

Low-threshold exciton-biexciton optical Stark effect in direct-gap semiconductors

A. L. Ivanov, L. V. Keldysh, and V. V. Panashchenko

Moscow State University

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A new mechanism of exciton-biexciton modification of semiconductor spectra in the presence of a polariton pump wave is examined. This mechanism is attributable to the direct virtual coupling of two excitons forming a biexciton by Coulomb-like interaction. It is demonstrated that it is precisely this mechanism that is responsible for the spectral modification previously attributed exclusively to the “giant” oscillator strength of the exciton-biexciton optical transition and, specifically, may lead to an effective dynamical red-shift of the exciton level. This makes it possible to attribute this phenomenon to the exciton-biexciton optical Stark effect which can be observed at substantially lower pump wave intensities compared to the ordinary exciton Stark effect. A consistent microscopic theory of the exciton-biexciton Stark effect is formulated both on the basis of the exciton quasiparticle approximation and an analysis of the photon-electron-hole Hamiltonian of the semiconductor.

1. INTRODUCTION

Dynamical coherent phenomena in semiconductors are receiving extensive theoretical and experimental attention currently. Such phenomena are manifested as a modification of the spectra of elementary semiconductor excitations in the presence of intensive coherent electromagnetic radiation. One specific field of research has been devoted to an analysis of the case where the electromagnetic wave frequency lies in the transparency region near the exciton ground level characterizing the fundamental absorption edge. In this case, an intense electromagnetic wave, which will henceforth be referred to as the pump wave, travels as a polariton wave with virtually no damping in the semiconductor and can be treated as an external source wave. Such a pump wave, specifically, can lead to a number of dynamical phenomena in the crystal.

We will first focus on the exciton optical Stark-effect which is an active area of research at present.^{1–6} This effect is manifested as a blue-shift of the exciton line in the presence of a high intensity polariton wave. This intensity is given by the parameter $Na_{ex}^3 \sim 1$, where a_{ex} is the Bohr radius of the exciton in the ground state, while N here is the exciton concentration of the pump wave.

Studies of the dynamical changes in the polariton and biexciton spectra of the semiconductor are more conventional.^{7–14} Here the discussion primarily concerns the pump wave stimulated exciton-biexciton splitting of the semiconductor spectra near the resonance frequency $\omega = \Omega_0^{biox} - \omega_k$, where ω_k is the pump wave frequency, while $\hbar\Omega_0^{biox}$ is the biexciton excitation energy relative to the valence band. A theoretical description of this effect^{7–11} has customarily been based on an analysis of a system of independent boson excitations: Photons α_p , excitons B_p and biexcitons A_p with ordinary polariton exciton-phonon mixing and a phenomenological incorporation of three-particle interaction of the form $V^{-1/2}M_1(\mathbf{p}, \mathbf{q})A_p^+ B_q \alpha_{p-q}$. In this case, the matrix element M_1 , was attributed to the “giant” oscillator strength of the exciton-biexciton optical transition:^{15–17}

$$M_1(\mathbf{p}, \mathbf{q}) = -i \frac{\Omega_c}{2} \Psi\left(\mathbf{q} - \frac{\mathbf{p}}{2}\right), \quad (1)$$

where Ω_c is the polariton parameter characterizing the exciton-photon interaction strength, $\Psi(\mathbf{p})$ is the Fourier trans-

form of the wave function of exciton relative motion in the biexciton, and V is the crystal volume. The subsequent introduction of the polariton pump wave into the quasiparticle system resulted in this spectral splitting, which is determined by the matrix element M_1 and pump intensity I .

The purpose of this paper is to formulate a consistent microscopic approach to analyzing the exciton-photon-biexciton system of a semiconductor in an intense polariton wave. Such a consistent analysis makes it possible to identify a new spectral modification mechanism determined by the direct virtual coupling of two excitons that form a biexciton by their Coulomb-like interaction.^{18,19} This therefore refers to a process described by terms of the form $V^{-1/2}M_2(\mathbf{p}, \mathbf{q})A_p^+ B_q B_{p-q}$ in the corresponding quasiparticle Hamiltonian. Here, as we will demonstrate below, the stimulated exciton-biexciton splitting is not determined by the giant oscillator strength (1) of the exciton-biexciton transition, but rather solely by the matrix element $M_2(\mathbf{p}, \mathbf{q})$.

However, the most interesting result obtained here is the dynamical red-shift of the exciton level, which is dependent on pump wave intensity. This effect is also directly determined by Coulomb-like exciton-exciton interaction and can be defined as the exciton-biexciton optical Stark effect. This exciton-biexciton Stark effect has a lower pump wave intensity threshold compared to the exciton optical Stark-effect, since in this case the condition for observing the shifts is determined by the parameter $Na_{biox}^3 \gg Na_{ex}^3$, where a_{biox} is the radius of the biexciton in the ground state.

The following is pertinent in this connection. On the one hand, the exciton-biexciton optical Stark-effect has the same fundamental nature as the exciton Stark-effect: an exciton-exciton interaction in the semiconductor excited by the pump wave. On the other hand, the specific nature of the exciton-biexciton Stark-effect, particularly, its low observation threshold, arisen because under moderate ($Na_{biox}^3 \ll 1$) semiconductor excitation, in addition to the single characteristic dimensions a_{ex} in the exciton system, another natural scale, a_{biox} , appears.

2. THE EXCITON-BIEXCITON STARK-EFFECT IN THE EXCITON QUASIPARTICLE APPROXIMATION

We begin the analysis of the exciton-biexciton optical Stark-effect with the exciton quasiparticle approximation

which treats excitons as unstructured quasiparticles, and holds for $Na_{ex}^3 \ll 1$ (Ref. 20). In our case this is a valid approach, since the effects of interest to us arise at a substantially lower semiconductor excitation $Na_{bixex}^3 \sim 1$. The character of exciton-exciton interaction is essentially dependent on the spin structure of the interacting excitons. This makes it necessary to introduce into the model two types of excitons whose annihilation operators will be denoted by B_{1p} and B_{2p} , respectively.

In the simplest case, the various dipole-active excitons correspond to the doubly-degenerate exciton term and the only difference in their internal structure is that the spins of both the electrons and the holes comprising the excitons are in opposite directions. Optical excitation of excitons of various types may be related to the use of photons of specific polarizations whose annihilation operators in turn will be denoted by α_{1p} and α_{2p} . A possible experimental example of such a model will be discussed below. We therefore use the following exciton-photon Hamiltonian as the initial Hamiltonian:

$$H = \sum_{p,\sigma=1,2} \left[\omega_p^{ex} B_{\sigma p} + B_{\sigma p} + \omega_p^{ph} \alpha_{\sigma p} + \alpha_{\sigma p} + i \frac{\Omega_c}{2} (\alpha_{\sigma p} + B_{\sigma p} - B_{\sigma p} + \alpha_{\sigma p}) \right] + \frac{1}{2} \sum_{p,1,q,\sigma,\sigma'} W_{\sigma\sigma'}(\mathbf{q}) B_{\sigma p} + B_{\sigma'1} B_{\sigma'1+q} B_{\sigma p-q}, \quad (2)$$

where

$$\omega_p^{ex} = \omega_l + \frac{\hbar p^2}{2M}, \quad \omega_p^{ph} = \frac{cp}{\varepsilon_g^{1/2}}$$

are the unperturbed exciton and photon dispersions, respectively; ε_g is the background permittivity, M is the translational exciton mass, $\hbar\omega_l$ is the energy of the exciton state, and $W_{\sigma\sigma'}(\mathbf{q})$ is the Fourier transform of the exciton-exciton interaction potential. A number of studies²¹⁻²³ have been devoted to determining the Coulomb-like potential $W_{\sigma\sigma'}(\mathbf{q})$ in connection with analyses of the character of exciton-exciton scattering and calculation of a series of biexciton states.

