur at the same resonant frequency ω_0. As it was stated in Sec. III D, depolarization shift occurs only in QDs exposed to the light with fluctuating number of photons (classical light is the limiting case of such states of electromagnetic field); in that situation, the energy defect $2\hbar\Delta\omega$ can physically be interpreted in the following way. The defect $2\hbar\Delta\omega$ is stipulated in the total nonclassical Hamiltonian (18) by the term ΔH defined by Eq. (A14). This equation describes electromagnetic interaction of oscillating electron and hole. In the QED, this interaction is transferred by a virtual photon with the energy $\hbar\Delta\omega$, which is extracted from the external field and returns back at random fashion. Obviously, such an interaction mechanism is impossible in external fields with a fixed number of photons, like the Fock states. Namely by this reason the depolarization field is not excited in QDs exposed to the Fock states and, consequently, the depolarization shift does not exist.

As it is mentioned in Appendix A, the term ΔH (A14), judging from its appearance, corresponds to the longitudinal electromagnetic oscillations in QD [50,64]. Thus, the exchange mechanism described above can be treated as physical interpretation from the QED standpoint of the formation in QD of the such longitudinal electromagnetic field. If we neglect the retardation inside the QD [see Eq. (A13)], the interaction process is reduced to the dynamical Coulomb interaction of electrons and holes in QD. Thus, effect of the depolarization field in QDs is analogous to effect of the spatial charge in free-electron beams (or, e.g., in microwave electronic devices) [65]. However, mathematical formalism utilized in our paper and key results of the theory are drastically different from that presented in Ref. [65]. The reason is that in microwave electronic devices, classical electrons interact with classical electromagnetic fields, whereas in QDs quantum carriers interact with quantum light.

4. To the problem of quantum states sources testing

Predicted in our paper effect of fine structure of exciton spectral line may find a number of challenging potential applications. Recent progress in quantum optics has made possible single-photon and single Bell state sources for the generation of entangled states of light [66]. However, such sources are not perfect. For a realistic single-photon source, the dominant state $|1_k\rangle$ is accompanied by a weak background of other states. Analogous situation takes place for generators of entangled states. As it has been shown in our paper, the QD electromagnetic response to a single-photon or single Bell state from hypothetical idealistic source would contain unshifted single spectral line; presence of the background will manifest itself in appearance of the shifted line. Intensity of this line will inform us about contribution of the background and, consequently, about quality of the source. Thus, interaction of QDs with quantum light can be applied for testing of quantum light sources.

5. Outlook

Let us discuss now some prospects for further development of the presented theory. First, note that our model does not account for the real lineshape in the electromagnetic response: practically everywhere in the paper (excluding Sec. III C) the line is approximated by the Dirac δ–function. As the next step, radiative and nonradiative mechanisms of relaxation should be involved into consideration. Nonradiative processes can be taken into account both phenomenologically (approximation of the longitudinal and transverse relaxation time [37]) and microscopically [a special term responsible for the phonon and electron-phonon interactions [67] is introduced in total Hamiltonian (18)]. To include radiative corrections into treatment, higher terms in the expansion in k of the kernel of integral (A1) must be taken into account. Even in this case we come to relation (A3); however, depolarization tensor $\underline{\mathbf{N}}$ becomes complex valued [16,68].

Another essential assumption of the present theory is that the electric field is assumed to be homogeneous over QD. Thus, nonlocal effects in QDs, related namely to the inhomogeneity, remain beyond the scope of the paper. For the classical light, theory of nonlocal effects in QDs is well developed [19,49,50,64,69,70]. Elaboration of corresponding theory for QDs exposed to quantum light is a self-maintained problem which will be considered elsewhere.

In the paper we have considered an isolated QD. Naturally, one can expect that the described effects will manifest themselves in different more complicated physical situations, where QDs interact with the quantum light, such as QD in a microcavity [9], response of a QD ensemble [71], role of the image effects [50], etc. The image effects originated from the discontinuity of the dielectric function nonresonant part at the QD boundary. In the general case of a dispersive and lossy medium, accounting for the image effects is a complicated problem which can be solved by introducing of the noise current [27,40]. The problem is significantly simplified if the nonresonant dielectric function is nondispersive and nonabsorptive.

