Photon-statistics dispersion in excitonic composites

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Abstract. Linear media are predicted to exist whose relative permeability is an operator in the space of quantum states of light. Such media are characterized by a photon statistics-dependent refractive index. This indicates a new type of optical dispersion: the photon-statistics dispersion. Interaction of quantum light with such media modifies the photon number distribution and, in particular, the degree of coherence of light. An excitonic composite—a collection of non-interacting quantum dots—is considered as a realization of the medium with the photon-statistics dispersion. Expressions are derived for generalized plane waves in an excitonic composite and input–output relations for a planar layer of the material. Transformation rules for different photon initial states are analyzed. Utilization of the photon-statistics dispersion in potential quantum-optical devices is discussed.

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1. Introduction

The search for and investigation of novel physical processes transforming the photon number distribution is one of the most important problems of quantum optics [1]. Processes of that type known so far can be separated into two groups. The first group comprises different interference effects, such as two-beam interference of photons in a beam-splitter [1]. In some schemes, the use of ancillary photons and conditional detection enables the realization of effective nonlinear interaction by means of linear optics [2]. Besides their fundamental importance, such processes have tremendous potential for quantum computing. In particular, transformations of photon statistics such as nonlinear sign shifts [2, 3] and arbitrary photon-number-state filtering [4] have been realized. The manifestation of phase-dependent photon statistics in the mixed field of a narrow band two-photon source and coherent field has been reported in [5]. Photon antibunching as well as photon bunching have been observed. One can expect that this scheme should be able to produce effectively sub-Poissonian photon statistics.

The second group is formed by the processes in systems with nonlinearity and gain, where the transformation occurs due to the strong light–matter coupling [1]. A major development is that of single-photon sources [6]. The strong light–matter coupling enables different methods of generation of the sub-Poissonian photon statistics, such as micromasers [1], single-atom resonance fluorescence [7], single-trapped-atom lasers [8] and cavity-QED lasers [9]. In [10], a method has been proposed for the transformation of a vacuum state of a cavity mode into an arbitrary quantum state of light by means of a succession of strong atom–field interaction processes inside the cavity.

In the present paper, we present a new type of opportunity: transformation of the photon statistics in a linear homogeneous medium whose refractive index depends on the photon...
number distribution. As the physical realization of a medium with a photon statistics-dependent refractive index, we propose a collection of non-interacting quantum dots (QDs) embedded in a host semiconductor. This composite is called an excitonic composite [11, 12].

The optical and electronic properties of QDs are currently in vogue, due to their promising applications in semiconductor device physics [13] and, in particular, for quantum information storage and processing [14, 15]. The application of QDs as potential quantum-light emitters [6], [15]–[19] is now being intensively discussed. In particular, Regelman et al [19] demonstrate a quantum light source of multicolor photons with tunable statistics. The coupling of two QDs has been demonstrated as an effective means of antibunching for emitted photons [20].

A peculiar property of a QD exposed to an electromagnetic field is the pronounced role of the dipole–dipole electron–hole interaction. Phenomenologically, this interaction can be introduced through the local fields [21]–[25]. Obviously, in quantum electrodynamics, the dipole–dipole interaction is due to the exchange between electrons and holes by virtual vacuum photons [26]. Slepyan and Maksimenko et al [21, 22] predict a fine structure of the absorption (emission) line in a QD interacting with quantum light. Instead of a single line at the exciton transition frequency $\omega_0$, a doublet appears with one component blue (red) shifted by a value $\Delta \omega$. The fine structure has no analog in classical electrodynamics. The value of the shift depends only on the QD shape, while the intensities of components are completely determined by the light statistics. In the limiting cases of classical light and single Fock states, the doublet reduces to a singlet shifted in the former case and unshifted in the latter one. In heterogeneous media comprising regular or irregular arrays of QDs embedded in a semiconducting host, this mechanism is responsible for the photon-statistics dispersion mentioned earlier.

The electromagnetic response properties of a composite medium comprising electrically small (of dimension much smaller than the wavelength) inclusions exposed to classical light can be modeled within the effective-medium approach modified [11, 12] to include specific properties of excitons as resonant states with a discrete energy spectrum. To solve the problem of the quantum light interaction with a composite medium, an adequate quantization technique must be developed. Conventionally, particular models of the classical light interaction with homogeneous media are exploited for this purpose. Non-homogeneities are implemented into models by means of homogenization procedures on the classical stage. However, the subsequent replacement of classical fields by corresponding field operators often leads to a fundamental problem, which is the lack of correct commutation relations for the field operators. To overcome this problem, some modifications of the quantization scheme have been proposed, such as the noise current concept [27]. Nevertheless, in all the cases, to our knowledge, the constitutive parameters in classical and quantum optics are assumed to be identical.

Here, we propose an alternative approach: first we consider the interaction of quantum light with an isolated scatterer (QD) and then we develop a corresponding homogenization technique. This approach exploits the well-known technique [28, 29] linking the macroscopic refractive index to the forward scattering amplitude $f(0)$ by

$$n^2 = 1 + \frac{4\pi\rho}{k^2} f(0),$$  \hspace{1cm} (1)

where $k = \omega/c$ is the vacuum wavenumber, $c$ is vacuum speed of light and $\rho$ is the density of scatterers in the medium. Within this approach, the quantum field operators satisfy the correct commutation relations automatically. Note that the approach is applicable not only to photons.
but also to quantum fields of other origination (atoms and atomic nuclei, neutrons, etc) and is based on nuclear optics of polarized media [30].

The paper is arranged as follows. In section 2, we present a model of the single QD–quantum light interaction and evaluate the forward scattering amplitude operator. Primary attention is paid to accounting for the local-field effects. The corresponding component of the total Hamiltonian is deduced by means of the BBGKY (Bogoliubov–Born–Green–Kirkwood–Yvone)-hierarchy analysis. The fundamentals of quantum optics of excitonic composites—constitutive relations for field operators, planewave solution of Maxwell equations and input–output relations for a planar layer—are formulated in section 3. Section 4 is devoted to the photon-statistics dispersion. A general formulation of the transformation law for the light density operator and the second-order coherence correlation function are set up and some particular examples of different initial quantum states are considered. A summary of this work and outlook are given in section 5. The appendix provides some details of the derivations presented in section 2.

2. Interaction of quantum light with a single QD

2.1. Local-field corrections: the BBGKY-hierarchy analysis

We start with the evaluation of the forward scattering amplitude \( f(0) \) of quantum light interacting with a single QD. As aforestated, in our analysis, we are not restricted to the phenomenological Hartree–Fock–Bogoliubov approximation utilized in [21, 23] to account for the local-field effects; instead, our analysis is based on more general principles of quantum many-body theory. Namely, here we present the BBGKY-hierarchy analysis [31] of the problem. Also, in order to eliminate fast oscillations, we make use of the rotating-wave approximation [1].

As has been pointed out by Cho [32], in the formulation of the Hamiltonian of the interacting system ‘electromagnetic field + condensed matter’, the decomposition of the system into matter and field is ambiguous. Following the approach called scheme B in [32], we introduce the total electromagnetic field assuming carriers are not interacting directly. According to [32], the Hamiltonian of a QD exposed to a quantum electromagnetic field can be written as

\[
\hat{H} = \hat{H}_0 + \hat{H}_{ph} + \hat{H}_{tot}^I.
\]

Here \( \hat{H}_0 \) is the Hamiltonian of the charge carriers’ free motion. The term ‘free’ implies that the carriers are spatially confined in the QD but are free of the influence of the electromagnetic field and interparticle interactions. The Hamiltonian

\[
\hat{H}_{ph} = \int_0^\infty \hbar \omega \hat{a}^\dagger\omega \hat{a}\omega \, d\omega
\]

describes photons in free space in the single-mode approximation [1]. The index \( \omega \) ascribes the creation/annihilation operators to a photon of frequency \( \omega \). The creation/annihilation operators satisfy the commutation relation \( [\hat{a}\omega, \hat{a}^\dagger\omega'] = \delta(\omega - \omega') \).

The total field inside a QD is identical to the local one; correspondingly, the total interaction Hamiltonian in the length gauge is given by

\[
\hat{H}_{tot}^I = -\frac{1}{2} \int_V (\hat{\mathbf{P}} \hat{\mathbf{E}}_I + \hat{\mathbf{E}}_I \hat{\mathbf{P}}) \, d^3\mathbf{r}.
\]
In the photon basis, the local field operator is represented by
\[ \hat{E}_L = i \sum_{k,k'} \sqrt{\frac{2 \pi \hbar \omega_0}{\Omega}} e_{k,k'} (\hat{c}_{k,k} e^{i k r} - \hat{c}_{k,k}^\dagger e^{-i k r}). \] (5)

Here \( k \) is the mode index, the index \( \lambda = 1 \) and 2 denotes the field polarization, \( \Omega \) is the normalization volume, and \( \hat{c}_{k,k} \) and \( \hat{c}_{k,k}^\dagger \) are the creation and annihilation operators of the bosonic field. It should be emphasized that these operators are not equivalent to the creation/annihilation operators of real photons, \( \hat{a}_{\omega}^\dagger / \hat{a}_{\omega} \), present for example in (3). Equation (5) is fulfilled only in the QD volume.