It is significant that identical excitons are always mutually repulsive, e.g., $W_{\sigma\sigma}(\mathbf{q}) > 0$, while excitons of different type experience attraction. A characteristic plot of the relation $W = W_{12}(q) = W_{21}(q)$ for the isotropic semiconductor examined in the case below is shown in Fig. 1. The given

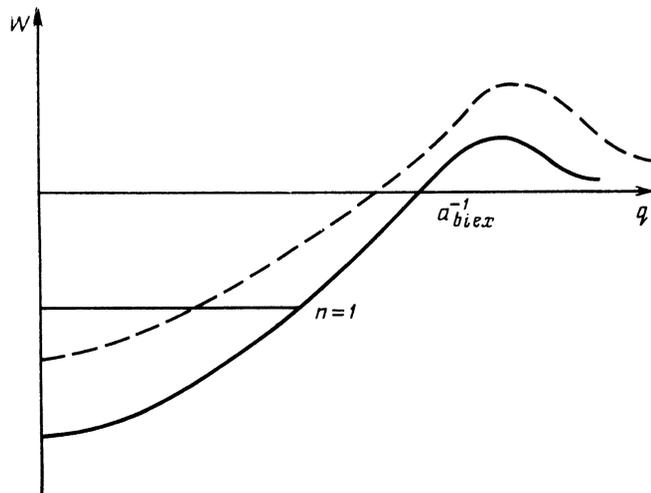


FIG. 1.

potential $W(q)$ may correspond to the coupled two-exciton complexes, biexcitons whose corresponding annihilation operator A_q is given by

$$A_q = \frac{1}{V^{1/2}} \sum_1 \Psi(1) B_{11+q/2} B_{2-1+q/2}, \quad (3)$$

where $\Psi(1)$ is the wave function in the momentum space relative to the motion of the two excitons in the biexciton normalized to unity. Note that such a representation is valid only for the case where the binding energy of the biexciton is substantially lower than the binding energy of its constituent excitons. An important element in the subsequent analysis will be the commutation relations characterizing the degree of independence of the quasiparticle excitations of the semiconductor

$$[A_q, B_{1p}^+] = \frac{1}{V^{1/2}} \Psi(p-q/2) B_{2q-p},$$

$$[A_q, B_{2p}^+] = \frac{1}{V^{1/2}} \Psi(p-q/2) B_{1q-p}, \quad (4)$$

$$[A_q, A_p] = \delta_{p,q} + \frac{1}{V} \sum_{1,\sigma} \Psi^*(1) \Psi\left(\frac{q-p}{2} - 1\right) B_{\sigma-1+p/2} B_{\sigma-1+q-p/2} = \delta_{p,q} [1 + O(B^+B)], \quad (5)$$

where $O(B^+B)$ here and below denotes the concentration corrections determined by the population level of the exciton states.

The complete biexciton series $\{\Psi_j(1)\}$ of both the coupled states and the states of the continuous spectrum can be found together with the corresponding self-energies Ω_{pj}^{bix} from the Heisenberg equation for the operator A_{jq} [A_{jq} is the annihilation operator of state j of the biexciton \mathbf{q} determined by Eq. (3)]. This yields

$$\frac{\hbar^2 l^2}{M} \Psi_j(1) + \sum_{1'} W(1-1') \Psi_j(1')$$

$$= \hbar \left[\Omega_{jq}^{bix} - 2\omega_l - \frac{\hbar q^2}{4M} \right] \Psi_j(1) + O(B^+B), \quad (6)$$

which for the case of a weakly-excited semiconductor is naturally the Schrodinger equation reduced to the center of mass of the two excitons in the momentum representation with the given potential $W(1)$. It follows from the completeness condition of the wave functions $\{\Psi_j\}$

$$B_{11+q/2} B_{2-1+q/2} = \sum_j \frac{1}{V^{1/2}} \Psi_j^*(1) A_{jq}. \quad (7)$$

Taking this into account the initial Hamiltonian (2) can be reduced to

$$H_M = \sum_{p,\sigma} \left[\omega_p^{ex} B_{\sigma p} + B_{\sigma p} + \omega_p^{ph} \alpha_{\sigma p} + \alpha_{\sigma p} + i \frac{\Omega_c}{2} (\alpha_{\sigma p} + B_{\sigma p} - B_{\sigma p} + \alpha_{\sigma p}) \right] + \sum_{i,j} \varepsilon_{ij} A_{i1p} + A_{j1p} + \frac{1}{2} \sum_{p,1,q,\sigma} W_{\sigma\sigma}(\mathbf{q}) B_{\sigma p} + B_{\sigma 1} + B_{\sigma 1+q} B_{\sigma p-q}, \quad (8)$$

where the self-adjoint matrix ε_{ij} is determined by the expansion of the potential $W(1-1')$ in a complete set of biexciton wave eigenfunctions

$$\varepsilon_{ij} = \frac{1}{V} \sum_{1,1'} \Psi_j^*(1) W(1-1') \Psi_i(1'). \quad (9)$$

Certain qualitative conclusions can already be drawn from the form of Hamiltonian (8) near the fundamental biexciton resonance, when only a single term can remain in the sum over the states i, j with $i=j=1$. First, in contrast to the conventional approach,⁷⁻¹¹ $\varepsilon = \varepsilon_{11}$ is not equal to the unperturbed biexciton energy Ω_p^{biex} , but rather is determined by the average potential exciton interaction energy in the biexciton

$$\varepsilon = \Omega_p^{\text{biex}} - \frac{1}{V} \sum_i [\omega_{-i+p/2}^{\text{ex}} + \omega_{i+p/2}^{\text{ex}}] |\Psi(1)|^2, \quad (10)$$

where $\Psi(1) = \Psi_{i=1}(1)$, $\Omega_p^{\text{biex}} = \Omega_{i=1}^{\text{biex}}(p) = \Omega_0^{\text{biex}} + \hbar p^2/4M$. This result has a clear physical meaning and derives directly from the representation (3). Second, the term $V^{-1/2} M_2(\mathbf{p}, \mathbf{q}) A_p^+ B_{1q} B_{2p-q}$ used in this analysis is in fact already found in the Hamiltonian (8). At the same time Eq. (8) can be recast by retaining this term in its explicit form and altering the isolated term $\varepsilon_{11} A_{1p}^+ A_{1p} \equiv \varepsilon A_p^+ A_p$ accordingly.