In the absence of depolarization field ($\underline{\mathbf{N}}\rightarrow 0$) Hamiltonian (18) is reduced to the Friedrichs Hamiltonian [72,73]. Eigenstates of this Hamiltonian are so-called dress states (dress particles as well as dress photons). Fundamental theory of dress states, covering both stable and unstable cases, has been elaborated in Refs. [72,73]. The concept of dress states can be applied for further development of the theory presented. Indeed, eigenfunctions of Hamiltonian (18) can be expanded in the Friedrichs Hamiltonian eigenfunctions. As a result, a new system of dress states would appear with local-field effects incorporated. At that, the component ΔH can be considered as a small perturbation of the Friedrichs Hamiltonian. Such an approach may be found extremely useful under treatment of different problems of quantum optics of QDs.

VI. CONCLUSION

In the paper we have developed consecutive formalism of the electromagnetic field quantization in electrically small scatterers taking into account local fields. The formalism has been applied for the analysis of the role of local fields in electrodynamics of an isolated QD which is modeled as a spatially confined two-level quantum oscillator. As the first step, we have formulated *general self-consistent microscopic nonlinear equations* (11) describing interaction of an isolated QD with classical electromagnetic field. Physically, system is analogous to the Bloch equations for optically dense media. The general system derived can be applied to investigation of different nonlinear processes in QDs which are expected to be strongly influenced by the local fields.

In this paper, we generalize the system to nonclassical light. We have shown that the resonant interaction of nonclassical light with QD is realized via two different mechanisms. The first, quasiclassical, one is related to macroscopic polarization of QD in external electromagnetic field. This mechanism provides *depolarization shift* of the resonant frequency, blue for the ground-state QD and red for the excited one. Value of the shift depends only on the geometrical and electronic properties of QD and is independent on the incident light statistics. For typical semiconductor QDs, the shift is predicted to be of the order of several meV. Second mechanism of the QD-light interaction has quantum-electrodynamical origination and cannot be interpreted in the framework of classical electrodynamics. This mechanism leaves the resonant frequency unshifted. Thus, in our paper we predicts a fine structure of the absorption (emission) line in a QD interacting with the quantum light. Instead of a single line with a frequency corresponding to the exciton transition, a doublet is appeared with one component shifted to the blue (red). Proportion between intensities of components is completely determined by the quantum light statistics.

Both components of the doublet corresponds to the same field polarization and, consequently, splitting occurs even for symmetrically shaped QDs (sphere, cub). This distinguishes the predicted effect from the polarization-dependent splitting considered in Refs. [47,17,18].

In the limiting case of classical light the doublet is transformed into shifted single line reproducing thus semiclassical results obtained earlier [14–16]. Unlike that, interaction of QD with a single Fock state or single Bell state is characterized by a single unshifted resonance. In particular, emission of photon from QD (interaction with vacuum state $|0\rangle$) or its absorption (interaction with the state $|1\rangle$) occurs at the unshifted frequency. Physically it can easily be understood from the fact that both Fock and Bell states are characterized by the zero mean electric field and thus they do not induce macroscopic polarization in QD. Since macroscopic polarization that is responsible for the difference between operators of local and acting fields, this difference disappear for the Fock and Bell states. In general case, both lines are presented in the spectrum and have comparable intensities.

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APPENDIX A: RELATION BETWEEN ACTING AND LOCAL FIELDS IN QDS

1. Classical fields

We start with the frequency-domain integral relation for complex-valued field amplitudes [44,64]

$$\mathcal{E}_L(\mathbf{r})=\mathcal{E}_0(\mathbf{r})+(\nabla\nabla\cdot+k^2)\int_V \mathcal{P}(\mathbf{r}')\frac{\exp(ik|\mathbf{r}-\mathbf{r}'|)}{|\mathbf{r}-\mathbf{r}'|},\ d^3\mathbf{r}' \tag{A1}$$

which follows from the Maxwell equations; here $\mathbf{r}$, $\mathbf{r}'\in V$ and $\mathcal{P}(\mathbf{r})$ is the polarization amplitude defined by $\mathbf{P}(\mathbf{r},t)=\mathrm{Re}[\mathcal{P}(\mathbf{r})\exp(-i\omega t)]$. This relation couples the complex-valued amplitudes of the local $\mathcal{E}_L(\mathbf{r})$ and the acting $\mathcal{E}_0(\mathbf{r})$ fields inside QD. Letting the QD to be electrically small, we can neglect retardation in this equation and transform it to the equation for fields as follows:

$$\mathbf{E}_L(\mathbf{r},t)=\mathbf{E}_0(\mathbf{r},t)+\nabla\nabla\cdot\int_V \mathbf{P}(\mathbf{r}',t)\frac{d^3\mathbf{r}'}{|\mathbf{r}-\mathbf{r}'|}. \tag{A2}$$

