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The bipolariton model and the concept of excitonic–biexcitonic mixed modes in semiconductors

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Abstract. An alternative approach to the so called bipolariton concept has been proposed. We have shown that the bipolariton approximation can indeed be used for calculating the elementary excitation spectrum in the photon–exciton–biexciton system in its biexciton part. The bipolariton concept does not provide the correct spectrum in the excitonic-polariton energy part, where the existence of four-particle electron–hole bound states should be considered. In order to go beyond the bipolariton approximation we developed a Green function formalism for the theory of an interacting electron–photon system in which photons, excitons and biexcitons are treated on an equal level. Our approach is a completely microscopic treatment and is based on the assumption that the excitons and biexcitons are two- and four-particle bound states, formed by electrons and holes. The method of Legendre transforms is used to derive a set of nine independent exact equations for the corresponding vertex and Green functions. Knowledge of the solutions of the above equations provide the excitation energies of the quasiparticles (excitonic–biexcitonic polaritons) formed by coupling of photons with excitons and biexcitons. Those elementary excitations manifest themselves as common poles of the photon propagator, two- and four-particle electron–hole Green functions.

1. Introduction

It is a well known fact that the interaction of light with the polarization of a crystal leads to the coupling of the photons with elementary excitations of the matter into new quasiparticles. The quanta of those mixed states are called polaritons. The polariton theory of light propagation in semiconductors has received much attention, because the theory is needed for the interpretation of optical spectra of crystals. In what follows we will consider the interaction of light with the polarization in semiconductors, considering the ions fixed at their equivalent positions, i.e. we assume the polarization is caused by the collective electronic excitations. In this case the system of interest consists of a radiation field and a material system. Since, we consider the ions fixed at their equilibrium positions, the material system is made up of electrons in a periodical lattice potential. The radiation and the matter interact via an electron–photon interaction. Although in an ideal crystallized semiconductor excitons and biexcitons are the energetically lowest electronic excitations, the biexciton contribution to the polariton formation and vice versa, the polariton contribution to the biexciton states, have been considered in a small number of articles. Most of them are concerned with the case when the semiconductor has been driven to a non-equilibrium state by an intensive coherent pump wave. If the pump wave is treated classically by replacing

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the exciton and photon pump operators by complex numbers, a strongly non-equilibrium excitonic–biexcitonic polariton eigenmode which follows dynamically the pump intensity can be introduced (Ivanov *et al* 1991). In the opposite case, i.e. without an intensive pump wave, there exist equilibrium thermodynamic states which can be described by using the equilibrium Green function technique. In this case the system of interest can be theoretically investigated by using two different methods: a phenomenological approach and a direct microscopic treatment. According to the first one, the initial electron–photon system is replaced by the system of interacting excitons and photons. In this approach the excitons are boson excitations with a dispersion depending on the number of phenomenological parameters, while the biexcitons are considered as compound particles formed by coupling of two excitons due to the exciton–exciton attractive potential. The phenomenological approach is based on the procedure of replacing the fermion creation and annihilation operators for one-electron states by Bose creation and annihilation operators for exciton states. This procedure is correct only in the limit $N a_x^3 \ll 1$, where N is the concentration of excitons excited by light and a_x is the exciton Bohr radius. But, as is well known, at low temperatures the system of Bose particles with attractive interaction between the boson particles (excitons) is unstable against spontaneous contraction. For this reason one has to introduce two equivalent types of exciton with the same parameters, both with total spin zero. The only distinction is that the excitons of the different types attract each other whereas excitons of the same type have a repulsive interaction. This assumption leads simultaneously to the existence of biexcitons as bound complexes of two excitons of different types and the stability of the many-particle (boson) system against spontaneous contraction. The two types of exciton may be considered as corresponding to singlet excitons with mutual opposite directions of electron (hole) spins, while in real systems triplet excitons also exist. According to the phenomenological approach, the initial electron–photon system can be replaced by the exciton–photon system, described by the following Hamiltonian (Ivanov *et al* 1991):

$$H_M = \sum_{\sigma,p} \left\{ \omega_p^x B_{\sigma,p}^+ B_{\sigma,p} + \omega_p^y \alpha_{\sigma,p}^+ \alpha_{\sigma,p} + \frac{i\Omega_c}{2} (\alpha_{\sigma,p}^+ B_{\sigma,p} - \alpha_{\sigma,p} B_{\sigma,p}^+) \right\} \\ + \frac{1}{2} \sum_{\sigma,\sigma',p,k,q} W_{\sigma\sigma'}(\mathbf{q}) B_{\sigma,p}^+ B_{\sigma',k}^+ B_{\sigma',k+q} B_{\sigma,p-q}. \quad (1a)$$

Here $\alpha_{\sigma,p}$ and $B_{\sigma,p}$ are the photon and exciton Bose operators, respectively. The symbols $\sigma, \sigma' = 1, 2$ refer to the above-mentioned two types of exciton and to the two possible circular polarizations of the photons; ω_p^x and ω_p^y are the exciton and photon dispersions, respectively. The polariton parameter Ω_c is defined in terms of the longitudinal–transverse splitting Δ_{LT} : $\Omega_c = \sqrt{2\omega_t \Delta_{LT}}$, where $\omega_t = \omega_{p=0}^x$. The explicit form of the potential $W_{\sigma\sigma'}(\mathbf{q})$ is given in the paper by Sheboul and Ekardt (1976). This potential is repulsive for two excitons with $\sigma = \sigma'$ and attractive for the excitons with $\sigma \neq \sigma'$, which leads to biexciton formation. The problem of the energy spectrum of the system, described by Hamiltonian (1a), has been solved by Ivanov and Haug (1993, 1995a, b). The authors have proposed and analysed the so-called bipolariton model. According to the bipolariton concept the excitonic–biexcitonic polariton eigenmodes do not exist. The elementary excitation spectrum consists of two types of quasiparticle: two excitonic-polariton branches $\omega_\mu(\mathbf{Q})$ (the upper ($\mu = +$) and the lower ($\mu = -$))

$$\omega_\mu(\mathbf{Q}) = \omega_\pm(\mathbf{Q}) = \frac{1}{2} [(\omega_Q^x + \omega_Q^y) \pm \sqrt{(\omega_Q^x - \omega_Q^y)^2 + \Omega_c^2}] \quad (1b)$$

and the renormalized biexcitons (bipolaritons) $\tilde{\Omega}_m(\mathbf{Q})$ (m is the complete quantum number). The bipolariton ground state energy $\tilde{\Omega}_0(\mathbf{Q})$ has a complex value:

$$\tilde{\Omega}_0(\mathbf{Q}) = \Omega_0(\mathbf{Q}) + \Delta_0(\mathbf{Q}) - i \frac{\Gamma_0(\mathbf{Q})}{2} \tag{1c}$$

where $\Omega_0(\mathbf{Q})$ is the biexciton ground state energy, $\Delta_0(\mathbf{Q})$ is the radiative renormalization of the biexciton ground state (the biexciton Lamb shift) and $\Gamma_0(\mathbf{Q})$ is the inverse biexciton radiative lifetime. The two polariton branches (1b) manifest themselves as common poles of photon and exciton Green functions, while the bipolaritons are the poles of the two-exciton Green function.

The other possible way to treat the system of interest is to use the direct microscopic approach, which is based on the assumption that the excitons and biexcitons are two- (one electron and one hole) and four-particle (two electrons and two holes) bound states, respectively. In view of the fact that in most of the papers related to our problem excitons are considered without internal structure, there may be a need to clarify the motivations for using the direct microscopic treatment. Firstly, this method does not make use of the complicated and approximate procedure of replacing the fermion operators for one-electron states by Bose operators for exciton states. Secondly, the functional technique combined with the method of the Legendre transforms can be successfully applied, because the corresponding action does not depend on the product of four field operators, as in the case of the Hamiltonian (1a). Thus, the many-body aspect of the bound state can be dealt with more systematically, so all possible correlation and screening effects are included in principle. Thirdly, this approach provides a simultaneous treatment of the exchange interaction, local-field corrections and band degeneracy. In the direct microscopic treatment one has to investigate the system which consists of a radiation field, described by the action $S_0^{(\omega)}$ and a material system. In our case the material system is the semiconductor, which can be described by the action for non-interacting electrons in a periodical lattice potential $S_0^{(e)}$. The radiation and the matter interact via an electron-photon interaction, described by the action $S^{(e-\omega)}$. In terms of the field theory we deal with a boson (photon) field $A_\alpha(z)$ interacting with a fermion field $\bar{\Psi}(y)$ (or $\Psi(x)$) at finite temperatures. Here $z = \{\rho, \nu\}$, $y = \{r, \sigma, u\}$ and $x = \{r', \sigma', u'\}$ are composite variables, where r, r', ρ are radius vectors and σ, σ' are spin indices. According to the finite-temperature field theory (Matsubara 1955), the variables u, u', ν range from 0 to $\beta = (k_B T)^{-1}$, where T is the temperature, k_B being the Boltzmann constant. The action of the system which takes into account the so-called ‘local-field’ effect has the following form (Koinov and Glinskii 1988):

$$S_1 = S_0^{(e)} + S_0^{(\omega)} + S^{(e-\omega)} + S^{(e-e)} \tag{2a}$$

where

$$S_0^{(e)} = \bar{\Psi}(y) G^{(0)-1}(y, x) \Psi(x) \tag{2b}$$

$$S_0^{(\omega)} = \frac{1}{2} A_\alpha(z) D_{\alpha\beta}^{(0)-1}(z, z') A_\beta(z') \tag{2c}$$

$$S^{(e-\omega)} = \bar{\Psi}(y) \Gamma_\alpha^{(0)}(y, x|z) \Psi(x) A_\alpha(z) \tag{2d}$$

$$S^{(e-e)} = -\frac{1}{2} \bar{\Psi}(y) \Psi(x) \tilde{\Gamma}_\alpha^{(0)}(y, x|z) \tilde{D}_{\alpha\beta}^{(0)}(z, z') \tilde{\Gamma}_\beta^{(0)}(y', x'|z') \tilde{\Psi}(y') \Psi(x'). \tag{2e}$$