Our approach is to analyze the properties of the exciton-photon-biexciton system of the semiconductor in the presence of a coherent polariton pump wave \mathbf{k} of frequency ω_k in the transparency region near the exciton absorption line. Such an intense wave will produce virtual electron-hole transitions, and therefore will drive the semiconductor to a strongly nonequilibrium state. Here, the pump wave, which for definiteness will be associated with the first type of excitons and photons, can be treated as a wave produced by an external source, while in our approximation its description is related to replacing the exciton and photon operators of the particular mode \mathbf{k} with C -numbers: $B_{1k} \rightarrow V^{1/2} P_0 \times \exp(-i\omega_k t)$; $\alpha_{1k} \rightarrow V^{1/2} E_0 \exp(-\omega_k t)$ which in fact represent the exciton and photon components of the polariton pump wave.

The nonequilibrium state in which the semiconductor ends up generally will make it impossible to treat the exciton and biexciton excitations as independent. This derives directly from the form of the commutators (4), (5) for the case where the exciton creation-destruction operators on the right side of these relations operate on the states of mode \mathbf{k} . In this case, even in the thermodynamic limit $V \rightarrow \infty$, finite corrections arise in the commutation relations that do not permit treating the excitons or biexcitons as independent boson excitations. In our approach this difficulty is easily overcome if the mode k is explicitly distinguished in the definition (3) of the biexciton operators as well as in the Hamiltonian (8); this mode can formally be regarded as macropopulated:

$$A_{1q} = P_0 \Psi_i(\mathbf{k} - \mathbf{q}/2) e^{-i\omega_k t} B_{2q-k} + \tilde{A}_{1q}, \quad (11)$$

$$\tilde{H} = \tilde{H}_1 + \tilde{H}_2,$$

$$\begin{aligned} \tilde{H}_1 = \sum_{\mathbf{p} \neq \mathbf{k}}' \left\{ (\omega_p^{\text{ph}} - \omega_k) \alpha_p^+ \alpha_p + [\omega_p^{\text{ex}} + |P_0|^2 V [W_{11}(0) \right. \\ \left. + W_{11}(\mathbf{p} - \mathbf{k})] B_{1p}^+ B_{1p} \right. \\ \left. + \left[i \frac{\Omega_c}{2} \alpha_{1p}^+ B_{1p} + \text{h.c.} \right] \right\} + \sum_{\mathbf{p}, \mathbf{1}, \mathbf{q} (\neq \mathbf{k})}' W_{11}(\mathbf{q}) B_{1p}^+ B_{11}^+ B_{11+q} B_{1p-q}, \end{aligned}$$

$$\begin{aligned} \tilde{H}_2 = \sum_{\mathbf{p}, \mathbf{1}, \mathbf{j}}' \left\{ \omega_p^{\text{ph}} \alpha_{2p}^+ \alpha_{2p} + \varepsilon_{ij} \tilde{A}_{1p+k}^+ \tilde{A}_{jp+k} \right. \\ \left. + \left[\omega_p^{\text{ex}} + \varepsilon_{ij} |P_0|^2 \Psi_i^* \left(\frac{\mathbf{p} - \mathbf{k}}{2} \right) \right. \right. \end{aligned}$$

$$\begin{aligned} \left. \times \Psi_j \left(\frac{\mathbf{p} - \mathbf{k}}{2} \right) \right] B_{2p}^+ B_{2p} \\ \left. + \left[i \frac{\Omega_c}{2} \alpha_{2p}^+ B_{2p} + \varepsilon_{ij} P_0 e^{-i\omega_k t} \Psi_j \left(\frac{\mathbf{p} - \mathbf{k}}{2} \right) \right. \right. \\ \left. \left. \times \tilde{A}_{1p+k}^+ B_{2p} + \text{h.c.} \right] \right\} + \sum_{\mathbf{p}, \mathbf{1}, \mathbf{q}} W_{22}(\mathbf{q}) B_{2p}^+ B_{21}^+ B_{21+q} B_{2p-q}. \quad (12) \end{aligned}$$

In fact such an explicit procedure for distinguishing the mode \mathbf{k} is nothing other than the construction of a new vacuum state of the semiconductor in the presence of the polariton pump. And if the vacuum state can be formally obtained in such an approach by the ordinary Glauber canonical transform that incorporates the polariton pump wave source in mode \mathbf{k} , the procedure for formulating the new vacuum state in the general case examined below will be more complicated.

It is important to emphasize that this introduction of a new vacuum state and the formulation of independent exciton and biexciton excitations are rigorous due specifically to the virtual nature of electron-hole pair creation by the pump wave, when the actual population processes of the electron-hole levels are suppressed. In this case the latter terms in the expressions for \tilde{H}_1 and \tilde{H}_2 function as concentration corrections in the new vacuum state, and can be neglected in what follows.

It follows from general Eq. (12) that the complete Hamiltonian \tilde{H} consists of two parts. Its first component \tilde{H}_1 describes the behavior of the photons and excitons of the first type in the presence of a polariton pump wave of the same type. The second part \tilde{H}_2 characterizes under an identical condition the second type of exciton-photons and the biexcitons. Using the canonical transformation

$$S = \exp \left[i\omega_k t \sum_{\mathbf{p}, \sigma} (\alpha_{\sigma p}^+ \alpha_{\sigma p} + B_{\sigma p}^+ B_{\sigma p}) \right],$$

which introduces explicit time dependence in the Hamiltonian (12), and summing over the indices i and j of the biexciton series in the third term in the expression for \tilde{H}_2 , we obtain

$$\begin{aligned} \tilde{H}_2 = \sum_{\mathbf{p}, \mathbf{1}, \mathbf{j}}' \left\{ (\omega_p^{\text{ph}} - \omega_k) \alpha_p^+ \alpha_p + \varepsilon_{ij} \tilde{A}_{1p+k}^+ \tilde{A}_{jp+k} \right. \\ \left. + [\omega_p^{\text{ex}} + |P_0|^2 V W(0) - \omega_k] B_p^+ B_p \right. \\ \left. + \left[i \frac{\Omega_c}{2} \alpha_p^+ B_p + \varepsilon_{ij} P_0 \Psi_j \left(\frac{\mathbf{p} - \mathbf{k}}{2} \right) \tilde{A}_{1p+k}^+ B_p + \text{h.c.} \right] \right\}, \quad (13) \end{aligned}$$

where the index 2 on the excitons and photons of the second type will be dropped in this section.