Also, the above made supposition allows us to guess the acting field and, consequently, the polarization $\mathbf{P}$ to be constant over the QD volume. As a result, Eq. (A2) is transformed to

$$\mathbf{E}_L=\mathbf{E}_0-4\pi\underline{\mathbf{N}}\mathbf{P}. \tag{A3}$$

Here $\underline{\mathbf{N}}$ is the depolarization tensor those components are defined by

$$N_{\alpha\beta}=-\frac{1}{4\pi}\frac{\partial^2}{\partial x_\alpha \partial x_\beta}\int_V \frac{d^3\mathbf{r}'}{|\mathbf{r}-\mathbf{r}'|}. \tag{A4}$$

This tensor is symmetrical [44] 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 [43]:

$$N_{zz}=\frac{e^2+1}{e^3}(e-\arctan e),$$

$$N_{xx}=N_{yy}=\frac{1}{2}(1-N_{zz}), \tag{A5}$$

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 formulas hold true for both disklike ($a_{el}>b_{el}$) and cigarlike spheroids ($a_{el}<b_{el}$). Infinite stretching of the spheroids ($a_{el}/b_{el}\to 0$) results in $N_{zz}\to 0$, $N_{xx}\to 1/2$ and Eq. (A5) reproduce the polarizabilities of the cylinders (see, e.g., Ref. [17]). It should be noted that for an arbitrary three-axis ellipsoid, the tensor $\mathbf{N}$ does not depend on the coordinates. Consequently, the local field $\mathbf{E}_L(\mathbf{r})$ is also constant over the QD volume. For nonellipsoidal QDs, the tensor $\mathbf{N}$ and thus the local field become spatially inhomogeneous what contradicts to the basic assumption used in Sec. II A under formulation of the Hamiltonian. To eliminate the contradiction, we should average relation (A3) over the QD volume. This leads us again to Eq. (A3) with $\mathbf{E}_L,\mathbf{E}_0,\mathbf{P}=\mathrm{const}$ and

$$N_{\alpha\beta}=-\frac{1}{4\pi V}\int_V\int_V \frac{\partial^2}{\partial x_\alpha \partial x_\beta}\frac{d^3\mathbf{r}d^3\mathbf{r}'}{|\mathbf{r}-\mathbf{r}'|}. \tag{A6}$$

2. Quantum fields

At the first glance, for nonclassical fields Eq. (A3) remains valid if we insert operators instead corresponding fields. However, such is not the case and relation between acting and local fields in QED require a special discussion. Indeed, correct procedure of insertion of operators should be carried out in the time-domain integral Maxwell equation

$$\hat{\mathbf{E}}_L(\mathbf{r},t)=\hat{\mathbf{E}}_0(\mathbf{r},t)+\left(\nabla\nabla\cdot-\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\times\int_{-\infty}^{t}\int_V G^{(ret)}(\mathbf{r}-\mathbf{r}',t-t')\hat{\mathbf{P}}(\mathbf{r}',t')d^3\mathbf{r}'dt', \tag{A7}$$

where retarded Green function satisfies the equation

$$\left(\nabla^2-\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)G^{(ret)}(\mathbf{r},t)=\delta(\mathbf{r})\delta(t) \tag{A8}$$

with the initial condition $G^{(ret)}(\mathbf{r},t)=0$ at $t<0$. In accordance with Ref. [35], the retarded Green function is given by

$$G^{(ret)}(\mathbf{r},t)=\frac{1}{(2\pi)^4}\int\int\frac{\exp[i(\mathbf{k}\mathbf{r}-\omega t)]}{\mathbf{k}^2-(\omega+i\epsilon)^2/c^2}\,d^3\mathbf{k}\,d\omega, \tag{A9}$$

with $\epsilon\to+0$.

Polarization operator in Eq. (A7) is given by the relation $\hat{\mathbf{P}}(\mathbf{r},t)=V^{-1}(-\boldsymbol{\mu}b^\dagger+\boldsymbol{\mu}^*b)=V^{-1}(\boldsymbol{\mu}|e\rangle\langle g|+\boldsymbol{\mu}^*|g\rangle\langle e|)$. Thus, eigenstates of electron-hole pair in QD are generating functions for the polarization operator. These functions cannot be considered as slowly varying over the QD volume. Consequently, polarization operator in Eq. (A7) cannot be approximated by its value in a certain point of the space and removed from the integrand. Finally, we can conclude that relation (A3) cannot be automatically extended to the case of field operators.