Here $G^{(0)-1}(y, x)$ is the inverse one-particle Green function for the system of non-interacting electrons in a periodical lattice potential. $D_{\alpha\beta}^{(0)-1}(z, z')$ and $\Gamma_\alpha^{(0)}$ are the inverse free photon propagator (in a gauge, when the scalar potential is equal to zero), and the ‘bare’ electron-photon vertex, respectively.

We notice that in the above equations there are quantities, such as $A_\alpha(z)$, $D_{\alpha\beta}^{(0)}(z, z')$, $\Gamma_\alpha^{(0)}(y, x|z)$, which depend on the photon variables z, z' . Their Fourier transforms contain

only components with wavevectors within the Brillouin zone. The Fourier transforms of the quantities with the tilde symbol $\tilde{A}_\alpha(z)$, $\tilde{D}_{\alpha\beta}^{(0)}(z, z')$, $\tilde{\Gamma}_\alpha^{(0)}(y, x|z)$ contain components with wavevectors $\mathbf{G}_n + \mathbf{Q}$ (\mathbf{G}_n denotes a reciprocal-lattice vector). The action $S^{(e-e)}$ describes the short-range part of the electron–electron interaction.

We have recently solved the problem of the energy spectrum of the system under consideration by using the direct microscopic treatment and the Green function method (Koinov 1996). The main result, obtained in this paper, is that even in a case without a pump wave the excitonic–biexcitonic-polariton eigenmodes exist. Those elementary excitations manifest themselves as common poles of photon, and two- and four-particle electron–hole, Green functions. In what follows we will show that the excitonic–biexcitonic-polariton spectrum consists of three branches $\tilde{\Omega}_i(\mathbf{Q})$, $i = 1, 2, 3$. They can be determined by solving the following equation:

$$\tilde{\Omega} - \Omega_0(\mathbf{Q}) - \frac{|A_+(\mathbf{Q})|^2}{\tilde{\Omega} - \omega_+^{exc.pol.}(\mathbf{Q})} - \frac{|A_-(\mathbf{Q})|^2}{\tilde{\Omega} - \omega_-^{exc.pol.}(\mathbf{Q})} = 0. \quad (3a)$$

Here $\omega_{\pm}^{exc.pol.}(\mathbf{Q})$ are the two (upper and lower) excitonic–polariton energies, $\Omega_0(\mathbf{Q})$ is the biexciton ground state energy and $A_{\pm}(\mathbf{Q})$ are the matrix elements of the interaction between the biexcitons and the exciton components of the corresponding excitonic polaritons. For small wavevectors $|\mathbf{Q}| \ll Q_0 = (\sqrt{\varepsilon_0}/c)\omega_t$ the first excitonic–biexcitonic-polariton branch is almost identical to the photon-like lower excitonic-polariton branch:

$$\tilde{\Omega}_1(\mathbf{Q}) \approx \omega_-^{exc.pol.}(\mathbf{Q}) \quad (3b)$$

while the other branches are defined as follows:

$$\tilde{\Omega}_{2,3}(\mathbf{Q}) = \frac{1}{2}[\omega_+^{exc.pol.}(\mathbf{Q}) + \Omega_0(\mathbf{Q}) \mp \sqrt{(\omega_+^{exc.pol.}(\mathbf{Q}) - \Omega_0(\mathbf{Q}))^2 + 4|A_+(\mathbf{Q})|^2}]. \quad (3c)$$

In the opposite case $|\mathbf{Q}| \gg Q_0 = (\sqrt{\varepsilon_0}/c)\omega_t$ the third excitonic–biexcitonic branch $\tilde{\Omega}_3(\mathbf{Q})$ is identical to the photon-like upper excitonic-polariton branch:

$$\tilde{\Omega}_3(\mathbf{Q}) \approx \omega_+^{exc.pol.}(\mathbf{Q}) \quad (3d)$$

while the other two branches are

$$\tilde{\Omega}_{1,2}(\mathbf{Q}) = \frac{1}{2}[\omega_-^{exc.pol.}(\mathbf{Q}) + \Omega_0(\mathbf{Q}) \mp \sqrt{(\omega_-^{exc.pol.}(\mathbf{Q}) - \Omega_0(\mathbf{Q}))^2 + 4|A_-(\mathbf{Q})|^2}]. \quad (3e)$$

The purpose of this paper is to clarify how the concept of excitonic–biexcitonic-polariton mixed modes is connected with the bipolariton model. We will also discuss some major problems of the bipolariton model, which in our opinion cannot be solved in the frame of the phenomenological approach.

2. Phenomenological approach

2.1. The elementary excitation spectra in the zero-order approximation

Quite generally, to determine the positions and widths of the excitation energies in the system under consideration, one may search for the poles of the corresponding exact Green functions. It is well known that all Green functions can be obtained by functional differentiation from the generating functional for the connected Green functions $Z[I, J]$ which is defined as follows: $Z[I, J] = i \ln W[I, J]$, where I and J are the sources of the photon and exciton fields, and

$$W[I, J] = \int D\mu(\alpha, B) \exp\{i[S_M + I_{\sigma,p}\alpha_{\sigma,p} + J_{\sigma,p}B_{\sigma,p}]\}. \quad (4a)$$

The action S_M , which corresponds to the Hamiltonian (1a), has the form:

$$S_M = \frac{1}{2} \alpha_{\sigma,p} D_0^{-1}(\mathbf{p}, \omega) \alpha_{\sigma,p} + \frac{1}{2} B_{\sigma,p} G_0^{-1}(\mathbf{p}, \omega) B_{\sigma,p} + \frac{\Omega_c}{2} \alpha_{\sigma,p} B_{\sigma,p} + \frac{1}{2} W_{\sigma\sigma'}(\mathbf{q}) B_{\sigma,k} B_{\sigma',p} B_{\sigma',p+q} B_{\sigma,k-q}. \quad (4b)$$

Here $\alpha_{\sigma,p}$ and $B_{\sigma,p}$ are boson fields. In this paper we use the summation–integration convention: that repeated variables are summed up or integrated over. The free-particle retarded photon $D_0(\mathbf{p}, \omega)$ and exciton $G_0(\mathbf{p}, \omega)$ Green functions are given by

$$D_0(\mathbf{p}, \omega) = \frac{1}{\omega - \omega_p^\gamma + i0^+} \quad G_0(\mathbf{p}, \omega) = \frac{1}{\omega - \omega_p^x + i0^+}. \quad (4c)$$

In (4a) the measure $D\mu$ is given by $D\mu = \text{constant} \times d\alpha dB$, where the normalization constant is chosen in such a manner that $\int D\mu \exp(iS_M) = 1$.