Further we restrict ourselves to the two-level approximation for the QD exciton with \( |g\rangle \) and \( |e\rangle \) as its ground and excited states, and neglect the intraband transition spectrum. The exciton is assumed to be strongly confined in the QD. In the two-level approximation, the excitonic component of the density operator is reduced to a single-particle function. The electromagnetic field is assumed to be weak. Thus, we can neglect the multi-photon processes and, correspondingly, photon–photon correlations. In that case, three variables figure in the BBGKY hierarchy: the excitonic density operator \( \hat{F}_{\text{ex}} \), the photonic density operator \( \hat{F}_{\text{ph}} \) and the operator of exciton–photon correlations \( \hat{K} \). The partial density operators are determined by \( \hat{F}_{\text{ex,ph}} = \text{Tr}_{\text{ex,ph}} \hat{\rho}_{\text{tot}} \), where \( \hat{\rho}_{\text{tot}} \) is the total multi-particle density operator of the system ‘exciton + quantum light’ in a QD, and \( \text{Tr}_{\text{ex,ph}} \) denotes traces with respect to excitonic and photonic variables, respectively. In the cluster expansion [31, 33] \( \hat{\rho}_{\text{tot}} = \hat{K} + \hat{F}_{\text{ex}} \hat{F}_{\text{ph}} \) and the BBGKY hierarchy takes the form following [31]:

\[
\begin{align*}
\ih \frac{\partial}{\partial t} \hat{F}_{\text{ex}} - [\hat{H}_1, \hat{F}_{\text{ex}}] &= \text{Tr}_{\text{ph}} [\hat{H}_{1\text{tot}}, \hat{K}], \\
\ih \frac{\partial}{\partial t} \hat{F}_{\text{ph}} - [\hat{H}_2, \hat{F}_{\text{ph}}] &= \text{Tr}_{\text{ex}} [\hat{H}_{2\text{tot}}, \hat{K}], \\
\ih \frac{\partial}{\partial t} \hat{K} - [\hat{H}_1 + \hat{H}_2, \hat{K}] &= [\hat{H}_{1\text{tot}}, \hat{F}_{\text{ex}} \hat{F}_{\text{ph}}].
\end{align*}
\]

(6)

(7)

(8)

Here, \( \hat{H}_1 = \hat{H}_0 + \hat{U}_1, \hat{H}_2 = \hat{H}_{\text{ph}} + \hat{U}_2, \) and

\[
\begin{align*}
\hat{U}_1 &= \text{Tr}_{\text{ph}} (\hat{H}_{1\text{tot}} \hat{F}_{\text{ph}}) = - \int_V \langle \hat{P} \rangle \hat{E}_L d^3r, \\
\hat{U}_2 &= \text{Tr}_{\text{ex}} (\hat{H}_{1\text{tot}} \hat{F}_{\text{ex}}) = - \int_V \langle \hat{P} \rangle \hat{E}_L d^3r.
\end{align*}
\]

(9)

(10)

The quantity \( \hat{E}_L \) is determined by equation (5), \( \langle \hat{E}_L \rangle = \text{Tr}_{\text{ph}} (\hat{E}_L \hat{F}_{\text{ph}}) \) and \( \langle \hat{P} \rangle = \text{Tr}_{\text{ex}} (\hat{P} \hat{F}_{\text{ex}}) \). The correlation operator \( \hat{K} \) describes the frequency shift and the decay rate induced by the scattering and reabsorption of spontaneous photons. Formally solving equations (7) and (8)\(^2\) and substituting the result into (6), we derive the master equation for the single-exciton density operator analogous to equation (26) in [34].

For a small excitation, the partial excitonic density operator can be represented by \( \hat{F}_{\text{ex}} = |g\rangle \langle g| + \delta \hat{F}(t) \), where \( \delta \hat{F}(t) \) is a small correction and \( \delta \hat{F}(\infty) = 0 \). Then, in the first-order

\(^2\) The initial condition for equation (8) is \( \hat{K}(-\infty) = 0 \).

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approximation, the contribution of the correlation operator is negligible. Indeed, in our model, we are not interested in the decay processes, whereas accordingly [34], the frequency shift is determined by $i[\hat{\sigma}_-\hat{\sigma}_+ - \hat{\sigma}_+\hat{\sigma}_- + \hat{F}_{ex}]$. The real coefficient of this term in the master equation [34] is proportional to the second-order cumulant of the electromagnetic field, which is a higher-order infinitesimal if a coherent component is prevalent in free-space light. However, even in a general case it can easily be shown that at small excitation this quantity dictates only a small renormalization of the resonant transition frequency, inessential with respect to the processes considered. Thus, the system (6)–(8) is reduced to a system of generalized Maxwell–Bloch equations [31, 34] accounting for the scattering of coherent photons stimulated by QD.

The next problem is to express the operator $\hat{U}_1$ (9) in terms of the polarization operator and the free-field operator. We start with operator (5) and present its mean value as

$$\langle \hat{\mathcal{E}}_k(r, t) \rangle = i \sum_{kl} e_{kl} \sqrt{\frac{2\pi \hbar \omega_k}{\Omega}} \langle \hat{c}_{kl}(t) \rangle e^{ikr} + \text{c.c.},$$

(11)

where $\langle \hat{c}_{kl}(t) \rangle = \text{Tr}_{ph}(\hat{c}_{kl} \hat{F}_{ph}(t))$. From equation (7), we then obtain

$$\frac{d}{dt} \langle \hat{c}_{kl}(t) \rangle = -i\omega_k \langle \hat{c}_{kl}(t) \rangle + \sqrt{\frac{2\pi \hbar \omega_k}{\hbar \Omega}} e_{kl} \int_V \langle \hat{\mathcal{P}}(r, t) \rangle e^{-ikr} d^3r.$$  

(12)

By solving this equation and substituting (11) into (9), we find the operator $\hat{U}_1$ as

$$\hat{U}_1 = -\int_V \hat{\mathcal{P}}(\hat{\mathcal{E}}_0) d^3r - \Delta \hat{H}.$$  

(13)

For details see the appendix. The free electric field operator $\hat{\mathcal{E}}_0$ in the first term of (13) is determined in the whole space and can be represented by the superposition $\hat{\mathcal{E}}_0(r) = \hat{\mathcal{E}}^{(+)}_0(r) + \hat{\mathcal{E}}^{(-)}_0(r)$, where superscripts $(\pm)$ mark the contributions from positive and negative frequencies, respectively. Thus, the time-domain operator $\hat{\mathcal{E}}^{(+)}_0(r)$ is associated with the frequency-domain field by the positive semi-axis Fourier transform:

$$\hat{\mathcal{E}}^{(+)}_0(r) = \int_0^\infty \hat{\mathcal{E}}_0(r, \omega) d\omega.$$  

(14)

The operators $\hat{\mathcal{E}}^{(-)}_0(r)$ and $\hat{\mathcal{E}}^{(+)}_0(r)$ are the Hermitian conjugates. We denote the spectral amplitude $\hat{\mathcal{E}}_0(r, \omega)$ by the unified symbol throughout the entire frequency axis and we imply that the amplitude satisfies the condition $\hat{\mathcal{E}}_0(r, -\omega) = \hat{\mathcal{E}}^{+}_0(r, \omega)$. In the single-mode approximation

$$\hat{\mathcal{E}}_0(r, \omega) = i\sqrt{\frac{\hbar}{A}} e_y \hat{a}_\omega e^{ikz},$$

(15)

where $e_y$ is the unit polarization vector of free photons and $A$ is the normalization cross-section in the $x$–$y$-plane. Differently from (5), equation (15) is fulfilled in the entire space.