At this point in the analysis it is appropriate to consider diagonalization of the quadratic form (12) subject to Eq. (13) to find the true spectrum of the exciton-photon-biexciton excitations of the semiconductor in the presence of the polariton pump field. Such diagonalization in the language of the diagram technique corresponds to an exact solution of the diagram equations shown in Fig. 2 relative to the one-particle and two-particle exciton Green's functions determined in the vacuum state of the semiconductor with the particular macropopulated mode \mathbf{k} . In these graph equations the solid and wavy lines denote the exciton and photon propagators, respectively; the dashed line represents the ex-

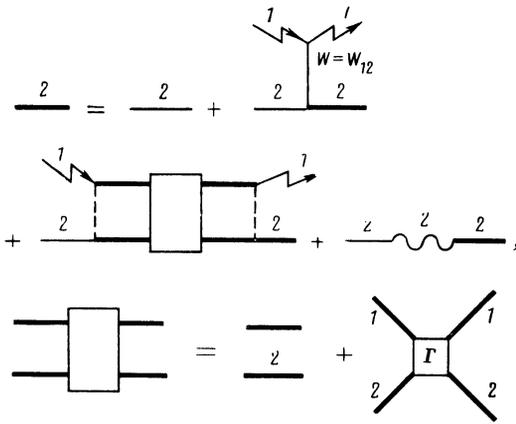


FIG. 2.

citon-exciton interaction potential W , and the arrows denote the exciton component P_0 or P_0^* of the pump wave; the square represents the biexciton vertex function Γ . We believe that the exciton resonance shift δ_p^{ex} in the presence of the pump wave is of primary interest; as follows from Eq. (13), this shift is given by

$$\delta_p^{ex} = |P_0|^2 V W(0) \quad (14)$$

and is nothing more or less than a way of accounting for the second diagram in the exciton diagram equation. Such a level shift of the excitons of the second type is a red-shift for the characteristic exciton momenta $p < a_{\text{biex}}^{-1}$ and in fact is due to the attraction of the test exciton of the second type \mathbf{p} to excitons of the first type \mathbf{k} of the pump wave.

If the analysis is limited to the biexciton ground level, in this approximation the Hamiltonian (12) is substantially simplified:

$$\begin{aligned} \bar{H}_2 = & \sum_{\mathbf{p}} \left\{ (\omega_{\mathbf{p}}^{ph} - \omega_{\mathbf{k}}) \alpha_{\mathbf{p}}^+ \alpha_{\mathbf{p}} + \varepsilon \bar{A}_{\mathbf{p}+\mathbf{k}}^+ \bar{A}_{\mathbf{p}+\mathbf{k}} \right. \\ & + \left[\omega_{\mathbf{p}}^{ex} + |P_0|^2 \varepsilon \left| \Psi \left(\frac{\mathbf{p}-\mathbf{k}}{2} \right) \right|^2 - \omega_{\mathbf{k}} \right] B_{\mathbf{p}}^+ B_{\mathbf{p}} \\ & \left. + \left[i \frac{\Omega_c}{2} \alpha_{\mathbf{p}}^+ B_{\mathbf{p}} + \varepsilon P_0 \Psi \left(\frac{\mathbf{p}-\mathbf{k}}{2} \right) \bar{A}_{\mathbf{p}+\mathbf{k}}^+ B_{\mathbf{p}} + \text{h.c.} \right] \right\}, \quad (15) \end{aligned}$$

where the parameter ε is determined by Eq. (10). Taking this approximation into account we naturally proceed to explicit diagonalization of Hamiltonian (12) which is quadratic in the independent boson operators $\alpha_{\mathbf{p}}$, $B_{\mathbf{p}}$ and $A_{\mathbf{p}+\mathbf{k}}$. Such diagonalization employs a canonical transformation and also involves introducing new elementary excitations of the semiconductor $\alpha_{\mathbf{p}}'$, $B_{\mathbf{p}}'$ and $A_{\mathbf{p}+\mathbf{k}}'$ consisting of photon, exciton and biexciton components:

$$\begin{pmatrix} \alpha_{\mathbf{p}}' \\ B_{\mathbf{p}}' \\ A_{\mathbf{p}+\mathbf{k}}' \end{pmatrix} = (C_{ij}) \begin{pmatrix} \alpha_{\mathbf{p}} \\ B_{\mathbf{p}} \\ \bar{A}_{\mathbf{p}+\mathbf{k}} \end{pmatrix}. \quad (16)$$

The transformation to diagonal form denotes in the familiar sense an exact treatment of both exciton-phonon and exciton-exciton interactions in this approximation. The corresponding new dispersion equation takes the form

$$\begin{aligned} v_1 v_2 v_3 - \frac{\Omega_c^2}{4} v_3 - |m|^2 v_1 &= 0, \quad (17) \\ v_1 &= \omega_{\mathbf{p}}^{ph} - \omega_{\mathbf{k}} - \omega, \quad v_2 = \omega_{\mathbf{p}}^{ex} + \delta_{\mathbf{p}}^{ex} - \omega_{\mathbf{k}} - \omega, \\ v_3 &= \Omega_{\mathbf{p}+\mathbf{k}}^{\text{biex}} + \delta_{\mathbf{p}+\mathbf{k}}^{\text{biex}} - 2\omega_{\mathbf{k}} - \omega \end{aligned}$$

and reflects the nature of unification with subsequent splitting of the exciton-photon-biexciton terms of the semiconductor in the presence of the polariton pump wave. The exciton $\delta_{\mathbf{p}}^{ex}$ and biexciton $\delta_{\mathbf{p}+\mathbf{k}}^{\text{biex}}$ shifts in Eq. (17) together with the matrix element m are determined by

$$\delta_{\mathbf{p}}^{ex} = \varepsilon |P_0|^2 \left| \Psi \left(\frac{\mathbf{p}-\mathbf{k}}{2} \right) \right|^2, \quad (18)$$

$$\begin{aligned} \delta_{\mathbf{p}+\mathbf{k}}^{\text{biex}} = & |P_0|^2 \left[V W(0) + V W_{11}(0) \right. \\ & \left. + \sum_1 W_{11} \left(1 + \frac{\mathbf{p}-\mathbf{k}}{2} \right) \left| \Psi(1) \right|^2 \right], \quad (19) \end{aligned}$$

$$m = \varepsilon P_0 \Psi \left(\frac{\mathbf{p}-\mathbf{k}}{2} \right). \quad (20)$$

The dispersion equation (17) has three branches of the solutions $\omega = \omega_i(\mathbf{p})$, $i = 1, 2, 3$ while the corresponding weight multipliers χ_i characterizing the specific contribution of the photon, exciton and biexciton components to the new elementary excitations, are given by

$$\chi_i^{ph} = |C_{i1}|^2 = \frac{v_3^2 \Omega_c^2 / 4}{v_1^2 v_3^2 + v_3^2 \Omega_c^2 / 4 + |m|^2 v_1^2}, \quad (21)$$

$$\chi_i^{ex} = |C_{i2}|^2 = \frac{v_1^2 v_3^2}{v_1^2 v_3^2 + v_3^2 \Omega_c^2 / 4 + |m|^2 v_1^2},$$

$$\chi_i^{\text{biex}} = |C_{i3}|^2 = 1 - |C_{i1}|^2 - |C_{i2}|^2 = \frac{|m|^2 v_1^2}{v_1^2 v_3^2 + v_3^2 \Omega_c^2 / 4 + |m|^2 v_1^2}.$$

Unlike the exact Eq. (14) for the exciton level shift, the approximate relation (18) contains the biexciton wave function of the ground state and the biexciton potential ε . As follows from Eq. (9) this corresponds to an approximation near the biexciton level $n = 1$ (see Fig. 1) of the exact potential W of its expansion in eigenfunctions $\{\Psi_j\}$ and retention of the principal term.