Since the functional measure $D\mu$ must be invariant under the translations $B \Rightarrow B + \delta B$, $\alpha \Rightarrow \alpha + \delta\alpha$, the following Schwinger equations (also known as Dyson equations) occur:

$$D^{-1}(\mathbf{p}, \omega) = D_0^{-1}(\mathbf{p}, \omega) - \Pi(\mathbf{p}, \omega) \quad (5a)$$

$$G^{-1}(\mathbf{p}, \omega) = G_0^{-1}(\mathbf{p}, \omega) - \Sigma(\mathbf{p}, \omega). \quad (5b)$$

Here $\Pi(\mathbf{p}, \omega)$ and $\Sigma(\mathbf{p}, \omega)$ are the proper self-energy part of the photons and the exciton mass operator, respectively. Both of them can be written as a sum of a polariton component ($\Pi_{\Omega_c}(\mathbf{p}, \omega)$ or $\Sigma_{\Omega_c}(\mathbf{p}, \omega)$) and a component which depends on the exciton–exciton attractive potential ($\Pi_{W_{12}}(\mathbf{p}, \omega)$ or $\Sigma_{W_{12}}(\mathbf{p}, \omega)$):

$$\Pi(\mathbf{p}, \omega) = \Pi_{\Omega_c}(\mathbf{p}, \omega) + \Pi_{W_{12}}(\mathbf{p}, \omega) = \frac{\Omega_c^2}{4} G_0(\mathbf{p}, \omega) + \Pi_{W_{12}}(\mathbf{p}, \omega) \quad (6a)$$

$$\Sigma(\mathbf{p}, \omega) = \Sigma_{\Omega_c}(\mathbf{p}, \omega) + \Sigma_{W_{12}}(\mathbf{p}, \omega) = \frac{\Omega_c^2}{4} D_0(\mathbf{p}, \omega) + \Sigma_{W_{12}}(\mathbf{p}, \omega). \quad (6b)$$

Due to the last term in the right-hand side of the action (4b), the explicit forms of $\Pi_{W_{12}}(\mathbf{p}, \omega)$ and $\Sigma_{W_{12}}(\mathbf{p}, \omega)$ contains terms $W_{12}(\alpha BBB)$ and $W_{12}(\alpha BBBB)$, respectively. In the lowest order of the exciton–exciton attraction the Bethe–Salpeter equation for the two-particle exciton Green function K has the form:

$$K(\mathbf{p}, \mathbf{q}, \mathbf{K}; \omega) = K_0(\mathbf{p}, \mathbf{q}, \mathbf{K}; \omega) + \sum_{l,l'} K_0(\mathbf{p}, l, \mathbf{K}; \omega) W_{12}(l - l') K(l', \mathbf{q}, \mathbf{K}; \omega) \quad (7a)$$

where K_0 is the free two-exciton propagator

$$K_0(\mathbf{p}, \mathbf{q}, \mathbf{K}; \omega) = \delta_{pq} \frac{1}{2\pi i} \int d\Omega G\left(-\mathbf{q} + \frac{\mathbf{K}}{2}, -\Omega + \frac{1}{2}\omega\right) G\left(\mathbf{p} + \frac{\mathbf{K}}{2}, \Omega + \frac{1}{2}\omega\right). \quad (7b)$$

We note that the terms $\Pi_{W_{12}}(\mathbf{p}, \omega)$ and $\Sigma_{W_{12}}(\mathbf{p}, \omega)$ lead to the biexciton contributions to the poles of the photon and exciton Green functions, while the term $\Sigma_{\Omega_c}(\mathbf{p}, \omega)$ in the exciton Green function (5b) gives the polariton contribution to the two-exciton bound states.

In the zero-order approximation, the polariton contribution to the two-exciton bound states and the biexciton contribution to the polariton states can be neglected. In the range of validity of this approximation $\Pi_{W_{12}}(\mathbf{p}, \omega)$ and $\Sigma_{W_{12}}(\mathbf{p}, \omega)$ should be vanishingly small. In this case $\Pi_{\Omega_c}(\mathbf{p}, \omega)$ and $\Sigma_{\Omega_c}(\mathbf{p}, \omega)$ are the dominant parts of the proper self-energy of the photons and the exciton mass operator, respectively. Thus, one has the specific case where the excitons and photons are coupled into excitonic polaritons. In this approximation there exist two excitonic-polariton branches $\omega_\mu(\mathbf{p})$ (the upper ($\mu = +$) and the lower ($\mu = -$)):

$$\omega_\mu(\mathbf{p}) = \omega_\pm(\mathbf{p}) = \frac{1}{2}[(\omega_p^x + \omega_p^\gamma) \pm \sqrt{(\omega_p^x - \omega_p^\gamma)^2 + \Omega_c^2}] \quad (8a)$$

which manifest themselves as common poles of the photon and exciton Green functions, defined as follows:

$$\tilde{G}(\mathbf{p}, \omega) = \left(G_0^{-1}(\mathbf{p}, \omega) - \frac{\Omega_c^2}{4} D_0(\mathbf{p}, \omega) \right)^{-1} = \sum_{\mu=\pm} \frac{\Phi_\mu(\mathbf{p}, \omega)}{\omega - \omega_\mu(\mathbf{p}) + i0^+} \quad (8b)$$

$$\tilde{D}(\mathbf{p}, \omega) = \left(D_0^{-1}(\mathbf{p}, \omega) - \frac{\Omega_c^2}{4} G_0(\mathbf{p}, \omega) \right)^{-1} = \sum_{\mu=\pm} \frac{\varphi_\mu(\mathbf{p}, \omega)}{\omega - \omega_\mu(\mathbf{p}) + i0^+}. \quad (8c)$$

The functions Φ_μ (or φ_μ) characterize the distribution of the exciton (or photon) component between the upper and lower polariton branches:

$$\Phi_\mu(\mathbf{p}, \omega) = (\mu \cdot 1) \frac{\omega_\mu(\mathbf{p})(\omega - \omega_p^y)}{\omega(\omega_+(\mathbf{p}) - \omega_-(\mathbf{p}))} \quad (8d)$$

$$\varphi_\mu(\mathbf{p}, \omega) = (\mu \cdot 1) \frac{\omega_\mu(\mathbf{p})(\omega - \omega_p^x)}{\omega(\omega_+(\mathbf{p}) - \omega_-(\mathbf{p}))}. \quad (8e)$$

In the zero-order approximation the free two-exciton propagator K_0 can be calculated by using the Green functions $G_0(\mathbf{p}, \omega)$ instead of the exact one:

$$K_0(\mathbf{p}, \mathbf{q}, \mathbf{K}; \omega) = \delta_{pq} (\omega - \omega_{p-K/2}^x - \omega_{-p-K/2}^x + i0^+)^{-1}. \quad (9a)$$

In this approximation the poles of the two-exciton Green function $\Omega_m(\mathbf{K})$ (m is the total quantum number) determine the biexciton energy. If we restrict the range of frequencies ω near to the neighbourhood of position $\Omega_m(\mathbf{K})$, we may write:

$$K(\mathbf{p}, \mathbf{q}, \mathbf{K}; \omega) \approx \frac{\Psi_{m\mathbf{K}}(\mathbf{p})\Psi_{m\mathbf{K}}^*(\mathbf{q})}{\omega - \Omega_m(\mathbf{K}) + i0^+} \quad (9b)$$

where $\Psi_{m\mathbf{K}}(\mathbf{p})$ is the Fourier transforms of the corresponding biexciton eigenfunctions. By comparing the residues of both sides of the Bethe–Salpeter equation (7a) we obtain the following Schrödinger equation in the momentum representation:

$$\sum_{\mathbf{q}} \{[\omega_{p+K/2}^x + \omega_{-p+K/2}^x] \delta_{pq} + W_{12}(\mathbf{p} - \mathbf{q})\} \Psi_{m\mathbf{K}}(\mathbf{q}) = \Omega_m(\mathbf{K}) \Psi_{m\mathbf{K}}(\mathbf{p}). \quad (9c)$$

Thus, in the zero-order approximation the quasiparticles in the system under consideration are of two types: excitonic polaritons and biexcitons.

2.2. Polariton contribution to the biexciton spectra

Due to the terms $\Pi_{W_{12}}(\mathbf{p}, \omega)$ and $\Sigma_{W_{12}}(\mathbf{p}, \omega)$ the analysis of Bethe–Salpeter equation (7a) is a complicated problem. A possible approximation to simplify the task is to assume that the branches in the low-energy part of the spectrum are mostly excitonic polaritons with a small biexciton contribution (that can be neglected), and the high-energy part is mostly a biexciton one with a small polariton content. This is the so-called bipolariton approximation. In the range of validity of this approximation $\Pi_{W_{12}}(\mathbf{p}, \omega)$ and $\Sigma_{W_{12}}(\mathbf{p}, \omega)$ should be vanishingly small. In this case $\Pi_{\Omega_c}(\mathbf{p}, \omega)$ and $\Sigma_{\Omega_c}(\mathbf{p}, \omega)$ are the dominant parts of the proper self-energy of the photons and the exciton mass operator, respectively. Thus, one has the specific case where the excitons and photons are coupled into excitonic polaritons, while the biexciton states are renormalized due to the polariton effect.