In accordance with (A.5), the second term in (13) is given by

$$\Delta \hat{H} = -\int_V \int_V \hat{\mathcal{P}}(r) \hat{G}(r - r\prime) \langle \hat{\mathcal{P}}(r\prime, t) \rangle d^3r d^3r\prime.$$  

(16)

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As a result, we come to the light–matter Hamiltonian
\[\hat{\mathcal{H}} = \hat{\mathcal{H}}_0 + \hat{\mathcal{H}}_{\text{ph}} + \hat{\mathcal{H}}_1 + \Delta \hat{\mathcal{H}},\] 
(17)
where \(\hat{\mathcal{H}}_0\) and \(\hat{\mathcal{H}}_1\) are determined by the standard expressions [1]:
\[\hat{\mathcal{H}}_0 = \frac{\hbar}{2} \omega_0 \hat{\sigma}_z,\] 
(18)
\[\hat{\mathcal{H}}_1 = \hbar \int_0^\infty \left( g_\omega \hat{\sigma}_+ \hat{a}_\omega + g_\omega^* \hat{\sigma}_- \hat{a}_\omega^\dagger \right) d\omega.\] 
(19)
Here, \(\omega_0\) is the resonant frequency of the transition in QD and \(\hat{\sigma}_{z,\pm}\) are the Pauli pseudospin operators. The quantity \(g_\omega = -i \sqrt{\omega / \hbar c A}\) is the coupling factor for photons and carriers in QD; \(\mu = (\mu_{eg} e_y) e_y = (\mu_{ge} e_y) e_y\) and \(\mu_{eg}\) is the transition dipole moment.

Taking into account that in the two-level approximation and in the strong confinement regime, the polarization operator is presented by
\[\hat{\mathcal{P}}(r) = |\xi(r)|^2 \mu^* \hat{\sigma}_- + \text{H.c.},\] 
(21)
the expression
\[\Delta \hat{\mathcal{H}} = \hbar \Delta \omega (\hat{\sigma}_- \langle \hat{\sigma}_+ \rangle + \hat{\sigma}_+ \langle \hat{\sigma}_- \rangle)\] 
(22)
can easily be obtained from (16). Here, the depolarization shift
\[\Delta \omega = \frac{4\pi}{\hbar V} \mu (\mathcal{N} \mu)\] 
(23)
characterizes the role of local fields inside a QD. For a QD exposed to a classical electromagnetic field, the depolarization shift \(\Delta \omega\) has been introduced in [35, 36]. The depolarization tensor is given by [22]
\[\mathcal{N} = -\frac{V}{4\pi} \int_V \int_V |\xi(r)|^2 |\xi(r')|^2 \nabla_r \otimes \nabla_{r'} \left( \frac{1}{|r - r'|} \right) d^3r d^3r',\] 
(24)
where \(\nabla_r \otimes \nabla_r\) is the operator dyadic acting on variables \(r\) and \(\xi(r)\) is the wavefunction of the strongly confined exciton, the same in excited and ground states [37].

Thus, the system (6) and (7) with \(\hat{K} = 0\) corresponds to the Hamiltonian (4) with \(\hat{\mathcal{E}}_L\), determined by
\[\hat{\mathcal{E}}_L(r) = \hat{\mathcal{E}}_0 - \frac{4\pi}{V} (\mathcal{N} \mu \langle \hat{\sigma}_+ \rangle + \text{H.c.}).\] 
(25)
This relation demonstrates that the local field manifests itself as the QD depolarization, i.e. the screening of external field by charges induced on the QD surface. This result demonstrates that the motion of an isolated exciton occurs in the effective local field of the form of (25).

Certainly, the QD exciton also interacts with the photon reservoir. This interaction dictates the natural linewidth and fluctuation currents in the QD. In this paper, these factors are not taken into consideration and, hence, the corresponding component of the Hamiltonian has been
omitted in (2) and, consequently, in (4). Note that the local-field impact cannot be reduced to the interaction of the exciton with the excitonic reservoir. Indeed, the reservoir is not affected by the QD exciton, while the local field correction results from the self-consistent interaction of an ensemble of particles in QD. The self-consistency is reflected by the structure of the Hamiltonian (4) and the subsequent equation (25).

Differently from previous considerations [21, 23], here we have obtained the depolarization field as a C-number without phenomenological mean-field averaging. A similar approach has been used by Fleichhauer and Yelin in [34] in their analysis of the Lorenz–Lorentz correction for quantum light in optically dense media (see equation (31) and the comments in [34]). It has been noted in [34] that the Lorenz–Lorentz correction for quantum light cannot be obtained by the direct replacement of classical fields by corresponding field operators. With respect to the QD exciton interacting with quantum light, the statement can be illustrated as follows. The replacement \( \langle \hat{\sigma}_+ \rangle \rightarrow \hat{\sigma}_+ \) in equation (25) means, in fact, the replacement \( \Delta \hat{H} \rightarrow \hbar \Delta \omega (\hat{\sigma}_+ \hat{\sigma}_- + \hat{\sigma}_- \hat{\sigma}_+) \). It can easily be found that in this case the Hamiltonian responsible for the local-field effects becomes a C-number, but that means the complete elimination of the local-field effects. Seemingly, on that ground one can conclude that local-field effects cannot be accounted for within the two-level model. However, such a conclusion would contradict the principle of correspondence since it does not provide a limit transition from quantum optics to the classical case. Indeed, in classical optics of QDs, the local-field effects are described within the two-level model and determined by the Hamiltonian (22) [11, 12]. Actually, the absence of quantum fluctuations of the depolarization field follows from the fact that the depolarization field in the quasi-static approximation is longitudinal and thus is not quantized. That is, this field is not expressed in terms of photonic operators and, consequently, does not experience quantum fluctuations.

The analysis presented clearly shows us that equation (25) holds true under two important restrictions: the system is assumed to be two-level and retardation of electromagnetic field in the QD is neglected (electrically small scatterer). Note that these two assumptions are generally not independent. Neglecting retardation is maybe not unreasonable beyond the two-level model, when excitonic lines are located closely enough. However, consistent accounting for the retardation requires, in the general case, the refusal of the two-level model. The reason is that, because of diffraction, the spatial distribution of the electromagnetic field inside a QD becomes so complicated that it is no longer described by the wavefunction of an isolated resonant exciton. As an example, we mention the role of Mie-resonances in spherical QDs [24, 32].

In the general case, operators of electromagnetic field and polarization are related to each other by the wave equation. Going to its integral form, the relation can be considered as the Lorenz–Lorentz correction

\[
\hat{E}_L^{(+)} = \hat{E}_0^{(+)} - 4\pi \hat{N} \hat{P}^{(+)},
\]

where the generalized (non-local in space and in time) depolarization tensor is determined by

\[
\hat{N} \hat{P}^{(+)}(\mathbf{r}, t) = -\frac{1}{4\pi c^3} \frac{\partial^2}{\partial t^2} \int_V \hat{P}^{(+)}(\mathbf{r'}, t - \frac{|\mathbf{r} - \mathbf{r'}|}{c}) \frac{d^3\mathbf{r'}}{|\mathbf{r} - \mathbf{r'}|}.
\]

An analogous expression can be written for the negative-frequency component of the local field with the replacement of the retarded potential by the advanced one. As one can see, the right-hand side of this equation contains the operator \( \hat{P}^{(+)} \) but not its mean value \( \langle \hat{P}^{(+)} \rangle \); this indicates the presence of quantum fluctuations of the polarization.

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The rejection of the assumptions prescribed leads to the conclusion that the excitonic component of the density operator $\hat{P}_{\text{tot}}$ becomes multi-particle. In that case, the transition to the BBGKY hierarchy implies the introduction of exciton–exciton correlations of different orders as independent dynamical variables and projecting equation (27) on different excitonic states by means of operation $\text{Tr}$. If one neglects the exciton–exciton correlations, the above mentioned projection means the replacement $\hat{P}^{(\mu)} \rightarrow \langle \hat{P}^{(\mu)} \rangle$; that is, the transition to the Hartree–Fock–Bogoliubov approximation \[.\] Physically this means the replacement of the actual local field (26) by an effective field determined by equation (25). Accounting for correlations of all orders corresponds to exact fulfillment of the relation (27). Restriction to low-order exciton–exciton correlations (e.g. two-particle) means approximate accounting for the quantum fluctuations of the depolarization field in a weak electromagnetic field. Further, we neglect the quantum fluctuations in view of their smallness within the assumptions prescribed.

The simplest procedure to take into account the multi-level effects is as follows. The joint contribution of all transitions lying far away from a given resonance is approximated by a non-resonant relative permittivity $\varepsilon_h$. If we assume the host semiconductor’s relative permittivity to be equal to $\varepsilon_h$ and, for analytical tractability, suppose $\varepsilon_h$ to be frequency-independent and real-valued, we can set $\varepsilon_h = 1$ without loss of generality. The substitutions $c \rightarrow c/\sqrt{\varepsilon_h}$ and $\mu \rightarrow \mu/\sqrt{\varepsilon_h}$ in the final expressions will restore the case $\varepsilon_h \neq 1$.