The dispersion equation (17) obtained here is essentially different from the equation used previously.⁷⁻¹¹ Above all we emphasize that the spectral modification of the elementary excitations of the semiconductor in the presence of a polariton pump wave examined here is not determined by the "giant" oscillator strength $M_1(\mathbf{p}, \mathbf{q})$ of the exciton-biexciton transition, but rather solely by the matrix element $M_2(\mathbf{p}, \mathbf{q})$ characterizing direct resonance coupling of two different excitons to form a biexciton. Such coupling of the excitons forming a biexciton in turn is determined by the potential W which may be due to the presence in the system of the third quasiparticles, photons, which resonate with the excitons. In other words the polariton effect itself may be responsible for satisfaction of the law of conservation of energy in the resonance pairing of excitons that forms a biexciton.

The most interesting new result is the dynamic shifts $\delta_{\mathbf{p}}^{ex}$ and $\delta_{\mathbf{p}+\mathbf{k}}^{\text{biex}}$ of the exciton and biexciton levels which are dependent on pump wave intensity. This effect is also related to Coulomb-like direct exciton-exciton interaction. Indeed, the dynamic red-shift of the exciton level given by Eq. (18) has the following origin. An exciton of the second type \mathbf{p} produced, for example, by an electromagnetic probe wave, is attracted to excitons of the first type of the \mathbf{k} -pump wave, which results in virtual formation of biexcitons. The effectiveness of this interaction is characterized by the corre-

sponding biexciton potential $\varepsilon < 0$, while $|\Psi((\mathbf{p} - \mathbf{k})/2)|^4$ determines the probability of virtual formation of the biexciton $\mathbf{p} + \mathbf{k}$ from the \mathbf{p} and \mathbf{k} excitons. Regarding the biexciton shift $\delta_{\mathbf{p}+\mathbf{k}}^{\text{biex}}$, evidently this is a blue shift consistent with Eq. (19).

This result represents direct evidence that the blue shift $\delta_{11}^{\text{ex}} \approx 2|P_0|^2 V W_{11}(0)$ of the level of the first type of exciton comprising the biexciton in the presence of the pump wave is more significant than the red-shift δ^{ex} of the second type of exciton. Here any further stabilization of the exciton-photon-biexciton system is associated with the absence of exciton complexes that are more complicated than the biexciton. It is generally well-known^{20,23} that interexciton repulsion predominates in a uniformly excited system in thermodynamic equilibrium. However this case of effective exciton attraction can be implemented in a "probe radiation-pump wave" experiment. Such an optical experiment can be formulated for a CdS semiconductor in a geometry with $\mathbf{p} \parallel \mathbf{k} \parallel \mathbf{c}$ using a pump wave and probe radiation of opposite circular polarizations.

Finally we emphasize one additional significant element. The dispersion equation (17) reflects the fact that all three components of the new excitations—the photon, exciton and biexciton components—are in a certain sense equivalent. In other words, the new spectrum cannot be treated as a modification of the polariton or biexciton spectra,⁷ but rather it is necessary to uniformly treat all three components of the initial excitations as constituent elements.

Regarding the polariton pump wave itself, a self-consistent description of this wave is given by

$$\begin{aligned} i \frac{\partial}{\partial t} \mathbf{E}_0 &= (\omega_{\mathbf{k}^{ph}} - \omega_{\mathbf{k}}) \mathbf{E}_0 + i \frac{\Omega_c}{2} \mathbf{P}_0, \\ i \frac{\partial}{\partial t} \mathbf{P}_0 &= (\omega_{\mathbf{k}^{\text{ex}}} - \omega_{\mathbf{k}}) \mathbf{P}_0 - i \frac{\Omega_c}{2} \mathbf{E}_0 + W_{11}(0) V |P_0|^2 \mathbf{P}_0. \end{aligned} \quad (22)$$

The carrier frequency $\omega_{\mathbf{k}}$ of the pump wave is established by an external source, in accordance with the actual experimental situation. Equations (22) in many respects resemble the equations obtained in Ref. 1 and reflect both the polariton character of pump wave propagation and its nonlinear self-action, which is manifested as a blue-shift of the exciton level due to the interaction W_{11} of like excitons. We note that the exciton \mathbf{P}_0 and photon \mathbf{E}_0 components are normalized to the concentration of the corresponding excitations, consistent with the method used to isolate the macropopulated mode \mathbf{k} . Thus, for example, $N = |P_0|^2 \sim I$ is the exciton concentration of the pump wave.

3. THE EXCITON-BIEXCITON STARK-EFFECT IN THE ELECTRON-HOLE APPROXIMATION

In this section we formulate a consistent microscopic theory of the exciton-biexciton optical Stark-effect based on an analysis of the electron-hole Hamiltonian of the semiconductor. This approach is required by the lack of a precise understanding of the exciton-exciton interaction potential $W(\mathbf{l})$. The corresponding initial microscopic Hamiltonian describing the Coulomb interaction of the electrons and holes in the semiconductor and the electromagnetic field generating the band-to-band electron transitions takes the form

$$\begin{aligned} H_m &= \sum_{\mathbf{p}} [\omega_c(\mathbf{p}) a_{\mathbf{p}}^+ a_{\mathbf{p}} + \omega_v(\mathbf{p}) b_{\mathbf{p}}^+ b_{\mathbf{p}} + \omega_{\mathbf{p}^h}(\mathbf{p}) \alpha_{\mathbf{p}}^+ \alpha_{\mathbf{p}}] \\ &+ \frac{1}{2} \sum_{\mathbf{p}, \mathbf{l}, \mathbf{q}} V_{\mathbf{q}} [a_{\mathbf{p}}^+ a_{\mathbf{l}+\mathbf{q}} a_{\mathbf{p}-\mathbf{q}} + b_{\mathbf{p}}^+ b_{\mathbf{l}+\mathbf{q}} b_{\mathbf{p}-\mathbf{q}} - 2a_{\mathbf{p}}^+ b_{\mathbf{l}+\mathbf{q}} a_{\mathbf{p}-\mathbf{q}}] \\ &+ \sum_{\mathbf{p}, \mathbf{l}} \{ [\beta_1(\mathbf{p}) \alpha_{\mathbf{l}} a_{\mathbf{p}+\mathbf{l}}^+ b_{-\mathbf{p}}^+ + \beta_1^*(\mathbf{p}) \alpha_{\mathbf{l}}^+ b_{-\mathbf{p}} a_{\mathbf{p}+\mathbf{l}}] \}, \end{aligned} \quad (23)$$

where $\omega_c(\mathbf{p}) = E_g + \hbar^2 p^2 / 2m_c$ and $\omega_v(\mathbf{p}) = \hbar^2 p^2 / 2m_c$ are the electron energy (operator $a_{\mathbf{p}}$) and hole energy (operator $b_{\mathbf{p}}$), respectively; E_g is the bandgap of the semiconductor; and $V_{\mathbf{q}} = 4\pi e^2 / V \varepsilon_g q^2$ is the Fourier transform of the Coulomb potential. In turn the matrix element $\beta_1(\mathbf{p})$ is determined by

$$\beta_1(\mathbf{p}) = \left(-\frac{e}{mc} \right) \langle \tilde{u}_{\mathbf{p}+\mathbf{l}, c} | \hat{\mathbf{p}} | \tilde{u}_{\mathbf{p}, v} \rangle \left(\frac{2\pi c \hbar}{V l} \right)^{1/2}, \quad (24)$$