By comparing the residues of both sides of the equation (7a) the following Bethe–Salpeter equation for determining the renormalized (bipolariton) wavefunction $\tilde{\Psi}_v(\mathbf{p}, \mathbf{K})$ and the corresponding energy $\tilde{\Omega}_v(\mathbf{K})$ is obtained:

$$\tilde{\Psi}_v(\mathbf{p}, \mathbf{K}) = K_0(\mathbf{p}, \mathbf{p}, \mathbf{K}; \tilde{\Omega}_v(\mathbf{K})) \sum_{\mathbf{q}} W_{12}(\mathbf{p} - \mathbf{q}) \tilde{\Psi}_v(\mathbf{q}, \mathbf{K}) \quad (10a)$$

where

$$K_0(\mathbf{p}, \mathbf{q}, \mathbf{K}; \omega) = \delta_{pq} \sum_{\mu, \mu'=\pm} \frac{\Phi_{\mu}(-\mathbf{p} + \mathbf{K}/2, \omega - \omega_{\mu'}(-\mathbf{p} + \mathbf{K}/2)) \Phi_{\mu}^*(\mathbf{p} + \mathbf{K}/2, \omega_{\mu'}(\mathbf{p} + \mathbf{K}/2))}{\omega - \omega_{\mu}(-\mathbf{p} + \mathbf{K}/2) - \omega_{\mu'}(\mathbf{p} + \mathbf{K}/2) + i0^+}. \quad (10b)$$

The renormalized biexciton ground-state wavefunction $\tilde{\Psi}_0(\mathbf{p}, \mathbf{K})$ and energy $\tilde{\Omega}_0(\mathbf{K})$ can be determined by solving the equation:

$$\tilde{\Psi}_0(\mathbf{p}, \mathbf{K}) = K_0(\mathbf{p}, \mathbf{p}, \mathbf{K}; \tilde{\Omega}_0(\mathbf{K})) \sum_{\mathbf{q}} W_{12}(\mathbf{p} - \mathbf{q}) \tilde{\Psi}_0(\mathbf{q}, \mathbf{K}). \quad (10c)$$

A similar equation was first derived by Ivanov and Haug (1993), but instead of (10b), the following propagator has been used:

$$K_0^{I,H}(\mathbf{p}, \mathbf{p}, \mathbf{K}; \omega) = \delta_{pq} \frac{\Phi_{-}(-\mathbf{p} + \mathbf{K}/2, \omega_{-}(-\mathbf{p} + \mathbf{K}/2)) \Phi_{-}^*(\mathbf{p} + \mathbf{K}/2, \omega_{-}(\mathbf{p} + \mathbf{K}/2))}{\omega - \omega_{-}(-\mathbf{p} + \mathbf{K}/2) - \omega_{-}(\mathbf{p} + \mathbf{K}/2) + i0^+}. \quad (10d)$$

This form of the propagator follows from (10b) when: (i) the lower polariton branch $\omega_{-}(\mathbf{p})$ is taken into consideration; (ii) the following approximation $\tilde{\Omega}_0(\mathbf{K}) \approx \omega_{-}(\mathbf{p} + \mathbf{K}/2) + \omega_{-}(-\mathbf{p} + \mathbf{K}/2)$ has been assumed.

Let $\mathbf{p}_0 = \mathbf{p}_0(\mathbf{K})$ denote the root of the equation

$$\tilde{\Omega}_0(\mathbf{K}) - \omega_{\mu'}\left(\mathbf{p}_0 + \frac{\mathbf{K}}{2}\right) + \omega_{\mu}\left(-\mathbf{p}_0 + \frac{\mathbf{K}}{2}\right) = 0.$$

The existence of the root of the above equation leads to the complex value of the renormalized biexciton energy $\tilde{\Omega}_0(\mathbf{K}) = \text{Re} \tilde{\Omega}_0(\mathbf{K}) + i \text{Im} \tilde{\Omega}_0(\mathbf{K})$ (in the case when $\text{Im} \tilde{\Omega}_0(\mathbf{K}) = 0$, the bipolariton energy $\tilde{\Omega}_0(\mathbf{K})$ is a pole not only of two-particle Green function K , but of the propagator (10b) as well).

The Bethe–Salpeter equation (10c) (with $K_0 = K_0^{I,H}$) has been numerically solved by Ivanov and Haug (1995a, b) for the case of CuCl (the biexciton ground state at the point $\mathbf{K} = \mathbf{0}$ is $\Omega_0 = 6.362$ eV). The obtained biexciton Lamb shift at the point $\mathbf{K} = \mathbf{0}$ and the corresponding radiative lifetime are: $\Delta_0 = 0.103$ meV and $\Gamma_0 = 27.4$ μeV .

The main problem with the bipolariton approximation is that this approximation does not provide the correct elementary excitation spectrum in the excitonic-polariton energy part, since the existence of the biexcitons may modify the excitonic-polariton states. Beyond the bipolariton approximation, the problem becomes much more complicated, since in this case, the excitonic polaritons are no longer stable states, as in the bipolariton model. $\Pi_{W_{12}}(\mathbf{p}, \omega)$ and $\Sigma_{W_{12}}(\mathbf{p}, \omega)$ describe both the decay of the excitonic-polariton states and the renormalization of the upper and lower polariton branches due to the biexciton states.

As emphasized earlier, due to the last term in the action (4b), going beyond the bipolariton approximation and including $\Pi_{W_{12}}(\mathbf{p}, \omega)$ and $\Sigma_{W_{12}}(\mathbf{p}, \omega)$ terms in calculations is very difficult to accomplish not only by using the diagrammatic perturbation theory, but by employing the field-theoretical technique as well. We, therefore, turn to the direct microscopic treatment of the initial electron–photon system.

3. Direct microscopic approach

3.1. Generating functional for connected Green functions and Schwinger equations

The method used in what follows is a completely microscopic treatment, which is based on the assumption that the biexcitons are four-particle (two electrons and two holes) bound

states. The starting point of the method we now wish to apply is the statement that all Matsubara Green functions can be obtained by functional differentiation from the generating functional for the connected Green function $Z[J, M, N]$ which is defined as follows:

$$Z[J, M, N] = \ln W[J, M, N] \quad (11a)$$

where J is the source of the photon field; M and N are the two- and four-particle electron-hole sources and

$$W[J, M, N] = \int D\mu(\bar{\Psi}, \Psi, A) \exp\{S + J_\alpha(z)A_\alpha(z) - \bar{\Psi}(y)M(y, x)\Psi(x) + \bar{\Psi}(y)\bar{\Psi}(y')N(y, y'; x, x')\Psi(x)\Psi(x')\}. \quad (11b)$$

The measure $D\mu$ is given by $D\mu + \text{constant } d\bar{\Psi} d\Psi dA$, where the normalization constant is chosen in such a manner that $\int D\mu \exp(S) = 1$. The source $N(y, y'; x, x')$ is antisymmetric over y, y' and x, x' arguments: $N(y, y'; x, x') = -N(y', y; x, x') = -N(y, y'; x', x)$.

As a preliminary step, we shall define the Green functions by using the generating functional and functional differentiation over the corresponding sources (all functional derivatives over the fermion fields and over the sources of the fermion type are left-handed ones and after the functional differentiation one should set $J = M = N = 0$).

Photon Green function.

$$D_{\alpha\beta}(z, z') = -\langle \hat{T}_v \{A_\alpha(z)A_\beta(z')\} \rangle = -\frac{\delta^2 Z}{\delta J_\alpha(z)\delta J_\beta(z')}. \quad (11c)$$

One-particle electron Green function.

$$G(x, y) = -\langle \hat{T}_u \{\Psi(x)\bar{\Psi}(y)\} \rangle = -\frac{\delta Z}{\delta M(y, x)}. \quad (11d)$$

Two-particle electron-hole Green function.

$$K \begin{pmatrix} x & y' \\ y & x' \end{pmatrix} = -\langle \hat{T}_u \{\bar{\Psi}(y)\Psi(x)\bar{\Psi}(y')\Psi(x')\} \rangle = -\frac{\delta^2 Z}{\delta N(y, x)\delta M(y', x')}. \quad (11e)$$

Four-particle electron-hole Green function.

$$R \begin{pmatrix} x_{21} & y_{43} \\ y_{21} & x_{43} \end{pmatrix} = -\langle \hat{T}_u \{\bar{\Psi}(y_3)\bar{\Psi}(y_4)\Psi(x_3)\Psi(x_4)\bar{\Psi}(y_1)\bar{\Psi}(y_2)\Psi(x_1)\Psi(x_2)\} \rangle \\ = -\frac{\delta^2 Z}{\delta N(y_{12}; x_{12})\delta N(y_{34}; x_{34})} \quad (11f)$$

where \hat{T} is a u - (or u' , v -) ordering operator, $y_{12} = \{y_1, y_2\}$, $x_{12} = \{x_1, x_2\}$ are composite variables and the brackets $\langle \hat{O} \rangle$ on an operator \hat{O} mean that the thermodynamic average is taken.