2.2. Equations of motion

In the previous section, we have formulated the light–matter interaction Hamiltonian (17) and defined all its components in terms of the operators $\hat{a}_\omega$, $\hat{a}^\dagger_\omega$, $\hat{\sigma}_\pm$ and $\hat{\sigma}_z$ by equations (3) and (18)–(22). The Hamiltonian (17) differs from Hamiltonians conventionally used in quantum optics of QDs by the term $\Delta \hat{H}$ accounting for the local-field effects. This term dictates peculiar quantum-optical properties (particularly, the effect of photonic-state dispersion) of excitonic composites, which are the focus of the present paper.

In the Heisenberg representation, the equations of motion for operators $\hat{a}_\omega$, $\hat{\sigma}_-$ and $\hat{\sigma}_z$ are given by

$$\frac{\partial}{\partial t} \hat{\sigma}_- = -i\omega_0 \hat{\sigma}_- + i\hat{\sigma}_z \int_0^\infty g_\omega \hat{a}_\omega \, d\omega + i\Delta \omega \hat{\sigma}_z \langle \hat{\sigma}_- \rangle,$$

(28)

$$\frac{\partial}{\partial t} \hat{a}_\omega = -i\omega \hat{a}_\omega - ig_\omega^* \hat{\sigma}_-,$$

(29)

$$\frac{\partial}{\partial t} \hat{\sigma}_z = 2i \int_0^\infty (g_\omega^* \hat{a}_\omega^\dagger \hat{\sigma}_- - g_\omega \hat{a}_\omega \hat{\sigma}_+^\dagger) \, d\omega + 2i\Delta \omega \langle \hat{\sigma}_- \langle \hat{\sigma}_+ \rangle \rangle,$$

(30)

At the initial time $t = -\infty$, let the QD be in the ground state $|g\rangle$ ($\langle \hat{\sigma}_z(-\infty) \rangle = -1$) and be exposed to an arbitrary quantum state of light. The interaction is assumed to be weak, i.e. only first-order terms in $g_\omega$ are of importance. From equation (28), it follows that $\hat{\sigma}_- \text{ and } \langle \hat{\sigma}_- \rangle \sim O(g_\omega)$, which means that $\partial \hat{\sigma}_z/\partial t \sim O(|g_\omega|^2)$ in (30). Consequently, the weak interaction model allows us to put $\hat{\sigma}_z(t) = \hat{\sigma}_z(-\infty)$ in the further analysis. Since the final expressions will obligatorily involve averaging over the ground state, i.e. the quantity $\langle \hat{\sigma}_z(t) \rangle \approx \langle \hat{\sigma}_z(-\infty) \rangle = -1$, New Journal of Physics 10 (2008) 023032 (http://www.njp.org/)
it is convenient to make the replacement $\hat{\sigma}_z \rightarrow -1$ straightaway in equation (28). As a result we come to

$$\frac{\partial}{\partial t} \langle \hat{\sigma}_- \rangle = -i(\omega_0 + \Delta \omega) \langle \hat{\sigma}_- \rangle - i \int_0^\infty g_\omega (\hat{A}_\omega) \, d\omega.$$  \hspace{1cm} (31)

The integration of this equation with respect to time yields

$$\langle \hat{\sigma}_-(t) \rangle = -i \int_0^t g_\omega \langle \hat{A}_\omega(t') \rangle \, e^{-i(\omega_0 + \Delta \omega)(t-t')} \, dt' \, d\omega.$$  \hspace{1cm} (32)

The substitution of (32) into (28) gives us

$$\frac{\partial}{\partial t} \hat{\sigma}_- = -i \omega_0 \hat{\sigma}_- - i \int_0^\infty g_\omega \hat{A}_\omega(t) \, d\omega,$$  \hspace{1cm} (33)

where

$$\hat{A}_\omega(t) = \hat{a}_\omega(t) - i \Delta \omega \int_{-\infty}^t \langle \hat{a}_\omega(t') \rangle \, e^{-i(\omega_0 + \Delta \omega)(t-t')} \, dt'.$$  \hspace{1cm} (34)

By integration of (33) we obtain the explicit relation

$$\hat{\sigma}_-(t) = \hat{\sigma}_-(-\infty) - i \int_0^\infty g_\omega \int_{-\infty}^t \hat{A}_\omega(t') \, e^{-i\omega_0(t-t')} \, dt' \, d\omega$$  \hspace{1cm} (35)

between the operators $\hat{a}_\omega$ and $\hat{\sigma}_-$, which defines the interaction of the QD exciton with quantum light in the weak-field limit. This equation serves as a basis for the further analysis, because it allows us to develop the correlation between the electric field and the polarization operators in QD. Going to the frequency domain and utilizing the relations (15), (21), (34) and (35), we come to the frequency-domain polarizability operator as

$$\hat{P}(\mathbf{r}) = -\frac{\mu^2}{\omega - \omega_0 + i0} \left[ \hat{E}_0(\mathbf{r}) + \frac{\Delta \omega}{\omega - \omega_0 - \Delta \omega + i0} \hat{E}_0(\mathbf{r}) \right]$$  \hspace{1cm} (36)

with $\hat{E}_0(\mathbf{r})$ given by equation (15). In (36), the quantity $\hat{P}(\mathbf{r}, \omega) = \hat{P}(\mathbf{r})$ is the spectral amplitude of the operator $\hat{P}(\mathbf{r})$, defined by the relation analogous to equation (14) for the electric field. In the same manner, further we introduce the spectral amplitude of the electric displacement $\hat{D}(\mathbf{r})$.

Let us restrict further consideration to monochromatic fields. This allows us to omit from here onwards the index $\omega$ in operators $\hat{a}_\omega$ and $\hat{a}_\omega^\dagger$ and omit explicit mention of the frequency dependence in complex-amplitude operators.

2.3. **Forward scattering amplitude**

Let the field $\hat{E}_0(\mathbf{r})$ be scattered by an isolated QD. Obviously, the total field $\hat{E}(\mathbf{r})$ outside the QD is a superposition of the incident and the scattered fields. In terms of the polarization induced in the QD, this field is expressed by the relation

$$\hat{E}(\mathbf{r}) = \hat{E}_0(\mathbf{r}) + (\nabla \nabla \cdot + k^2) \int \hat{P}(\mathbf{r}') \frac{e^{ik|\mathbf{r}'-\mathbf{r}|}}{|\mathbf{r}' - \mathbf{r}|} \, d^3 \mathbf{r'},$$  \hspace{1cm} (37)
which follows from the Maxwell equations. In turn, the induced polarization is related to a weak incident field by the linear relation (36). Substitution of (36) into (37) allows to couple the incident and the total field, that is, to define the scattering operator [38].

The analogous relation with operators replaced by corresponding C-numbers holds for classical fields. For electrically small scatterers, the coupling between the polarization and the electric field is local. This allows the introduction of the forward scattering amplitude by

\[ P(r_0) = \frac{V}{k^2} f(0) E(r_0), \]  

where \( r_0 \) is the scatterer radius-vector. In the far zone, the scattered field is a diverging spherical wave; thus,

\[ E(r) = E_0(r) + \frac{V}{k^2} (\nabla \cdot + k^2) \frac{e^{ik|r-r_0|}}{|r-r_0|} f(0) E_0(r_0). \]  

Then, the forward scattering amplitude \( f(0) \) is found to be proportional to the QD polarizability [21] and is given by

\[ f(0) = - \frac{k^2 |\mu|^2}{\hbar V} \frac{1}{\omega - \omega_0 - \Delta \omega + i0}. \]  

Going to quantum light, we cannot restrict ourselves to the formal replacement of the electric field and polarization in (38) and (39) by corresponding operators, keeping the forward scattering amplitude in the form of (40). As follows from equation (36), because of the local field impact, the polarization operator is represented by a superposition of the electric field operator and its mean value. As a result, equation (36) cannot be reduced to (38) written for operators.