where $\tilde{u}_{\mathbf{p}, c}$, $\tilde{u}_{\mathbf{p}, v}$ are the Bloch electron functions in the conduction and valence bands, respectively and $\hat{\mathbf{p}}$ is the momentum operator. The exciton and biexciton destruction operators are introduced as

$$B_{\mathbf{q}} = \frac{1}{V^{1/2}} \sum_{\mathbf{l}} \varphi(\mathbf{l}) a_{\mathbf{l}+\mathbf{q}} b_{-\mathbf{l}+\mathbf{q}}, \quad (25)$$

$$\begin{aligned} A_{\mathbf{q}} &= \frac{1}{4V^{1/2}} \sum_{\mathbf{p}, \mathbf{l}, \mathbf{s}} \Gamma(\mathbf{p}, \mathbf{l}, \mathbf{s}) a_{\mathbf{l}+\alpha(\mathbf{p}+\mathbf{q}/2)} b_{-\mathbf{l}+\beta(\mathbf{p}+\mathbf{q}/2)} \\ &\quad \times a_{\mathbf{s}+\alpha(-\mathbf{p}+\mathbf{q}/2)} b_{-\mathbf{s}+\beta(-\mathbf{p}+\mathbf{q}/2)}, \end{aligned} \quad (26)$$

where $\varphi(\mathbf{l})$ and $\Gamma(\mathbf{p}, \mathbf{l}, \mathbf{s})$ are the wave functions of the exciton and biexciton normalized to unity, respectively; $\alpha = m_c / (m_c + m_e)$, $\beta = m_v / (m_c + m_v)$. Regarding the spin structure of these elementary excitations we consider, as in the preceding section, exclusively the dipole-active excitons with total spin $S = 0$ and biexcitons for which the spins of the electrons and the holes cancel. Accounting for the simple relationship between the results obtained below and the relations examined previously, we will not write out the indices 1, 2 on the different exciton or photon operators nor the spin indices.

The commutation relations analogous to the commutators (4), (5) examined in the exciton quasiparticle approximation take the form

$$\begin{aligned} [A_{\mathbf{q}}, B_{\mathbf{p}}^+] &= \frac{1}{V^2} \sum_{\mathbf{l}, \mathbf{s}} \Gamma(-\mathbf{p}+\mathbf{q}/2, \mathbf{l}, \mathbf{s}) \varphi^*(\mathbf{s}) a_{\mathbf{l}+\alpha(\mathbf{q}-\mathbf{p})} b_{-\mathbf{l}+\beta(\mathbf{q}-\mathbf{p})}, \\ [A_{\mathbf{q}}, A_{\mathbf{p}}^+] &= \delta_{\mathbf{q}, \mathbf{p}} [1 + O(a^+ a) + O(b^+ b)], \end{aligned} \quad (27)$$

where the following properties of the biexciton wave function were used in deriving Eq. (28):

$$\begin{aligned} \Gamma(\mathbf{p}, \mathbf{l}, \mathbf{s}) &= \Gamma(-\mathbf{p}, \mathbf{s}, \mathbf{l}) = -\Gamma(\mathbf{s} - \mathbf{l} + \beta \mathbf{p} - \alpha \mathbf{p}, \\ &\beta \mathbf{s} + \alpha \mathbf{l} - 2\alpha \beta \mathbf{p}, \quad \alpha \mathbf{s} + \beta \mathbf{l} + 2\alpha \beta \mathbf{p}) \\ &= -\Gamma(-\mathbf{s} + \mathbf{l} - \beta \mathbf{p} + \alpha \mathbf{p}, \beta \mathbf{l} + \alpha \mathbf{s} + 2\alpha \beta \mathbf{p}, \beta \mathbf{s} + \alpha \mathbf{l} - 2\alpha \beta \mathbf{p}), \end{aligned} \quad (29)$$

which reflect the antisymmetry properties of the total function $\Gamma(\mathbf{p}, \mathbf{l}, \mathbf{s})$ with respect to permutation of electrons or holes. It is possible to find the corresponding Schrödinger equation for the wave function $\Gamma(\mathbf{p}, \mathbf{l}, \mathbf{s})$ of a weakly-excited semiconductor based on the equation of motion for the biexciton operator $A_{\mathbf{q}}$ (26) obtained by means of Hamiltonian (23):

$$\begin{aligned}
& [\omega_c(s-\alpha p+\alpha q/2)+\omega_c(1+\alpha p+\alpha q/2)+\omega_v(-s-\beta p+\beta q/2) \\
& +\omega_v(-1+\beta p+\beta q/2)]\Gamma(\mathbf{p}, \mathbf{l}, \mathbf{s}) \\
& \sum_r \{V_r[\Gamma(\mathbf{p}, \mathbf{l}, \mathbf{s}+\mathbf{r})+\Gamma(\mathbf{p}, \mathbf{l}+\mathbf{r}, \mathbf{s})] \\
& +V_r[\Gamma(\mathbf{p}-\mathbf{r}, \mathbf{l}+\alpha \mathbf{r}, \mathbf{s}-\alpha \mathbf{r})+\Gamma(\mathbf{p}-\mathbf{r}, \mathbf{l}-\beta \mathbf{r}, \mathbf{s}+\beta \mathbf{r}) \\
& -\Gamma(\mathbf{p}-\mathbf{r}, \mathbf{l}+\alpha \mathbf{r}, \mathbf{s}+\beta \mathbf{r}) \\
& -\Gamma(\mathbf{p}-\mathbf{r}, \mathbf{l}-\beta \mathbf{r}, \mathbf{s}-\alpha \mathbf{r})]\}=\Omega_q^{biox}\Gamma(\mathbf{p}, \mathbf{l}, \mathbf{s}). \quad (30)
\end{aligned}$$

The terms under the summation sign can be arbitrarily grouped into two parts by brackets. In this case, the first term will characterize the potential self-energy of each of the two excitons comprising the biexciton, while the second will correspond to exciton-exciton interaction. It is precisely this second term that will determine the dynamical shift of the exciton level in the presence of the pump wave.

The polariton pump wave \mathbf{k} is formally introduced to this photon-electron-hole system of the semiconductor by adding the Glauber macroscopic photon source to the initial Hamiltonian (23):

$$H_s=\omega_k^{ph}[V^{1/2}\exp(i\omega_k t)E_0^*\alpha_k+h.c.], \quad (31)$$

which henceforth will be examined self-consistently by relating the generated classical electromagnetic field $E_0 \exp(-i\omega_k t)$ to the photon component of the pump wave. The new vacuum state of the semiconductor in the presence of the pump wave can be introduced by means of two successive canonical transformations of the Hamiltonian (23). The first such transformation is an ordinary Glauber transformation which eliminates the photon source (31). The second transformation, analogous to that proposed in Ref. 20, is implemented by the operator

$$S=\exp\left\{\sum_p \varphi_p[a_{p+k}^+b_{-p}^+-b_{-p}a_{p+k}]\right\}, \quad (32)$$

which results in the following substitution of electron-hole operators:

$$\begin{aligned}
a_{p+k} &\rightarrow Sa_{p+k}S^+=u_p a_{p+k}+v_p b_{-p}^+, \\
b_{-p} &\rightarrow Sb_{-p}S^+=u_p b_{-p}-v_p a_{p+k}^+, \quad (33)
\end{aligned}$$