Electron-photon vertex function.

$$\Gamma_\alpha(y, x|z) = -\frac{\delta G^{-1}(y, x)}{\delta J_\beta(z')} D_{\alpha\beta}^{-1}(z', z). \quad (11g)$$

The rest of the functional derivatives of Z with respect to the sources are:

$$R_\alpha(z) = -\frac{\delta Z}{\delta J_\alpha(z)} \quad \Delta(x_{21}; y_{21}) = -\frac{\delta Z}{\delta N(y_{12}; x_{12})} \quad (11h) \\ Y_\alpha(x, y|z) = -\frac{\delta^2 Z}{\delta J_\alpha(z)\delta M(y, x)} \quad X_\alpha(x_{21}, y_{21}|z) = -\frac{\delta^2 Z}{\delta J_\alpha(z)\delta N(y_{12}, x_{12})}$$

$$Z(x_{21}, y_{21}|x, y) = -\frac{\delta^2 Z}{\delta M(y, x)\delta N(y_{12}, x_{12})}. \tag{11i}$$

As a consequence of the fact that the measure $d\bar{\Psi} d\Psi dA$ is invariant under the translations $\bar{\Psi} \Rightarrow \bar{\Psi} + \delta\bar{\Psi}$; $A \Rightarrow A + \delta A$ one can derive the Schwinger equations which in our case have the form:

$$-D_{\alpha\beta}^{(0)-1}(z, z')R_\beta(z') + \Gamma_\alpha^{(0)}(y, x|z)G(x, y) + J_\alpha(z) = 0 \tag{12a}$$

$$G^{-1}(y_1, x_1) = G^{(0)-1}(y_1, x_1) - M(y_1, x_1) = \Sigma(y_1, x_1) - 2N(y_{12}; x_{34})\Delta(x_{43}; y_{25})G^{-1}(y_5, x_1) \tag{12b}$$

$$[G^{(0)-1}(y_1, x_2) - M(y_1, x_2)]\Delta(x_{21}; y_{43}) = \underset{[y_3, y_4]}{\text{Asymm}}\{\delta(y_1 - y_3)G(x_1, y_4)\} + T(y_1, x_1|y_{43}) + 2N(y_{15}; x_{56})Z(x_{65}, y_{43}|x_1, y_5) \tag{12c}$$

where the symbol $\underset{[y_3, y_4]}{\text{Asymm}}\{\delta(y_1 - y_3)G(x_1, y_4)\}$ means antisymmetrization over the set of variables $[y_1, y_2]$.

The mass operator Σ in (12b) has the form:

$$\begin{aligned} \Sigma(y_1, x_1) = & \Gamma_\alpha^{(0)}(y_1, x_1|z)R_\alpha(z) - \Gamma_\alpha^{(0)}(y_1, x_2|z)G(x_2, y_2)\Gamma_\beta(y_2, x_1|z')D_{\alpha\beta}(z, z') \\ & - \tilde{\Gamma}_\alpha^{(0)}(y_1, x_2|z)\tilde{D}_{\alpha\beta}^{(0)}(z, z')\tilde{\Gamma}_\beta^{(0)}(y_3, x_3|z')K \begin{pmatrix} x_3 & y_2 \\ y_3 & x_2 \end{pmatrix} G^{-1}(y_2, x_1) \\ & - \tilde{\Gamma}_\alpha^{(0)}(y_1, x_1|z)\tilde{D}_{\alpha\beta}^{(0)}(z, z')\tilde{\Gamma}_\beta^{(0)}(y_2, x_2|z')G(x_2, y_2). \end{aligned} \tag{13a}$$

The operator $T(y_1, x_1|y_{43})$ in (12c) is called the polarization operator of the four-particle Green function and has the form:

$$\begin{aligned} T(y_1, x_1|y_{43}) = & \Gamma_\alpha^{(0)}(y_1, x_2|z)\Delta(x_{21}; y_{43})R_\alpha(z) - \Gamma_\alpha^{(0)}(y_1, x_2|z)\frac{\delta\Delta(x_{21}; y_{43})}{\delta J_\alpha(z)} \\ & - \tilde{\Gamma}_\alpha^{(0)}(y_1, x_3|z)\tilde{D}_{\alpha\beta}^{(0)}(z, z')\tilde{\Gamma}_\beta^{(0)}(y_2, x_2|z')Z(x_{32}, y_{43}|x_1, y_2). \end{aligned} \tag{13b}$$

Since all variables in the Schwinger equations (12) are essentially the functional derivatives of Z with respect to the sources, one can say that the Schwinger equations are equations for the generating functional Z .

3.2. Legendre transform and equations for Green and vertex functions

The basic idea in our method is to use the Legendre transform in order to derive a set of exact equations for the Green functions and vertex functions. Legendre transforms have appeared in a wide variety of roles in both quantum field theory and statistical mechanics. As we will see in what follows there is probably no need to advertise the utility of the Legendre transform in polariton theory. It is convenient to treat (11d) and (11h) as definitions for functional $R[J, M, N]$, $G[J, M, N]$ and $\Delta[J, M, N]$. If those functionals are invertible with inverses $J[R, G, \Delta]$, $M[R, G, \Delta]$ and $N[R, G, \Delta]$, then we define the Legendre transform by

$$V[R, G, \Delta] = Z[R, G, \Delta] - J_\alpha(z)\frac{\delta Z}{\delta J_\alpha(z)} - M(y, x)\frac{\delta Z}{\delta M(y, x)} - N(y_{12}; x_{12})\frac{\delta Z}{\delta N(y_{12}; x_{12})}. \tag{14}$$

After going over from functional Z to the Legendre transform, the mass operator Σ and the polarization operator T must be considered as functionals as R , G and Δ . From definition (14) one can obtain the dual relations to (11d) and (11h)

$$\frac{\delta V}{\delta R_\alpha(z)} = J_\alpha(z) \quad \frac{\delta V}{\delta G(y, x)} = M(y, x) \quad \frac{\delta V}{\delta \Delta(x_{21}; y_{21})} = N(y_{12}; x_{12}).$$

We now wish to return to our statement that the Green functions are the thermodynamic average of the \hat{T}_u -ordered products of field operators. The standard procedure for calculating the Green functions is to apply Wick's theorem that enables us to evaluate the \hat{T}_u -ordered products of field operators as a perturbation expansion involving only wholly contracted field operators. These expansions can be summed formally to yield different equations of Green functions. The main disadvantage of this procedure is that the validity of the equations must be verified diagram by diagram. For this reason we will derive a set of nine independent exact equations for corresponding vertex and Green functions by using the important property of the Legendre transform—its orthogonality,

$$\frac{\delta^2 Z}{\delta A_i(x) \delta A_j(x')} \frac{\delta^2 V}{\delta B_j(x') \delta B_k(x'')} = -\delta_{ik} \delta(x - x'').$$

Here $A_1(x) = J_\alpha(z)$, $A_2(x) = M(y, x)$, $A_3(x) = N(y_{12}; x_{12})$ are the sources of the corresponding fields and $B_1(x) = R_\alpha(z)$, $B_2(x) = G(x, y)$, $B_3(x) = \Delta(x_{21}; y_{21})$ are the corresponding dual quantities. Thus taking $i = 1, 2, 3$ and $k = 1, 2, 3$ we obtain a set of nine independent exact equations:

$$D_{\alpha\beta}(z, z') = D_{\alpha\beta}^{(0)}(z, z') + D_{\alpha\gamma}^{(0)}(z, z'') \Gamma_\gamma^{(0)}(y, x|z'') K^{(0)} \begin{pmatrix} x & y' \\ y & x' \end{pmatrix} \Gamma_\delta(y', x'|z''') D_{\delta\beta}(z''', z') \quad (15a)$$

$$D_{\alpha\beta}(z, z') \Gamma_\beta^{(0)}(y, x|z') - K^{(0)} \begin{pmatrix} x' & y'' \\ y' & x'' \end{pmatrix} \Gamma_\beta(y'', x''|z') D_{\beta\alpha}(z', z) \frac{\delta^2 V}{\delta G(x', y') \delta G(x, y)} + X_\alpha(x_{21}; y_{21}|z) \frac{\delta^2 V}{\delta \Delta(x_{21}; y_{21}) \delta G(x, y)} = 0 \quad (15b)$$

$$-K^{(0)} \begin{pmatrix} x & y' \\ y & x' \end{pmatrix} \Gamma_\beta(y', x'|z') D_{\beta\alpha}(z', z) \frac{\delta^2 V}{\delta \Delta(x_{21}; y_{21}) \delta G(x, y)} + X_\alpha(x_{43}; y_{43}|z) \frac{\delta^2 V}{\delta \Delta(x_{43}; y_{43}) \delta \Delta(x_{21}; y_{21})} = 0 \quad (15c)$$

$$K^{(0)} \begin{pmatrix} x & y' \\ y & x' \end{pmatrix} \Gamma_\beta(y', x'|z') D_{\beta\alpha}(z', z) = K \begin{pmatrix} x & y' \\ y & x' \end{pmatrix} \Gamma_\beta^{(0)}(y', x'|z') D_{\beta\alpha}^{(0)}(z', z) \quad (15d)$$