The necessary generalization of (38) exploits representation of the forward amplitude as an operator in the space of quantum states of light:

\[ \hat{P}(r_0) = \frac{V}{2k^2} [\hat{f}(0) \hat{E}_0(r_0) + \hat{E}_0(r_0) \hat{f}(0)]. \]  

As one can see, the product of the operators \( \hat{f}(0) \) and \( \hat{E}_0 \) enters this equation in symmetrized form, analogously to the interaction Hamiltonian (4). Using (15), (36) and (41), we derive the equation

\[ \hat{f}(0) \hat{a} + \hat{a} \hat{f}(0) = 2 f_2(0) \hat{a} + 2 \langle \hat{a} \rangle [f_1(0) - f_2(0)], \]  

for the forward scattering amplitude operator \( \hat{f}(0) \), where

\[ f_{1,2}(0) = - \frac{k^2 |\mu|^2}{\hbar V} \frac{1}{\omega - \omega_{1,2} + i0} \]  

are the partial forward scattering amplitudes with \( \omega_1 = \omega_0 + \Delta \omega \) and \( \omega_2 = \omega_0 \). To solve (42) with respect to \( \hat{f}(0) \), we introduce the operator

\[ \hat{\xi} = \sum_{m=0}^{\infty} \frac{|2m+1\rangle \langle 2m|}{\sqrt{2m+1}}. \]
where \( |n\rangle \) stands for the \( n\)th Fock state. The operator satisfies the identity
\[
\hat{\xi} \hat{a} + \hat{a} \hat{\xi} = 1.
\] (45)

We shall seek a solution of (42) in the form of power series of the operator \( \hat{\xi} \); since \( \hat{\xi}^n = 0 \) at any \( n \geq 2 \), we come to the relation \( \hat{f}(0) = X \hat{\xi} + Y \). Substituting this representation into equation (42) and equating the terms of the same order in \( \hat{a} \), we find coefficients \( X \) and \( Y \) and, thus, write the forward scattering amplitude operator explicitly:
\[
\hat{f}(0) = 2\langle \hat{a} \rangle \left[ f_1(0) - f_2(0) \right] \hat{\xi} + f_2(0).
\] (46)

The total electric field operator is expressed by (37) with the polarization operator defined by (41).

3. Quantum optics of an excitonic composite

3.1. Constitutive relations for the field operators

Consider a heterogeneous medium comprising a cubic lattice of identical spherical QDs embedded in a semiconducting host. The QD radius and distance between QDs are assumed to be much less than the wavelength in the host medium. The optical properties of such a composite medium are isotropic and can be described by an effective index of refraction. Let the optical density of the composite be small enough to neglect the electromagnetic coupling of QDs, i.e. to neglect the Mossotti–Clausius correction. Since the forward scattering amplitude turns out to be an operator, the optical response of the composite is described, instead of by (1), by the index of refraction
\[
\hat{n}^2 = 1 + \frac{4\pi \rho}{k^2} \hat{f}(0),
\] (47)

which is an operator in the space of quantum states of light. Obviously, the same is relevant to the relative permittivity \( \hat{\varepsilon} = \hat{n}^2 \). Utilizing the well-known relation \( \hat{D} = \hat{E} + 4\pi \hat{P} \) and (41), we come to the operator constitutive relation
\[
\hat{D} = \frac{1}{2} \left( \hat{\varepsilon} \hat{E} + \hat{E} \hat{\varepsilon} \right),
\] (48)

with the effective operator permittivity
\[
\hat{\varepsilon} = 2\langle \hat{a} \rangle \hat{\xi} n_1^2 + \left( 1 - 2\langle \hat{a} \rangle \hat{\xi} \right) n_2^2,
\] (49)

where \( n_{1,2}^2 = 1 + 4\pi \rho f_{1,2}(0) / k^2 \). The constitutive relations (48) and (49) satisfy the principle of correspondence: in the classical limit \( \hat{D} \rightarrow \langle \hat{D} \rangle \) we come to \( \langle \hat{D} \rangle = n_1^2 \langle \hat{E} \rangle \), i.e. the correct constitutive relation for an excitonic composite exposed to classical light [11, 12]. Correspondingly, in that case the forward scattering amplitude is a \( C \)-number and is expressed by (40). Even so, the local fields may result in physically observable effects, such as polarization-dependent splitting of the gain line [35, 36] in asymmetric QDs. The splitting was observed experimentally in [39, 40] and was referred to as a manifestation of the exchange interaction. Let us emphasize once more that here we deal with the same effect presented in phenomenological and microscopic languages. In the limit of zero local field, \( \Delta \omega \rightarrow 0 \) and,
obviously, \( n_1 \rightarrow n_2 \). As a result, equation (49) is reduced to \( \hat{\varepsilon} = n_2^2 \), giving \( \hat{D} = n_2^2 \hat{E} \). Thus, the constitutive relation for field operators takes the form usual for transparent media.

Because of (45), the operators \( \hat{\varepsilon} \) (49) and \( \hat{a} \) do not commutate and, consequently, the symmetrized representation of the constitutive relation (48) is of fundamental importance. Note that the operator \( \hat{\varepsilon} \) is non-Hermitian even in the absence of dissipation. We discuss its physical meaning later on. Here, we only stress that the non-hermitian nature does not result in any physical contradiction since the operator \( \hat{\varepsilon} \) does not represent any observable quantity. In contrast, the electric displacement operator \( \hat{D} \), related to its Fourier amplitude \( \hat{D} \) by a procedure analogous to equation (14), is Hermitian as it must be for observable quantities. The hermicity is due to the symmetrized notation on the right-hand side of equation (48).

3.2. Generalized plane waves

Consider the propagation in the excitonic composite of free (transverse) electromagnetic waves. The propagation is described by the 1D frequency-domain operator Maxwell equations

\[
i k \hat{H} = e_x \times \frac{\partial \hat{E}}{\partial z}, \quad -\frac{i k}{2} (\hat{\varepsilon} \hat{E} + \hat{E} \hat{\varepsilon}) = e_x \times \frac{\partial \hat{H}}{\partial z}. \tag{50}
\]

This system can easily be transformed to a set of wave equations for the operator \( \hat{E} - \langle \hat{E} \rangle \) and the mean value \( \langle \hat{E} \rangle \):

\[
\left( \frac{\partial^2}{\partial z^2} + k^2 n_1^2 \right) \langle \hat{E} \rangle = 0, \tag{51}
\]

\[
\left( \frac{\partial^2}{\partial z^2} + k^2 n_2^2 \right) (\hat{E} - \langle \hat{E} \rangle) = 0. \tag{52}
\]

The elementary solution of these equations leads to the expansions

\[
\hat{E} = i \sqrt{\frac{\hbar k}{A}} e_y \left[ \frac{e^{i k n_1 z}}{\sqrt{n_1}} \langle \hat{a} \rangle + \frac{e^{i k n_2 z}}{\sqrt{n_2}} (\hat{a} - \langle \hat{a} \rangle) \right], \tag{53}
\]

\[
\hat{H} = -i \sqrt{\frac{\hbar k}{A}} e_z \left[ e^{i k n_1 z} \sqrt{n_1} \langle \hat{a} \rangle + e^{i k n_2 z} \sqrt{n_2}(\hat{a} - \langle \hat{a} \rangle) \right], \tag{54}
\]

for the mode traveling in the direction \( z > 0 \). Equations (53) and (54) display the local field effect in the excitonic composite: decomposition of a single mode into coherent and incoherent parts with different refractive indices. Thus, as quantum light propagates through the excitonic composite, the light statistics is altered and spatial modulation of the amplitude with the period

\[
L_0 = \frac{2\pi}{n_1 - n_2 |k|} \tag{55}
\]
occurs. The period is strongly dependent on frequency due to the frequency dependence of partial refractive indices \( n_{1,2} \) determined by equation (43). In turn, the alteration leads to transformation of the light quantum coherence of the second and higher degrees. To some extent, the situation is similar to that in nuclear optics [30]: the existence of two spin-dependent refractive indices of the particle’s wavefunction causes the spin rotation as the particle travels through a polarized medium.
Physically, the result obtained can be understood as follows. There are two mechanisms of quantum light scattering by a QD. The first one—quasi-classical—is due to the induction in the QD of the observable polarization and, therefore, it persists in the classical limit. The corresponding frequency turns out to be shifted by the value $\Delta \omega$. This mechanism changes the QD state due to the local-field-induced electron–hole correlations and thus provides an inelastic channel of the scattering. The first term on the right-hand side of (53) conforms to this channel.

The aforementioned non-hermicity of the operator $\hat{\varepsilon}$ even in the absence of dissipation is due to inelastic scattering. The second mechanism manifests itself in the scattering of states of light having zero observable field (e.g. a single Fock state). Such states do not induce observable polarization and, consequently, any frequency shift. This mechanism has no classical analog and provides the elastic channel of the scattering. The presence of the inelastic scattering channel signifies the change of the scattered light’s quantum state, i.e. the photon-statistics dispersion. Thus, the photon-statistics dispersion and the resulting phenomenon of the electromagnetic field spatial modulation are the direct manifestations of the fine structure of the excitonic line, inherent in an isolated QD [21].