where $u_p=\cos\varphi_p$, $v_p=\sin\varphi_p$, $u_p^2+v_p^2=1$. After these operations, including a time transformation, the Hamiltonian takes the form

$$H_m=H_0+H_e+H_h+H_{ph}+H_{vac}+H_{int}+H_{e-h}, \quad (34)$$

$$\begin{aligned}
H_0 &= \sum_p \left\{ [\omega_c(\mathbf{p}+\mathbf{k})+\omega_v(\mathbf{p})-\omega_k]v_p^2+[\beta_k(\mathbf{p})E_0+\beta_k^*(\mathbf{p})E_0^*]u_p v_p \right. \\
& \left. -\sum_1 V_{p-1}[v_p^2 v_1^2+u_p u_1 v_p v_1] \right\} + [\omega_k^{ph}-\omega_k]|E_0|^2, \quad (35)
\end{aligned}$$

$$\begin{aligned}
H_e &= \sum_p \left\{ [\omega_c(\mathbf{p}+\mathbf{k})-\omega_k]u_p^2-\omega_v(\mathbf{p})v_p^2 \right. \\
& \left. -[\beta_k(\mathbf{p})E_0+\beta_k^*(\mathbf{p})E_0^*]u_p v_p \right. \\
& \left. +\sum_1 V_{p-1}[2u_p u_1 v_p v_1-u_p^2 v_1^2+v_p^2 v_1^2] \right\} a_{p+k}^+ a_{p+k}, \quad (36)
\end{aligned}$$

$$\begin{aligned}
H_h &= \sum_p \left\{ \omega_c(\mathbf{p})u_p^2-[\omega_c(\mathbf{p}+\mathbf{k})-\omega_k]v_p^2 \right. \\
& \left. -[\beta_k(\mathbf{p})E_0+\beta_k^*(\mathbf{p})E_0^*]u_p v_p \right. \\
& \left. +\sum_1 V_{p-1}[2u_p u_1 v_p v_1-u_p^2 v_1^2+v_p^2 v_1^2] \right\} b_{-p} b_{-p}, \quad (37)
\end{aligned}$$

$$H_{ph}=\sum_p [\dot{\omega}_p^{ph}-\omega_k]\alpha_p^+\alpha_p, \quad (38)$$

$$H_{vac}=\sum_p \{L(\mathbf{p})a_{p+k}^+b_{-p}^++L^*(\mathbf{p})b_{-p}a_{p+k}\}, \quad (39)$$

where

$$\begin{aligned}
L(\mathbf{p}) &= [\omega_c(\mathbf{p}+\mathbf{k})+\omega_v(\mathbf{p})-\omega_k]u_p v_p+\beta_k(\mathbf{p})E_0 u_p^2-\beta_k^*(\mathbf{p})E_0^* v_p^2 \\
& +\sum_1 V_{p-1}[u_1 v_1 v_p^2-u_p^2 u_1 v_1-2u_p v_p v_1^2],
\end{aligned}$$

$$\begin{aligned}
H_{int} &= \sum_{p,1} \{ \alpha_1 \beta_1(\mathbf{p}) [u_{p-k+1} u_p a_{p+1}^+ b_{-p}^+ - v_{p-k+1} v_p b_{-p-1+k} a_{1+k} \\
& - u_{p-k+1} v_p a_{p+1}^+ a_{p+k} + u_p v_{p-k+1} b_{-p-1+k} b_{-p}^+] + h.c. \}, \quad (40)
\end{aligned}$$

$$\begin{aligned}
H_{e-h} &= \sum_{p,1,s} \left\{ \frac{1}{2} \bar{V}(\mathbf{p}, \mathbf{l}, \mathbf{s}) [a_{p+k}^+ a_{1+k}^+ a_{1+k+s} a_{p+k-s} \right. \\
& \left. + b_{-p}^+ b_{-1}^+ b_{-1-s} b_{-p+s} \right. \\
& \left. - 2a_{p+k}^+ b_{-1-s}^+ b_{-1} a_{p+k-s} \right. \\
& \left. + \frac{1}{2} \bar{W}(\mathbf{p}, \mathbf{l}, \mathbf{s}) [a_{p+k}^+ a_{1+k}^+ b_{-1-s}^+ b_{-p+s}^+ + a_{p+k} a_{1+k} b_{-1-s} b_{-p+s} \right. \\
& \left. - 2a_{p+k}^+ b_{-p+s}^+ b_{-1} a_{1+k+s} \right. \\
& \left. + \bar{U}(\mathbf{p}, \mathbf{l}, \mathbf{s}) [a_{p+k}^+ a_{1+k}^+ a_{1+k+s} b_{-p+s}^+ + b_{-p+s} a_{1+k+s} a_{1+k} a_{p+k} \right. \\
& \left. + a_{p+k}^+ b_{-p}^+ b_{-1}^+ b_{-1-s}^+ + b_{-1-s}^+ b_{-1} b_{-p} a_{p+k-s} \right\}, \quad (41)
\end{aligned}$$

where

$$\begin{aligned}
\bar{V}(\mathbf{p}, \mathbf{l}, \mathbf{s}) &= V_s [u_p u_1 u_{1+s} u_{p-s} + v_p v_1 v_{1+s} v_{p-s} \\
& + u_p u_{p-s} v_1 v_{1+s} + u_1 u_{1+s} v_p v_{p-s}], \quad (42a)
\end{aligned}$$

$$\begin{aligned}
\bar{W}(\mathbf{p}, \mathbf{l}, \mathbf{s}) &= V_s [u_p u_1 v_{1+s} v_{p-s} + u_{p-s} u_{1+s} v_p v_1 \\
& - u_p u_{1+s} v_1 v_{p-s} - u_{p-s} u_1 v_p v_{1+s}], \quad (42b)
\end{aligned}$$

$$\begin{aligned}
\bar{U}(\mathbf{p}, \mathbf{l}, \mathbf{s}) &= V_s [u_p u_1 u_{1+s} v_{p-s} + u_p v_{p-s} v_1 v_{1+s} \\
& - u_1 u_{p-s} u_{1+s} v_p - u_{p-s} v_p v_1 v_{1+s}]. \quad (42c)
\end{aligned}$$

The remaining unknown parameter φ_p of the transformation (32) is determined by the stability requirement of the new vacuum state relative to creation of the hole pair electron. This condition in fact results in the requirement $L(\mathbf{p})=0$ in Eq. (39) for H_{vac} . As we shall demonstrate below the parameter φ_p is determined by $|P_0|^2 a_{ex}^3 \ll 1$, which validates approximate relations ($\varphi_p \ll 1$)

$$u_p=\varphi_p; \quad u_p=1-\varphi_p^2/2. \quad (43)$$

In order to determine the parameter φ_p we therefore obtain the following equation:

$$[\omega_c(\mathbf{p}+\mathbf{k})+\omega_v(-\mathbf{p})-\omega_k]\varphi_p-\sum_1 V_{p-1}\varphi_1=-\beta_k(\mathbf{p})V^{1/2}E_0. \quad (44)$$