$$K^{(0)} \begin{pmatrix} x & y'' \\ y & x'' \end{pmatrix} \Gamma_\beta(y'', x''|z') D_{\beta\alpha}(z', z) \Gamma_\alpha^{(0)}(y', x'|z) - K \begin{pmatrix} x & y'' \\ y & x'' \end{pmatrix} \frac{\delta^2 V}{\delta G(x'', y'') \delta G(x', y')} + Z(x_{21}; y_{21}|x, y) \frac{\delta^2 V}{\delta \Delta(x_{21}; y_{21}) \delta G(x', y')} = -\delta(x - x') \delta(y - y') \quad (15e)$$

$$-K \begin{pmatrix} x & y' \\ y & x' \end{pmatrix} \frac{\delta^2 V}{\delta G(x', y') \delta \Delta(x_{21}; y_{21})} + Z(x_{43}; y_{43}|x, y) \frac{\delta^2 V}{\delta \Delta(x_{43}; y_{43}) \delta \Delta(x_{21}; y_{21})} = 0 \quad (15f)$$

$$X_\beta(x_{21}; y_{21}|z') D_{\beta\alpha}^{(0)-1}(z', z) - Z(x_{21}; y_{21}|x, y) \Gamma_\alpha^{(0)}(y, x|z) = 0 \quad (15g)$$

$$-X_\alpha(x_{21}; y_{21}|z) \Gamma_\alpha^{(0)}(y, x|z) + Z(x_{21}; y_{21}|x', y') \frac{\delta^2 V}{\delta G(x', y') \delta G(x, y)} - R \begin{pmatrix} x_{21} & y_{43} \\ y_{21} & x_{43} \end{pmatrix} \frac{\delta^2 V}{\delta \Delta(x_{43}; y_{43}) \delta G(x, y)} = 0 \quad (15h)$$

$$Z(x_{21}; y_{21}|x, y) \frac{\delta^2 V}{\delta G(x, y) \delta \Delta(x_{43}; y_{43})} - R \begin{pmatrix} x_{21} & y_{65} \\ y_{21} & x_{65} \end{pmatrix} \frac{\delta^2 V}{\delta \Delta(x_{65}; y_{65}) \delta \Delta(x_{43}; y_{43})} = -\delta(x_{21} - x_{43}) \delta(y_{21} - y_{43}) \quad (15i)$$

where the symbol $\delta(x_{21} - x_{43})$ means

$$\delta(x_{21} - x_{43}) = \text{Asymm}_{[x_1, x_2]} \{ \delta(x_1 - x_3) \delta(x_2 - x_4) \}.$$

The above nine equations assume a central role in our theory, as can be seen in the rest of this paper.

3.3. Analytic properties of the Green functions and the elementary excitation spectra

Upon combining (15d) and (15e) one sees that the two-particle electron-hole Green function satisfies the Bethe-Salpeter equation:

$$K^{-1} \begin{pmatrix} y & x' \\ x & y' \end{pmatrix} = K_{e-\omega}^{-1} \begin{pmatrix} y & x' \\ x & y' \end{pmatrix} - I \begin{pmatrix} y & x' \\ x & y' \end{pmatrix}. \quad (16a)$$

Here we have defined the propagator $K_{e-\omega}^{-1} \begin{pmatrix} y & x' \\ x & y' \end{pmatrix}$ as follows

$$K_{e-\omega}^{-1} \begin{pmatrix} y & x' \\ x & y' \end{pmatrix} = \frac{\delta^2 V}{\delta G(x', y') \delta G(x, y)} - \Gamma_{\alpha}^{(0)}(y, x|z) D_{\alpha\beta}^{(0)}(z, z') \Gamma_{\beta}^{(0)}(y', x'|z'). \quad (16b)$$

The kernel $I \begin{pmatrix} y & x' \\ x & y' \end{pmatrix}$ has the form

$$I \begin{pmatrix} y & x' \\ x & y' \end{pmatrix} = \frac{\delta^2 V}{\delta G(x', y') \delta \Delta(x_{21}; y_{21})} R_0 \begin{pmatrix} x_{21} & y_{43} \\ y_{21} & x_{43} \end{pmatrix} \frac{\delta^2 V}{\delta \Delta(x_{43}; y_{43}) \delta G(x', y')} \quad (16c)$$

where we have introduced the four-particle propagator $R_0 \begin{pmatrix} x_{21} & y_{43} \\ y_{21} & x_{43} \end{pmatrix}$ defined by the equation

$$R_0 \begin{pmatrix} x_{21} & y_{65} \\ y_{21} & x_{65} \end{pmatrix} \frac{\delta^2 V}{\delta \Delta(x_{65}; y_{65}) \delta \Delta(x_{43}; y_{43})} = \delta(x_{43} - x_{21}) \delta(y_{43} - y_{21}). \quad (17)$$

By means of (17) one may rewrite the inverse two-particle Green function (16a) in the form

$$K^{-1} \begin{pmatrix} y & x' \\ x & y' \end{pmatrix} = K_M^{E-1} \begin{pmatrix} y & x' \\ x & y' \end{pmatrix} - \Gamma_{\alpha}^{(0)}(y, x|z) D_{\alpha\beta}^{(0)}(z, z') \Gamma_{\beta}^{(0)}(y', x'|z') \quad (18a)$$

where

$$K_M^{E-1} \begin{pmatrix} y & x' \\ x & y' \end{pmatrix} = \frac{\delta^2 V}{\delta G(x, y) \delta G(x', y')} - \frac{\delta^2 V}{\delta G(x, y) \delta \Delta(x_{21}, y_{21})} R_0 \begin{pmatrix} x_{21} & y_{43} \\ y_{21} & x_{43} \end{pmatrix} \times \frac{\delta^2 V}{\delta \Delta(x_{43}, y_{43}) \delta G(x', y')}. \quad (18b)$$

In a diagrammatic language K_M^E can be obtained by subtracting from the two-particle Green function K any diagrams that may be separated into two parts by cutting only a long-wavelength phonon line. In the case when the existence of the four-particle states can be neglected (if we set $\delta^2 V / (\delta \Delta \delta G) = 0$ in (18b)) the propagator K_M^E assumes the standard form of the two-particle Green function for ‘mechanical’ excitons when the Elliott exchange interaction is included.

From the exact equations (15d) and (15c) one can obtain the Dyson equation for the long-wavelength photon Green function

$$D_{\alpha\beta}(z, z') = D_{\alpha\beta}^{(0)}(z, z') + D_{\alpha\gamma}^{(0)}(z, z'') \Pi_{\gamma\delta}^{(L)}(z'', z''') D_{\delta\beta}(z''', z') \quad (18c)$$

where $\Pi_{\alpha\beta}^{(L)}$ is the self-energy part of the long-wavelength photons:

$$\Pi_{\alpha\beta}^{(L)}(z, z') = \Gamma_{\alpha}^{(0)}(y, x|z) K_M^E \begin{pmatrix} x & y' \\ y & x' \end{pmatrix} \Gamma_{\beta}^{(0)}(y', x'|z'). \quad (18d)$$

The propagator (18b) is connected with $\Pi_{\alpha\beta}^{(L)}$ and, therefore, it is of the first importance for calculation of the dielectric tensor $\varepsilon_{\alpha\beta}(\mathbf{Q}, \omega)$ (Koinov 1996).

Further, we will derive an equation for the four-particle Green function using the method of Legendre transforms. If we combined equations (15g) and (15h) the following relationship can be obtained

$$Z(x_{21}; y_{21}|x, y) = R \begin{pmatrix} x_{21} & y_{43} \\ y_{21} & x_{43} \end{pmatrix} \frac{\delta^2 V}{\delta \Delta(x_{43}; y_{43}) \delta G(x', y')} K_{e-\omega} \begin{pmatrix} x' & x \\ y' & y \end{pmatrix}.$$

Insertion of the last equation into (15i) allows us to obtain the following Bethe–Salpeter equations for the four-particle Green function

$$R \begin{pmatrix} x_{21} & y_{43} \\ y_{21} & x_{43} \end{pmatrix} = R_0 \begin{pmatrix} x_{21} & y_{43} \\ y_{21} & x_{43} \end{pmatrix} + R_0 \begin{pmatrix} x_{21} & y_{65} \\ y_{21} & x_{65} \end{pmatrix} P \begin{pmatrix} y_{65} & x_{87} \\ x_{65} & y_{87} \end{pmatrix} R \begin{pmatrix} x_{87} & y_{43} \\ y_{87} & x_{43} \end{pmatrix} \quad (19a)$$

where the inhomogeneous term R_0 is defined by (17) and the kernel P has the form:

$$P \begin{pmatrix} y_{21} & x_{43} \\ x_{21} & y_{43} \end{pmatrix} = \frac{\delta^2 V}{\delta \Delta(x_{21}; y_{21}) \delta G(x, y)} K_{e-\omega} \begin{pmatrix} x & y' \\ y & x' \end{pmatrix} \frac{\delta^2 V}{\delta G(x', y') \delta \Delta(x_{43}; y_{43})}. \quad (19b)$$