Going to the time-domain representation of operators (53) and (54), $\hat{E}(r, t)$ and $\hat{H}(r, t)$, by the procedure defined by equation (14), we can show that the condition

$$\int_A \langle \hat{E}(r, t) \times \hat{H}(r, t) \rangle e_z \, dx \, dy = \text{const.} \quad (56)$$

holds true for an arbitrary quantum state. Physically, this condition reflects the momentum conservation for fields determined by operators (53) and (54): a mean value of the energy flux through an arbitrary closed surface is equal to zero for an arbitrary quantum state of light. That is, dissipation is absent in the medium in spite of the non-hermicity of the relative permittivity operator $\hat{\varepsilon}$. The conservation law (56) testifies to the physical correctness of electromagnetic waves defined by the operators (53) and (54).

### 3.3. Input–output relations

Consider a planar layer of the QD-based composite material of thickness $d$ located at $|z| < d/2$ exposed to a quantum field. The problem to be solved is to ascertain relations between operators of incoming, reflected and transmitted waves. For that, we make use of equations (53) and (54) and utilize the layer’s reflection $R$ and transmission $T$ amplitudes (see e.g. equations (6.130)–(6.132) in [27]). The reflection and transmission amplitudes satisfy the property $|R|^2 + |T|^2 = 1$, which is a reflection of the scattering matrix’s unitarity. Obviously, the total field outside the layer can be represented by [27]

$$\hat{E} = \hat{E}_+^{\text{in}} + \hat{E}_-^{\text{out}} + \hat{E}_+^{\text{in}} + \hat{E}_-^{\text{out}}, \quad (57)$$

where $\hat{E}_\pm^{\text{in}}$ are the operators of incoming waves propagating along the positive (index ‘+’) and negative (index ‘−’) directions of the $z$-axis. The superscript ‘out’ marks outgoing waves. By analogy with (15), the incoming waves are given by

$$\hat{E}_\pm^{\text{in}} = i \sqrt{\frac{\hbar k}{A}} e_y \hat{a}_\pm e^{\pm ikz}, \quad (58)$$

where creation/annihilation operators $\hat{a}_\pm/\hat{a}_\pm$ satisfy ordinary bosonic commutation relations.
In view of the commutativity of operators on the right-hand sides of equations (53) and (54), the layer’s transmission/reflection balance can be written separately for each of the waves \( \exp(ikn_{1,2}z) \):

\[
\begin{align*}
\langle \hat{a}^\text{out}_\pm \rangle &= T_1 \langle \hat{a}_\pm \rangle + R_1 \langle \hat{a}_\mp \rangle, \\
\hat{a}^\text{out}_\pm - \langle \hat{a}^\text{out}_\pm \rangle &= T_2 (\hat{a}_\pm - \langle \hat{a}_\pm \rangle) + R_2 (\hat{a}_\mp - \langle \hat{a}_\mp \rangle).
\end{align*}
\]

Here \( T_{1,2} \) and \( R_{1,2} \) are the transmission and reflection coefficients of layers of the thickness \( d \) and refractive indices \( n_1 \) and \( n_2 \), respectively. Using (59) and (60), we can represent outgoing fields as

\[(\hat{E}^\text{out})^2 \sim |T_2|^2 \langle \hat{a}_\pm \rangle^2 + (|T_1|^2 - |T_2|^2) |\langle \hat{a}_\pm \rangle|^2.
\]

It can easily be shown that, in view of \(|R_{1,2}|^2 + |T_{1,2}|^2 = 1\), the outgoing electromagnetic field satisfies the correct commutation relations for field operators and the energy–momentum balance condition for incoming and outgoing waves for light in the arbitrary quantum state.

Equation (61) shows that the quantum light transmission through a layer of excitonic composite leads to the alteration of light coherence. For example, using the obvious identity

\[
\langle (\hat{a} - \langle \hat{a} \rangle)(\hat{a} - \langle \hat{a} \rangle) \rangle = \langle \hat{a}_\pm \hat{a}_\mp \rangle - |\langle \hat{a}_\pm \rangle|^2,
\]

we derive

\[(\langle \hat{E}^\text{out}_+ \rangle^2) \sim |T_2|^2 \langle \hat{a}_\pm \rangle^2 + (|T_1|^2 - |T_2|^2) |\langle \hat{a}_\pm \rangle|^2.
\]

The photocurrent measured by a detector located in the region \( z > d/2 \) is proportional to \( \langle |\hat{E}^\text{out}_+ |^2 \rangle \). Thus, the photodetector signal is no longer completely determined by the mean number of photons (first term in (62)) but depends also on the light statistics (second term in (62)). This effect can be exploited for experimental verification of theoretical predictions of the present paper. The distinction of the reflection/transmission properties of the excitonic composite layer for coherent and non-coherent components can be used for the design of new quantum-optical light transformers.

4. Photonic-state dispersion

The complete characterization of a quantum state of light is given by the density operator. In this section, we evaluate this operator in an excitonic composite medium. Along with that, the issues of interest are the light characteristics measurable in typical photodetection processes. As an example of such characteristics, further we consider the normally ordered normalized time-zero second-order correlation function \( g^{(2)}(z) \) of the excitonic composite, which characterizes the quantum-mechanical second-order degree of coherence at a given cross-section \( z \) [1].

4.1. Density operator

Let us rewrite equation (53) as

\[
\hat{E} = i \sqrt{\frac{\hbar}{A n_2}} e^{i kn_2 z} \hat{a}(z) e_y,
\]

where \( \hat{a}(z) = \hat{a} + \kappa(z) \) and

\[
\kappa(z) = \langle \hat{a} \rangle \left[ \sqrt{\frac{n_2}{n_1}} e^{i (n_1 - n_2)kz} - 1 \right].
\]
The quantity $\kappa(z)$ is a periodic function of the spatial coordinate $z$ with oscillation period $L_0$ (55). Operators $\hat{a}(z)$ and $\hat{a}^\dag(z)$ satisfy the ordinary bosonic commutation relations. That is, they can be treated as annihilation and creation operators of the quanta of some bosonic field. Utilizing then operator identities (3.47) and (3.48) from [27], we come to

$$\hat{a} = \hat{D}^\dagger_\kappa \hat{a} \hat{D}_\kappa,$$

(65)

where

$$\hat{D}_\kappa = \exp(\kappa \hat{a}^\dagger - \kappa^* \hat{a})$$

is the displacement operator with $\kappa$ as the displacement factor. Hereafter we omit the $z$-dependence. According to (65), as light passes through the excitonic composite the spatial evolution of the light density operator is governed by the unitary transform

$$\hat{\varrho}' = \hat{D}^\dagger_\kappa \hat{\varrho} \hat{D}_\kappa,$$

(67)

where $\hat{\varrho}$ is the initial operator of the light density induced by a source at the input. The distinguishing property of the transform (67) is that it actually is not linear: the displacement factor $\kappa$ depends on the state of the initial quantum light through the relation

$$\kappa \sim \langle \hat{a} \rangle = \sum_{n=0}^{\infty} \sqrt{n+1} \varrho_{n+1,n}.$$

(68)

Physically, this means that the field acting on the QD-exciton is self-consistent in the Hartree–Fock–Bogoliubov sense (see equation (22)). As a result of self-consistency, nonlinearity of the quantum-mechanical motion of carriers in QD arises; see the second term on the right-hand side of equation (28).

The operator transformation law (67) can be rewritten in terms of familiar matrix elements. As the first step we represent $\hat{\varrho}'$ by the expansion

$$\hat{\varrho}' = \sum_{m,n} \varrho_{mn} \hat{D}_{-\kappa} |m\rangle \langle n| \hat{D}^\dagger_{-\kappa}.$$

(69)

In accordance with [27],

$$\hat{D}_{-\kappa} |m\rangle = \exp\left(\frac{-|\kappa|^2}{2}\right) \sum_p S_{mp}(|\kappa|^2) |p\rangle,$$

(70)

where

$$S_{mp}(\kappa) = \begin{cases} (\kappa^*)^{m-p} \frac{p!}{m!} L^{(m-p)}(|\kappa|^2), & m \geq p, \\ (-\kappa)^{p-m} \frac{p!}{m!} L^{(p-m)}(|\kappa|^2), & m < p, \end{cases}$$

(71)

and $L^{(n)}_{m}(x)$ are the generalized Laguerre polynomials. Then, using (69)–(71) we can proceed from (67) to the law of transformation of matrix elements:

$$\varrho_{pq}' = \exp\left(\frac{-|\kappa|^2}{2}\right) \sum_{m,n} \varrho_{mn} S_{mp}(\kappa) S^*_{nq}(\kappa).$$