To derive a relation for the field and polarization operators, we first construct the interaction Hamiltonian

$$H_{IL}=-\frac{1}{2}V(\hat{\mathbf{P}}\hat{\mathbf{E}}_L+\hat{\mathbf{E}}_L\hat{\mathbf{P}}). \tag{A10}$$

Here we took into account that the operators $\hat{\mathbf{P}}$ and $\hat{\mathbf{E}}_L$ are generally noncommutative since the field $\hat{\mathbf{E}}_L$ is not transversal [48] [second term in Eq. (A7) contains longitudinal component]. Next, we substitute into Eq. (A10) representation (A7) and separate out the Hamiltonian component corresponding to the depolarization field:

$$\Delta H = -\frac{1}{2}\left(\frac{\partial^2}{\partial x_\alpha \partial x_\beta} - \delta_{\alpha\beta}\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right) \times \int_{-\infty}^{t}\int_V G^{(ret)}(\mathbf{r}-\mathbf{r}',t-t')[\hat{P}_\alpha(\mathbf{r}',t')\hat{P}_\beta(\mathbf{r},t) + \hat{P}_\beta(\mathbf{r},t)\hat{P}_\alpha(\mathbf{r}',t')]d^3\mathbf{r}'\,dt'. \tag{A11}$$

This notation implies summation over repetitive indices.

The last equation can be drastically simplified by the field averaging over the QD volume. This procedure is analogous to that was used for the classical light under derivation of Hamiltonian (5). The spatial averaging technique turns out to be similar to the Hartry approximation in the self-consistent field method for electron-electron interactions [74]. In accordance with the method, we insert

$$\hat{P}_\alpha(\mathbf{r}',t') \rightarrow \langle \hat{P}_\alpha(\mathbf{r}',t')\rangle \hat{I} \tag{A12}$$

into Eq. (A11) and, then, approximate averaged value of the operator by a constant. This allows us to remove the averaged operator from the integrand of Eq. (A11). Further we take into account that QD is an electrically small object and neglect the retardation effects inside the QD. In such a case, the Green function reduces to

$$G^{(ret)}(\mathbf{r}-\mathbf{r}',t-t') \approx \frac{\delta(t-t')}{|\mathbf{r}-\mathbf{r}'|} \tag{A13}$$

and terms $O(\partial^2/\partial t^2)$ in Eq. (A11) can be omitted. As a result, Eq. (A11) reduces to the expression

$$\Delta H = 4\pi N_{\alpha\beta}\hat{P}_\beta\langle \hat{P}_\alpha\rangle, \tag{A14}$$

which corresponds to Hamiltonian (18) describing interaction of QD with nonclassical light. Analogous approximations being applied to Eq. (A7) lead to the formula

$$\hat{\mathbf{E}}_L = \hat{\mathbf{E}}_0 - 4\pi \underline{\mathbf{N}}\;\mathbf{P}\;\hat{I}, \tag{A15}$$

which offers for the nonclassical light an alternative to Eq. (A3). Note that this relation has been derived under assumption that the oscillators and the vacuum are separate systems. This assumption lies in the basis of the approach applied in Ref. [34] under construction of QED of an atom imbedded in a lossy dispersive dielectric medium. In accordance with Ref. [34], relation (A15) provides the relation $\Gamma'_{sp} = \sqrt{\epsilon_h}\Gamma_{sp}$ for the QD radiative linewidth in the medium.

APPENDIX B: CORRELATION BETWEEN SCHRÖDINGER AND HEIZENBERG REPRESENTATIONS

Interaction of QDs with classical and quantum light can be performed both in Schrödinger and Heizenberg representations. Both approaches are completely equivalent. However, for correlation between different publications, it would be helpful to establish correspondence of basic parameters and equations for these approaches. Since the paper uses the Schrödinger picture, below we formulate Heizenberg equations for Hamiltonian (5), carry out their transformation to the Bloch equations and then establish their correspondence to Eqs. (11). Classical electromagnetic field is only considered.