It should be noted that the three Green functions $D_{\alpha\beta}$, K and R are not independent. Insertion of (15d) into (15a) allows us to obtain the following relationship between photon and two-particle Green functions

$$D_{\alpha\beta}(z, z') = D_{\alpha\beta}^{(0)}(z, z') + D_{\alpha\gamma}^{(0)}(z, z'') \Gamma_{\gamma}^{(0)}(y, x|z'') K \begin{pmatrix} x & y' \\ y & x' \end{pmatrix} \Gamma_{\delta}^{(0)}(y', x'|z''') D_{\delta\beta}^{(0)}(z''', z'). \quad (20a)$$

In a similar way, upon combining (18) with (15d) and using (15e) one sees that the following relationship occurs

$$K \begin{pmatrix} x & y' \\ y & x' \end{pmatrix} = K_M^E \begin{pmatrix} x & y' \\ y & x' \end{pmatrix} + K_M^E \begin{pmatrix} x & y'' \\ y & x'' \end{pmatrix} \Gamma_{\alpha}^{(0)}(y'', x''|z) D_{\alpha\beta}(z, z') \Gamma_{\beta}^{(0)}(y''', x'''|z') \\ \times K_M^E \begin{pmatrix} x''' & y' \\ y''' & x' \end{pmatrix}. \quad (20b)$$

In order to obtain a relationship between two- and four-particle Green functions we use (15f) and (19a). The result is:

$$R \begin{pmatrix} x_{21} & y_{43} \\ y_{21} & x_{43} \end{pmatrix} = R_0 \begin{pmatrix} x_{21} & y_{43} \\ y_{21} & x_{43} \end{pmatrix} + R_0 \begin{pmatrix} x_{21} & y_{65} \\ y_{21} & x_{65} \end{pmatrix} \frac{\delta^2 V}{\delta \Delta(x_{65}; y_{65}) \delta G(x, y)} \\ \times K \begin{pmatrix} x & y' \\ y & x' \end{pmatrix} \frac{\delta^2 V}{\delta G(x', y') \delta \Delta(x_{87}; y_{87})} R_0 \begin{pmatrix} x_{87} & y_{43} \\ y_{87} & x_{43} \end{pmatrix}. \quad (20c)$$

From equations (20) one can conclude that the three Green functions $D_{\alpha\beta}$, K and R have common poles. It has long been known (Gell-Mann and Low 1951) that any well defined elementary excitation of wavevector \mathbf{Q} and energy $\omega_v(\mathbf{Q})$ of the system under consideration manifests itself as a pole near the real axis in the frequency plane of the function $K \begin{pmatrix} r_1\sigma_1 & r_3\sigma_3 \\ r_2\sigma_2 & r_4\sigma_4 \end{pmatrix} |u_2 - u_1; u_4 - u_3; z\rangle$. The latter is obtained from the Fourier transform of the two-particle Green function $K \begin{pmatrix} r_1\sigma_1 & r_3\sigma_3 \\ r_2\sigma_2 & r_4\sigma_4 \end{pmatrix} |u_2 - u_1; u_4 - u_3; i\omega_p\rangle$ by the

analytic continuation of the set of points $i\omega_p$ along the imaginary axis into the appropriate half of the z plane. Similarly, one can define the analytic continuation of the photon Green function $D_{\alpha\beta}(\mathbf{Q}, z)$. If we restrict the range of frequencies ω to the neighbourhood of position $\omega_v(\mathbf{Q})$, we may write:

$$K \begin{pmatrix} r_1\sigma_1 & r_3\sigma_3 \\ r_2\sigma_2 & r_4\sigma_4 \end{pmatrix} |u_2 - u_1; u_4 - u_3; \omega \rangle \approx \frac{\Phi^{vQ}(r_2\sigma_2; r_1\sigma_1; u_{21})\Phi^{vQ*}(r_4\sigma_4; r_3\sigma_3; u_{43})}{\omega - \omega_v(\mathbf{Q}) + i0^+} \quad (21a)$$

$$D_{\alpha\beta}(\mathbf{Q}, \omega) \approx \frac{A_\alpha^{vQ}(\mathbf{Q})A_\beta^{vQ*}(\mathbf{Q})}{\omega - \omega_v(\mathbf{Q}) + i0^+} \quad (21b)$$

where $\Phi^{vQ}(r_2\sigma_2; r_1\sigma_1; u_{21})$ and $A_\alpha^{vQ}(\mathbf{Q})$ are electron-hole and photon amplitudes, respectively.

Further, we continue with the analysis of the analytic properties of the four-particle Green function

$$R \begin{pmatrix} x_{21} & y_{65} \\ y_{43} & x_{87} \end{pmatrix} = R \begin{pmatrix} (\mathbf{r}, \sigma)_{21} & (\mathbf{r}, \sigma)_{65} \\ (\mathbf{r}, \sigma)_{43} & (\mathbf{r}, \sigma)_{87} \end{pmatrix} |u_4 - u_2; u_3 - u_1; \frac{1}{2}(u_4 + u_2 - u_3 - u_1)|u_8 - u_6; \\ u_7 - u_5; \frac{1}{2}(u_8 + u_6 - u_7 - u_5) | \frac{1}{4}(u_4 + u_3 + u_2 + u_1 - u_8 - u_6 - u_7 - u_5) \rangle.$$

It is well known that in the case of n particles, when $n \geq 3$, the kernels of the n -particle equations contain disconnected parts. Since the two-particle kernel and the two-particle Green function do not contain disconnected parts, one should find from (20c) a relationship between the connected part of the four-particle Green function

$$R^c \begin{pmatrix} (\mathbf{r}, \sigma)_{21} & (\mathbf{r}, \sigma)_{65} \\ (\mathbf{r}, \sigma)_{43} & (\mathbf{r}, \sigma)_{87} \end{pmatrix} |i\omega_{m_1}; i\omega_{m_2}; i\omega_{p_1}|i\omega_{m_3}; i\omega_{m_4}; i\omega_{p_2}|i\omega_p \rangle$$

and the two-particle ones. From this relationship we can conclude that the two Green functions K and R^c have identical poles. The function

$$R^c \begin{pmatrix} (\mathbf{r}, \sigma)_{21} & (\mathbf{r}, \sigma)_{65} \\ (\mathbf{r}, \sigma)_{43} & (\mathbf{r}, \sigma)_{87} \end{pmatrix} |i\omega_{m_1}; i\omega_{m_2}; i\omega_{p_1}|i\omega_{m_3}; i\omega_{m_4}; i\omega_{p_2}|z \rangle$$

is obtained by the analytic continuation of set of points $i\omega_p$ along the imaginary axis into the appropriate half of the z plane. If we restrict the range of frequencies ω to the neighbourhood of position $\omega_v(\mathbf{Q})$, we may write:

$$R^c \begin{pmatrix} (\mathbf{r}, \sigma)_{21} & (\mathbf{r}, \sigma)_{65} \\ (\mathbf{r}, \sigma)_{43} & (\mathbf{r}, \sigma)_{87} \end{pmatrix} |i\omega_{m_1}; i\omega_{m_2}; i\omega_{p_1}|i\omega_{m_3}; i\omega_{m_4}; i\omega_{p_2}|\omega \rangle \\ \approx \frac{\Psi^{vQ}((\mathbf{r}, \sigma)_{43}; (\mathbf{r}, \sigma)_{21}|i\omega_{m_1}; i\omega_{m_2}; i\omega_{p_1})\Psi^{vQ*}((\mathbf{r}, \sigma)_{87}; (\mathbf{r}, \sigma)_{65}|i\omega_{m_3}; i\omega_{m_4}; i\omega_{p_2})}{\omega - \omega_v(\mathbf{Q}) + i0^+} \quad (21c)$$

where $\Psi^{vQ}((\mathbf{r}, \sigma)_{43}; (\mathbf{r}, \sigma)_{21}|i\omega_{m_1}; i\omega_{m_2}; i\omega_{p_1})$ are the four-particle amplitudes, which can be obtained from the connected-kernel equation for the four-particle Green function.