(72)
Equations (67) and (72) determine the transformation law for quantum light with arbitrary statistics. Consider several particular cases as examples. For a single Fock state \(|n\rangle\), \(|\hat{a}\rangle = 0\) and \(\kappa = 0\). Taking \(\hat{D}_0 = 1\) into account we conclude that the excitonic composite does not transform single Fock states. An analogous situation occurs for thermal light where \(\varrho_{nn} \sim \delta_{mn}\) and, accordingly (68), \(|\hat{a}\rangle = 0\). The spatial evolution of a coherent state \(|\beta\rangle\) is characterized by the transform \(q' = \hat{D}^\dagger_\kappa |\beta\rangle |\beta\rangle = \hat{D}_{-\kappa} \hat{D}_{-\beta} |0\rangle |0\rangle \langle 0| \hat{D}_{-\beta}^\dagger \hat{D}_{-\kappa}^\dagger\). Taking into account the well-known identity [27] \(\hat{D}_{-\kappa} \hat{D}_{-\beta} = \hat{D}_{-\beta - \kappa} \exp[i\text{Im}(\kappa \beta^*)]\), we come to

\[
q' = |\kappa + \beta\rangle \langle \kappa + \beta| = \left| \sqrt{\frac{n_2}{n_1}} \beta e^{i(n_1 - n_2)kz} \right| \left| \sqrt{\frac{n_2}{n_1}} \beta e^{i(n_1 - n_2)kz} \right|. \tag{73}
\]

This result implies that the initial coherent state is transformed into another coherent state with modified coherent amplitude. The coherent amplitude of the transformed state experiences spatial beating as light passes through the composite (factor \(\exp[i(n_1 - n_2)kz]\) in (73)). The mean photon number in the composite \(|n_2\beta^2/n_1\rangle\) is invariable and significantly differs from that the source produces in vacuum, \(|\beta|^2\). The alteration is determined by the ratio \(|n_2/n_1|\), which varies in a broad range depending on the photon energy. There are different energy bands in the excitonic composite, where the classical source efficiency either rises (\(|n_2| > |n_1|\)) or falls (\(|n_2| < |n_1|\)), in comparison with the source efficiency in vacuum.

As one more example, consider the transformation of the Fock qubit. Let \(|\Psi\rangle = \beta_0 |0\rangle + \beta_1 |1\rangle\), where the coefficients \(\beta_0, \beta_1\) are coupled by the ordinary normalization \(|\beta_0|^2 + |\beta_1|^2 = 1\). According to (67) and (68), the transformation is given by

\[
q' = |\Psi'\rangle \langle \Psi'\rangle = \hat{D}_{-\kappa} |\Psi\rangle \langle \Psi| \hat{D}_{-\kappa}^\dagger, \tag{74}
\]

with \(|\hat{a}\rangle = \beta_0^* \beta_1\) in (64) for the displacement factor \(\kappa\). Obviously, in the coherent-state basis \(|\Psi'\rangle = (\beta_0 \hat{D}_{-\kappa} + \beta_1 \hat{D}_{-\kappa} \hat{a}^\dagger) |0\rangle\). Taking then into account the relation \([\hat{a}^\dagger, \hat{D}_{-\kappa}] = -\partial \hat{D}_{-\kappa} / \partial \hat{a} = -\kappa^* \hat{D}_{-\kappa}\) (see equation (C.17) of [27]), we obtain \(|\Psi'\rangle = (\beta_0 \beta_1 \kappa^* + \beta_1 \hat{a}^\dagger)|\kappa\rangle\). Then, proceeding to the Fock-state representation, we come to the expression

\[
|\Psi'\rangle = \exp \left( -\frac{|\kappa|^2}{2} \right) \sum_{n=0}^{\infty} \frac{\kappa^n}{\sqrt{n!}} \left( \beta_0 + \beta_1 \kappa^* + \beta_1 \hat{a}^\dagger \right) |n\rangle. \tag{75}
\]

Equations (74) and (75) define the spatial evolution of the Fock qubit in an excitonic composite. Let us stress that (75) includes a full set of Fock states. An essential physical conclusion can be deduced from this: new states absent in light in the beginning arise as light travels through the excitonic composite.

### 4.2. Second-order coherence

To illustrate the effect of the photon statistics dispersion, we calculate the second-order correlation function \(g^{(2)}(z)\), which is represented in terms of the field operators by [1]

\[
g^{(2)}(z) = \langle \hat{E}^\dagger \hat{E}^\dagger \hat{E} \hat{E} \rangle / \langle \hat{E}^\dagger \hat{E} \rangle^2.
\]
Let us consider light prepared in an arbitrary quantum state $|\Psi\rangle$. Then, after some algebraic manipulations, we obtain the second-order correlation function for the field operator (53)

$$ g^{(2)}(z) = \frac{\sum_n |\kappa(z)A_{n-1} + 2\kappa(z)\sqrt{n}A_n + \sqrt{n(n+1)}A_{n+1}|^2}{\left[ \sum_n n|A_n|^2 + (n_2/n_1 - 1)|\langle \hat{\alpha} \rangle|^2 \right]^2}, \quad (76) $$

where $A_n = \langle n|\Psi\rangle$. Without taking local fields into account, the correlation function $g^{(2)}(z)$ coincides with the correlation function $g_0^{(2)} = \sum_n n(n+1)|A_{n+1}|^2/(\sum_n n|A_n|^2)^2$ of the incident field $E_0$. The distinguishing property of $g^{(2)}(z)$ is its frequency dependence, which is due to the pronounced frequency dispersion $n_{1,2} = n_{1,2}(\omega)$ of the excitonic composite. Also, the second-order correlation function experiences the spatial modulation with a period $L_0$.

The most intriguing peculiarity in the photon statistics appears in the vicinity of the resonance $\omega = \omega_2$. For the light states with a large enough $|\langle \hat{\alpha} \rangle|$, the inequality $n_2|\langle \hat{\alpha} \rangle|^2 \gg 1$ holds true because of the growth of $n_2$ in the vicinity of the resonance. As follows from (76), in that case the spatial modulation becomes a high-order infinitesimal and $g^{(2)}(z) \rightarrow 1$ independently of the light state $|\Psi\rangle$; that is, the photon statistics tends to Poissonian (completely uncorrelated photons) as light propagates through the excitonic composite. This means that the excitonic composite reduces the photon correlation degree for photons with both positive ($g_0^{(2)} > 1$) and negative ($g_0^{(2)} < 1$) correlation. Indeed, $g^{(2)}(z) < g_0^{(2)}$ in the former case, whereas $g^{(2)}(z) > g_0^{(2)}$ in the last one. Another situation arises for a pure Fock state $|n\rangle$ with $|\langle \hat{\alpha} \rangle| = 0$. In this case, equation (76) gives $g^{(2)} = 1 - 1/n$. That is, the correlation function (76) coincides with the free-space correlation function. In general, negligible changing of the correlation function takes place for all quasi-Fock states characterized by the condition $n_2|\langle \hat{\alpha} \rangle|^2 \ll 1$.

A spatial modulation of the second-order correlation function of light is characteristic for the Hanbury–Brown–Twiss (HBT) interference [1] of quantum light emitted by two sources. As different from the HBT effect, the spatial modulation $g^{(2)}(z)$ (76) in the excitonic composite arises as a result of the interference of coherent and incoherent components of the field. The transformation of the statistics of quantum light interacting with a finite-thickness dielectric slab with a $C$-number relative permittivity was considered in [27]. The statistics transformation is provided by the light reflection/refraction at boundaries, similar to the fact that a dispersionless dielectric slab modifies the frequency spectrum of a signal due to the interference of waves reflected from boundaries. Another effect of such a type has been predicted in [41]: appearance of spatial quantum correlation in light propagating through a multiple scattering random medium. The quantum correlations depend on the state of quantum light and are due to the cross-correlation between different output modes. In our case, the physical mechanism of the transformation is completely different: we predict the transformation effect to be inherent to a homogeneous medium. That allows us to identify the effect as a new type of optical dispersion.

It should be noted that the notions ‘linear regime’ and ‘weak coupling regime’ as applied to excitonic composite media are not identical. In the presence of a local field, the polarization (36) is linear in the incident field (linear regime) but contains a quadratic term in the oscillator strength, i.e. the term $O(|\mu|^4)$. Nonlinearity of that type violates the weak coupling regime and leads to the transformation of the photon statistics similar to what takes place in the standard Jaynes–Gummings dynamics [1]. In other words, in the presence of a local field, the light–QD interaction is characterized by two coupling parameters: the standard Rabi frequency and the new depolarization shift $\Delta \omega$.
5. Summary and outlook

In the present paper, we have presented a theory of the quantum light interaction with excitonic composites whose relative permittivity is predicted to be an operator in the space of quantum states of light. As a result, the medium’s refractive index turns out to be dependent on the photon number distribution providing, by analogy with the frequency dispersion and the spatial dispersion, the photon statistics dispersion of light. Self-organized lattices of ordered QD-molecules and 1D-ordered (In,Ga)As QD-arrays [42, 43] serve as examples of high-quality excitonic composites.