In accordance with Ref. [62], we present the Heizenberg equation in the following form:

$$i\hbar\frac{dO}{dt} = -[H,O], \tag{B1}$$

where O is an arbitrary operator. Letting $O=b$, we obtain

$$i\hbar\frac{db}{dt} = (\epsilon_e-\epsilon_g)b - 4\pi N_x P_x \mu(d_g^\dagger + d_e - \hat{I}) + E_{0x}\mu(d_g^\dagger + d_e - \hat{I}). \tag{B2}$$

By analogy, letting $O=d_g^\dagger = a_g a_g^\dagger$ and $O=d_e = a_e^\dagger a_e$, we derive

$$i\hbar\frac{d}{dt}d_g^\dagger = i\hbar\frac{d}{dt}d_e = -4\pi N_x P_x(\mu^* b + \mu b^\dagger) + E_{0x}(\mu^* b + \mu b^\dagger). \tag{B3}$$

Equations of motion for operators create a basis for derivation of modified Bloch equations. Indeed, let us present the acting field by $E_{0x} = \frac{1}{2}[\mathcal{E}(t)\exp(-i\omega t) + \text{c.c.}]$ with $\mathcal{E}(t)$ as slow-varying amplitude. The change over in Eqs. (B2) and (B3) to averaged values after averaging over the period $T = 2\pi/\omega$ gives the set of equation as follows:

$$i\hbar\frac{d\langle b\rangle}{dt} = \hbar\omega_0\langle b\rangle - \hbar\Delta\omega\langle b\rangle[\langle d_g^\dagger\rangle + \langle d_e\rangle - 1] + \frac{1}{2}\mathcal{E}(t)\mu[\langle d_g^\dagger\rangle + \langle d_e\rangle - 1]e^{-i\omega t}, \tag{B4}$$

$$i\hbar\frac{d\langle d_g^\dagger\rangle}{dt} = i\hbar\frac{d\langle d_e\rangle}{dt} = \frac{1}{2}[\mathcal{E}^*(t)\mu^*\langle b\rangle e^{i\omega t} - \text{c.c.}], \tag{B5}$$

where $\omega_0 = (\epsilon_e - \epsilon_g)/\hbar$. Note that the term related to the depolarization is absent in Eq. (B5). This is a result of averaging of Eq. (B3) over the period $T=2\pi/\omega$. Physically, system (B4)–(B5) is analogous to the system of Bloch equations for optically dense media derived in Ref. [32] [Eqs. (25) and (26)] on the basis of the Liouville equations.

Note that Eq. (10) for the macroscopic polarization gives the relations $\langle b^\dagger(t)\rangle \rightarrow -A^*(t)B(t)\exp(i\omega_0 t)$ and $\langle b(t)\rangle \rightarrow$

$A(t)B^*(t)\exp(-i\omega_0 t)$. Then, multiplying the first Eq. (11) by $B^*(t)$ and an equation complex conjugated to second Eq. (11) by $A(t)$, after summation of these equations we obtain

$$i\hbar\frac{d\langle b\rangle}{dt}=\hbar\omega_0\langle b\rangle-4\pi N_x\langle b\rangle\mu(|A|^2-|B|^2)+E_{0x}\mu(|A|^2-|B|^2). \tag{B6}$$

Next, let us multiply Eq. (11) by $A(t)$, and the complex conjugated equation by $A^*(t)$. Summation of these equations gives us

$$i\hbar\frac{d|A|^2}{dt}=\frac{1}{2}[\mathcal{E}^*(t)\mu^*\langle b\rangle e^{i\omega t}-\text{c.c.}]. \tag{B7}$$

Analogous procedure being applied to second Eq. (11) leads to

$$i\hbar\frac{d|B|^2}{dt}=-\frac{1}{2}[\mathcal{E}^*(t)\mu^*\langle b\rangle e^{i\omega t}-\text{c.c.}]. \tag{B8}$$

Composition of the last two equation results in the conservation law

$$\frac{d}{dt}(|A(t)|^2+|B(t)|^2)=0, \tag{B9}$$

which, obviously, can also be represented by $|A(t)|^2+|B(t)|^2=1$. The relations derived allow us to reveal the correspondence $|A|^2-|B|^2\rightarrow\langle d_g^\dagger\rangle+\langle d_e\rangle-1$, which leads us to conclusion that Eq. (B6) is identical to the Bloch equation for polarization while Eqs. (B7) and (B8) correspond to the Bloch equation for the charge density. Thus, the derived Eqs. (11) are completely equivalent to the Bloch equations (B2) and (B3).

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