The excitonic-biexcitonic-polariton spectrum can be calculated by searching for the poles of the photon, two- and four-particle Green functions, which satisfy equations (18c), (18a) and (19a), respectively. In what follows we will use the Bethe-Salpeter equation (19a). To solve this equation we assume the following two simplifications. The first one is to replace the inhomogeneous term R_0 by the usual biexciton Green function. The second simplification that is used concerns the kernel (19b): we take into account the contribution to the kernel which involve only terms, proportional to the second order of

$\delta^2 V / (\delta \Delta \delta G)$. In this case the exact propagator (16b) can be replaced by the following two-particle Green function:

$$K_{e-\omega}^{exc.pol.} \left(\begin{array}{cc} \mathbf{r}_1 \sigma_1 & \mathbf{r}_3 \sigma_3 \\ \mathbf{r}_2 \sigma_2 & \mathbf{r}_4 \sigma_4 \end{array} \middle| u_{21}; u_{43}; \omega \right) = \sum_{\mu} \frac{\varphi^{\mu Q}(\mathbf{r}_2 \sigma_2; \mathbf{r}_1 \sigma_1; u_{21}) \varphi^{\mu Q*}(\mathbf{r}_4 \sigma_4; \mathbf{r}_3 \sigma_3; u_{43})}{\omega - \omega_{\mu}^{exc.pol.}(\mathbf{Q}) + i0^+}. \quad (22)$$

Here the poles $\omega_{\mu}^{exc.pol.}(\mathbf{Q})$ of the above function determine the excitonic-polariton spectrum because this Green function describes the light propagation in crystals when the biexciton contribution to the polariton formation is neglected and $\varphi^{\mu Q}$ are the corresponding electron-hole amplitudes. The excitonic-polariton spectrum $\omega_{\mu}^{exc.pol.}(\mathbf{Q})$ can be calculated from the dielectric function $\varepsilon(\mathbf{Q}, \omega)$. In the case of direct-gap cubic semiconductors the simple one-oscillator model for the dielectric function can be used:

$$\varepsilon(\mathbf{Q}, \omega) = \varepsilon_b \left(1 + \frac{4\pi\beta_0\omega_t^2(\mathbf{Q}=\mathbf{0})}{\omega_t^2(\mathbf{Q}) - \omega^2} \right) \quad (23a)$$

where $\omega_t(\mathbf{Q})$ is the transverse exciton dispersion, ε_b is the background dielectric constant and $4\pi\beta_0 = 2\Delta_{LT}/\omega_t(\mathbf{Q}=\mathbf{0})$ is the oscillator strength. In this model the excitonic-polariton spectrum consists of two branches (the upper ($\mu = +$) and the lower ($\mu = -$)):

$$\omega_{\mu}^{exc.pol.}(\mathbf{Q}) = \frac{1}{\sqrt{2}} [A \pm \sqrt{A^2 - 4B^2}]^{1/2} \quad A = \frac{c^2 Q^2}{\varepsilon_b} + \omega_t^2(\mathbf{Q}) [1 + 4\pi\beta_0] \\ B^2 = \frac{c^2 Q^2 \omega_t^2(\mathbf{Q})}{\varepsilon_b}. \quad (23b)$$

The above two simplifications allow us to derive from the Bethe-Salpeter equation (19a) the following equation for the excitonic-biexcitonic-polariton spectrum:

$$\det \left\| [\omega - \Omega_m(\mathbf{Q})] \delta_{mm'} - \sum_{\mu} \frac{A_{m\mu}(\mathbf{Q}) A_{\mu m'}^*(\mathbf{Q})}{\omega - \omega_{\mu}^{exc.pol.}(\mathbf{Q})} \right\| = 0. \quad (24a)$$

Here $\Omega_m(\mathbf{Q})$ (m is the complete quantum number) denote the poles of the Green function R_0 and they determine the biexciton dispersion curves. The matrix element $A_{m\mu}(\mathbf{Q})$ describes the interaction between the biexciton with wavefunction $\Psi^{mQ}(\mathbf{r}_1, \sigma_1; \mathbf{r}_2, \sigma_2; \mathbf{r}_3, \sigma_3; \mathbf{r}_4, \sigma_4)$ and the excitonic component of the (μ, \mathbf{Q}) -excitonic polariton:

$$A_{m\mu}(\mathbf{Q}) = \langle \Psi^{mQ} | \frac{\delta^2 V}{\delta \Delta \delta G} | \varphi^{\mu Q} \rangle. \quad (24b)$$

If we take into account only the interaction between the biexciton ground state $\Omega_0(\mathbf{Q})$ and the two excitonic-polariton branches, then the equation (24a) assumes the form (3a), where $A_{\pm}(\mathbf{Q}) = A_{0\pm}(\mathbf{Q})$.

4. Discussion

As emphasized earlier, the bipolariton model does not take into consideration the important fact that the existence of four-particle bound states causes the additional polarization of the crystal, and, therefore, the biexcitons modify the excitonic-polariton spectrum. For this reason the bipolariton concept does not provide the correct spectrum of the system of interest. The last statement can be illustrated by considering the elementary excitation spectrum of

the system at the point $\mathbf{Q} = \mathbf{0}$. According to the bipolariton model the excitation spectrum consists of two excitonic-polariton states $\tilde{\Omega}_{1,2}$ and the renormalized biexciton state $\tilde{\Omega}_3$:

$$\begin{aligned}\tilde{\Omega}_1 &= \omega_-(\mathbf{Q} = \mathbf{0}) = \frac{1}{2}[\omega_t - \sqrt{\omega_t^2 + \Omega_c^2}] \approx -\frac{\Omega_c^2}{4\omega_t} = -\frac{\Delta_{LT}}{2} \\ \tilde{\Omega}_2 &= \omega_+(\mathbf{Q} = \mathbf{0}) = \frac{1}{2}[\omega_t + \sqrt{\omega_t^2 + \Omega_c^2}] \approx \omega_t + \frac{\Omega_c^2}{4\omega_t} = \omega_t + \frac{\Delta_{LT}}{2} \\ \tilde{\Omega}_3 &= \text{Re } \tilde{\Omega}_0(\mathbf{Q} = \mathbf{0}) = \Omega_0 + \Delta_0.\end{aligned}\quad (25)$$

We note that not only does the excitonic-polariton spectrum (8a) differ from (23b), obtained by the direct microscopic treatment, but near to the point $\mathbf{Q} = \mathbf{0}$ the lower branch $\omega_-(\mathbf{Q})$ becomes negative as well. Therefore, one can conclude that the procedure of replacing the initial electron-hole-photon Hamiltonian by the Hamiltonian (1a) leads to the wrong results, especially in the low-energy part of the spectrum.

According to the direct microscopic treatment, the elementary excitation spectrum at the point $\mathbf{Q} = \mathbf{0}$ is defined by (3b) and (3c):

$$\tilde{\Omega}_1 \approx \omega_-^{exc.pol.}(\mathbf{Q} = \mathbf{0}) = 0 \quad \tilde{\Omega}_2 \approx \tilde{\omega}_t + \Delta_{LT} \quad \tilde{\Omega}_3 \approx \Omega_0 + \Delta_0 \quad (26a)$$

where $\tilde{\omega}_t = \omega_t(\mathbf{Q} = \mathbf{0}) - \Delta_0$ and

$$\Delta_0 \approx \frac{|A_+(\mathbf{Q} = \mathbf{0})|^2}{\Omega_0 - \omega_t(\mathbf{Q} = \mathbf{0})}. \quad (26b)$$

The quantity Δ_0 determines both the biexciton ground-state energy shift and the transverse exciton energy shift. In our previous paper (Koinov 1996), based on the Dyson equation for the photon Green function a similar exciton energy shift has been already discussed. In this paper the shift (6b) has been introduced by means of the Bethe-Salpeter equation for the four-particle Green function. By comparing Δ_{biexc} , introduced in the previous paper with the shift (26b), one can see that the values of these two quantities are almost equal. This statement follows from the fact that at the point $\mathbf{Q} = \mathbf{0}$ the upper excitonic polariton branch has a very small photon component, and, therefore, the calculation of Δ_0 can be done by using the corresponding exciton wavefunction $F_{exc}^{n\mathbf{Q}=\mathbf{0}}$ of the upper excitonic polariton.

5. Summary

In this paper we have analysed the bipolariton concept, which has been recently used to calculate the elementary excitation spectrum of the electron-hole-photon system. In order to go beyond this approximation we have developed a finite-temperature Green function theory for the elementary excitation spectrum in the system of interacting electrons and photons, which takes into account the four-particle electron-hole bound states. We have shown that the bipolariton approximation can indeed be used for calculating the elementary excitation spectrum in the photon-exciton-biexciton system for the biexciton part. This concept does not provide the correct spectrum in the excitonic-polariton energy part, where the existence of biexcitons must be considered. By using the direct microscopic treatment we have obtained that even in a case without a pump wave the excitonic-biexcitonic-polariton eigenmodes exist and those elementary excitations manifest themselves as common poles of photon, and two- and four-particle electron-hole, Green functions.

References

- Gell-Mann M and Low F 1951 *Phys. Rev.* **84** 350
Ivanov A L and Haug H 1993 *Phys. Rev. B* **48** 1490
———1995a *Phys. Rev. Lett.* **74** 438
———1995b *Phys. Status Solidi b* **188** 61
Ivanov A L, Keldysh L V and Lanashchenko V V 1991 *Zh. Eksp. Teor. Fiz.* **99** 641
Koinov Z 1996 *J. Phys.: Condens. Matter* **8** L391
Koinov Z G and Glinskii G F 1988 *J. Phys. A: Math. Gen.* **21** 3431
Matsubara T 1955 *Prog. Theor. Phys.* **14** 351
Sheboul M I and Ekardt W 1976 *Phys. Status Solidi b* **73** 165