Light transformation in the excitonic composite is carried out by means of the displacement operator $\hat{D}_\kappa$—one of the canonical transforms of quantum optics [1]. However, differently from ordinary cases, the displacement factor $\kappa$ in an excitonic composite depends not only on the photon energy and momentum (in media with spatial dispersion) but also on the photon statistics. In particular, an excitonic composite provides different propagation regimes for coherent and non-coherent components of the total field. That is, the frequency response characteristics of excitonic composite-based structures may differ in coherent and non-coherent light. For example, a distributed Bragg reflector with layered components made of excitonic composite materials would manifest shifted reflection bands for coherent and non-coherent light. In the same manner, an excitonic composite-based microcavity will possess, depending on the light statistics, two alternate sets of eigenfrequencies. An interferometer consisting of two collinear plane mirrors of an excitonic composite material can serve as the simplest model. The excitonic composite provides a new (not related to anisotropy) birefringence mechanism: coherent and non-coherent components of a plane wave obliquely incident on the plane excitonic composite interface will propagate at different angles. Equations (59) and (60) show that an excitonic composite layer can serve as a beam splitter with a modified law of transformation of the field operators. Along with the second-order coherence transformation, the optical effects mentioned create the potential for experimental verification of the predicted photon-statistics dispersion.

The theory elaborated in the present paper deals with the single-mode case that implies the single pair of creation/annihilation operators sufficient to characterize the fields (53) and (54). In the multi-mode case, each mode will be characterized by its own mode-number-dependent value $\langle \hat{a} \rangle$ and, correspondingly, permittivity operator. As a result, then the photon-statistics dispersion will be accompanied by spatial dispersion. In classical crystal optics [44], spatial dispersion necessarily entails optical anisotropy even in structurally isotropic media. The preferential direction is determined by the propagating wavevector and refractive indexes are different for transverse and longitudinal field components. In an isotropic medium with the multi-mode photon-statistics dispersion, we meet another situation: in spite of the spatial non-locality the permittivity operator is scalar. The situation can be understood from the following. A preferential direction is absent in an excitonic composite since it can be related only to a physically observable wave. In contrast, in the case considered, the spatial dispersion mechanism concerns operators and disappears on proceeding to observable fields.

The physical picture of the photon-statistics dispersion can be essentially modified by the influence of multi-level effects. As was discussed in section 2.1, the influence manifests itself in exciton–exciton correlations which lead to quantum fluctuations of the depolarization field. Obviously, accounting for the multi-level effects modifies Hamiltonian (22) and,
correspondingly, equation (25). The incorporation into analysis of two-particle correlations is an original problem that could be the subject of an independent study.

The excitonic composite considered in this paper is an example that demonstrates the existence of media characterized by the operator constitutive relation (48) and, consequently, displaying the photon-statistics dispersion (49). It would be of interest to search for other artificial (and, possibly, natural) materials with photon-statistics dispersion and statistics transformation law not necessarily defined by (49) and (67), respectively. Since the photonic-state dispersion in excitonic composites is due to dipole–dipole interactions in QDs, one can expect dispersion of that type in media with strong dipole–dipole interactions, such as Bose–Einstein condensates [26] and ultracold atomic ensembles [45].

The developed model of quantum–optical properties of excitonic composites leads to a set of functional equations, in which both quantum fields to be found and predetermined coefficients are operators in the same space of quantum states. The investigation of general properties and methods of solution of such equations constitutes an intriguing mathematical problem. One can expect that the equations of that type will find application in studying quantum fields of different physical origin. A matter of special interest would be the case when the equation solution and the coefficients are non-commutative—as happens in our paper.

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Appendix. Derivation of equation (13)

The solution of equation (12) is represented by

\[ \langle \tilde{c}_{k\lambda}(t) \rangle = \beta_{k\lambda} e^{-i\omega_{k} t} + \frac{2\pi \omega_{k}}{\hbar \Omega} e_{k\lambda} \int_{-\infty}^{t} e^{-i\omega_{k}(t-\tau)} \int_{V} \langle \hat{P}(r, \tau) \rangle e^{-i\mathbf{k} \cdot \mathbf{r}} \, d^{3}r \, d\tau, \quad (A.1) \]

where \( \beta_{k\lambda} = \langle \tilde{c}_{k\lambda}(\infty) \rangle \) are arbitrary coefficients satisfying the continuity at the QD boundary of the expansion (11) for the quantity \( \langle \hat{E}_{L}(r, -\infty) \rangle \). In view of (A.1), equation (9) is written as

\[ \hat{U}_{1} = -i \sum_{k\lambda} \sqrt{\frac{2\pi \hbar \omega_{k}}{\Omega}} e_{k\lambda} \beta_{k\lambda} \int_{V} \hat{P}(r) e^{i\mathbf{k} \cdot \mathbf{r}} \, d^{3}r - i \sum_{k} \frac{2\pi \omega_{k}}{\Omega} \int_{-\infty}^{t} \int_{V} \hat{P}_{\alpha}(r) \langle \hat{P}_{\beta}(r', \tau) \rangle \sum_{\lambda} e_{k\lambda}^{(\alpha)} \times e_{k\lambda}^{(\beta)} e^{i\mathbf{k} \cdot \mathbf{r}' - i\omega_{k}(t-\tau)} \, d^{3}r \, d^{3}r' \, d\tau \right) + \text{H.c.}, \quad (A.2) \]

where indices \( \alpha \) and \( \beta \) denote projections of corresponding vectors in a Cartesian basis.

The first term in (A.2) can be transformed in the following way. Let us assume that \( \beta_{k\lambda} \neq 0 \) only for modes with \( \mathbf{k} \mathbf{e}_{x} = \mathbf{k} \mathbf{e}_{y} = 0 \) and \( \lambda \rightarrow y \) (i.e. \( \mathbf{e}_{k\lambda} \rightarrow \mathbf{e}_{y} \)). Next we assume \( \Omega = AL \) and carry out the standard limit transition \( L \rightarrow \infty \). Details of the procedure can be found, e.g. in [27]. Letting \( \langle \hat{a}_{\omega} \rangle = \lim_{L \rightarrow \infty} (\beta_{k\lambda} \sqrt{L} / 2\pi \epsilon) \), we obtain for the first term in (A.2) formula (9) with the replacement \( \hat{E}_{L} \rightarrow \hat{E}_{0} \), where \( \hat{E}_{0} \) is determined by equations (14) and (15).
For transformation of the second term in equation (A.2) we make use of the identity \[ \sum_\lambda \lambda e^{(\alpha)}_{\lambda} e^{(\beta)}_{\lambda} = \delta_{\alpha\beta} - \frac{k_\alpha k_\beta}{k^2} = \frac{1}{k^2} \left( \frac{\partial^2}{\partial x_\alpha \partial x_\beta} - \delta_{\alpha\beta} \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right), \]

and perform the limit transition \( \Omega \to \infty \), which corresponds to the substitution

\[ \sum_k \rightarrow \frac{\Omega}{(2\pi)^3} \int \cdot \, d^3k. \]

Then, the equation

\[ \hat{U}_1 = -\int_V \hat{P}(r) \langle \hat{E}_0(r,t) \rangle \, d^3r - 4\pi \int_0^\infty \int_V \int_V \left( \frac{\partial^2}{\partial x_\alpha \partial x_\beta} - \delta_{\alpha\beta} \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right) G^{(0)}(r-r',t') \]

\[ \times \hat{P}_\alpha(r)\langle \hat{P}_\beta(r',t-t') \rangle \, dt' \, d^3r \, d^3r' \]  

(A.3)

follows from (A.2) with

\[ G^{(0)}(r,t) = \frac{i e^2}{2(2\pi)^3} \int \frac{e^{ikr}}{\omega_k} \left( e^{-i\omega t} - e^{i\omega t} \right) \, d^3k \]

\[ = \frac{1}{4\pi |r|} \left[ \delta \left( \frac{|r|}{c} - t \right) - \delta \left( \frac{|r|}{c} + t \right) \right] \]  

(A.4)

as the free-space Green function [38]. As the next step we introduce the quasi-static approximation, which implies the limit transition \( c \to \infty \) in (A.3) and (A.4). Integration in (A.3) over \( t' \) leads us to

\[ \hat{U}_1 = -\int_V \hat{P}(r) \langle \hat{E}_0(r,t) \rangle \, d^3r - \int_V \int_V \hat{P}(r)\hat{G}(r-r')\langle \hat{P}(r',t) \rangle \, d^3r \, d^3r'. \]  

(A.5)

In view of equation (21), which determines the polarizability in the two-level approximation, and the rotating-wave approximation, we see that (A.5) is identical to equation (13).

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