Accounting for the conditions of this problem, we can approximate the isolated exciton level $n=1$ in Eq. (44). Specifically, in this case the following representation is valid:

$$\varphi_{p-\beta k}=V^{1/2}\varphi(\mathbf{p})P_0, \quad (45)$$

where the exciton wave function $\varphi(\mathbf{p})$, as follows from Eq. (44), is determined by the Schrödinger equation

$$\frac{\hbar^2 p^2}{2\mu}\varphi(\mathbf{p})-\sum_1 V_{p-1}\varphi(\mathbf{l})=\hbar[\omega_k^{ex}-E_g-\hbar^2 k^2/2M]\varphi(\mathbf{p}), \quad (46)$$

$$M=m_c+m_v; \quad \mu=m_c m_v/(m_c+m_v).$$

The parameter P_0 in fact represents the exciton polarization of the pump wave and is related to E_0 by Eq. (47), which can be obtained from Eq. (44):

$$(\omega_k^{ex} - \omega_k) P_0 = i \frac{\Omega_c}{2} E_0, \quad (47)$$

where the polariton parameter Ω_c is given by

$$\Omega_c = -i \sum_p \beta_k (\mathbf{p} - \beta \mathbf{k}) \varphi(\mathbf{p}) \approx \left(\frac{e}{2m} \right) \frac{\hbar^{1/2} p_{cv}}{\pi^3 (2\omega_i)^{1/2} a_{ex}^{3/2}},$$

$$p_{cv} = i \langle \tilde{u}_{\mathbf{p}+\alpha\mathbf{k},c} | \hat{p} | \tilde{u}_{\mathbf{p}-\beta\mathbf{k},v} \rangle. \quad (48)$$

It is evident that Eq. (47) directly corresponds to the second equation of the previous system (22) in a linear approximation. The source H_s of the electromagnetic wave E_0 introduced in the initial Hamiltonian (23) is self-consistently related to the exciton polarization P_0 by means of the first equation of the polariton system (22).

After determining the parameter φ_p , the subsequent analysis involves formulating new exciton and biexciton quasiparticle excitations by means of Eqs. (25), (26) where, however, it is necessary to use new exciton and hole operators. It is this approach that makes it possible to treat the excitons and biexcitons of the semiconductor in the presence of the polariton pump wave as independent boson excitations: $[A_q, B_p^+] = 0$, $[A_q, A_p^+] = \delta_{q,p}$, i.e., to neglect the concentration corrections.

Using the approximation (43) in conjunction with the transformed Hamiltonian \tilde{H}_m , it is possible to find the corresponding equations of motion for the photon operator α_p and the new exciton for the biexciton operators B_p and A_{p+k} . The corresponding dispersion equation for the intrinsic exciton-photon-biexciton excitations of the semiconductor in the presence of the pump wave can be obtained in turn from a closed system of these equations. This dispersion equation is entirely analogous to Eq. (17), which was previously considered in the exciton quasiparticle approximation. In this case some rather involved calculations make it possible to express (for the case $|P_0|^2 a_{biex}^3 \ll 1$) the dynamical shift δ_p^{ex} and the matrix element m , previously determined by Eqs. (18) and (20), respectively, through the microscopic characteristics of the electron-hole system of the semiconductor:

$$\delta_p^{ex} = \frac{|P_0|^2}{V^4} \sum_{p,l,q,s} \Gamma \left(\frac{\mathbf{k}-\mathbf{q}}{2}, \mathbf{l}, \mathbf{s} \right) \varphi(\mathbf{l}) \varphi(\mathbf{s})$$

$$\times [\Omega_{q+k}^{biex} - \omega_q^{ex} - \omega_k^{ex}] \Gamma \left(\frac{\mathbf{q}-\mathbf{k}}{2}, \mathbf{q}, \mathbf{p} \right) \varphi(\mathbf{q}) \varphi(\mathbf{p}); \quad (49)$$

$$m = P_0 \frac{1}{V^3} \sum_{l,s,r,x,y} \Gamma \left(\frac{\mathbf{p}-\mathbf{k}}{2}, \mathbf{l}, \mathbf{s} \right) \varphi(\mathbf{l}) \varphi(\mathbf{s}) [\Omega_{p+k}^{biex} - \omega_{r+k}^{ex} - \omega_{-r+k}^{ex}]$$

$$\times \Gamma \left(\mathbf{r} + \frac{\mathbf{p}-\mathbf{k}}{2}, \mathbf{x}, \mathbf{y} \right) \Gamma \left(\mathbf{r} + \frac{\mathbf{p}-\mathbf{k}}{2}, \mathbf{x}, \mathbf{y} \right), \quad (50)$$

where the exciton $\varphi(\mathbf{p})$ and biexciton $\Gamma(\mathbf{p}, \mathbf{l}, \mathbf{s})$ wave functions of the ground states are determined from the corresponding Schrödinger equations (46) and (30). It can be demonstrated that the final equations (49), (50) become the previous results (18), (20) in the limiting case of a weakly-coupled biexciton, when it is possible to factor (when the symmetry properties are properly accounted for) the function Γ : $\Gamma \rightarrow \Psi \varphi \varphi$.

4. DISCUSSION OF RESULTS

We return to a simpler model based on the exciton quasiparticle approximation for analyzing the exciton-biexciton optical Stark-effect. Figure 3b, provides a graph of the dispersion curves of the modified spectrum of a CdS semiconductor with the following parameters: $\omega_i = 2552$ MeV, $\Omega_0^{biex} = 5100$ MeV, $M = 0.9m_0$, $\epsilon_g = 8.87$, $\epsilon = -5$ MeV, $|\Psi(0)|^2 = 2 \cdot 10^{-18}$ cm³ in the presence of a polariton pump wave of frequency $\omega_k = 2547$ MeV and intensity $I = 5$ MW/cm². The dispersion curves were found by numerically solving Eq. (17), while the unperturbed terms of the semiconductor, i.e., in the absence of the pump wave, are shown in Fig. 3a, for comparison purposes. A numerical calculation was carried out for the case of double resonance, when the frequency $\Omega_0^{biex} - \omega_k$ falls within the unperturbed exciton line, i.e., in the range $\omega_i \pm \omega_{it}$, where ω_{it} represents the polariton longitudinal-transverse splitting. In the inverse case of distributed resonances it is natural to divide the exciton-biexciton induced splitting range at the frequency $\Omega_0^{biex} - \omega_k$ and the pump-induced effective shift δ_p^{ex} of the exciton level. The following equation can be obtained from the dispersion equation (17) for this purpose:

$$\delta_p^{ex} = \delta_p^{ex} - \frac{|m|^2}{\Omega_{p+k}^{biex} + \delta_{p+k}^{biex} - \omega_k - \omega_t}$$

$$= \epsilon |P_0|^2 \left| \Psi \left(\frac{\mathbf{p}-\mathbf{k}}{2} \right) \right|^2 \left[1 - \frac{\epsilon}{\Omega_{p+k}^{biex} + \delta_{p+k}^{biex} - \omega_k - \omega_t} \right]. \quad (51)$$

Therefore the effective dynamical shift of the exciton level is determined by two components. Specifically, the first component in Eq. (51) represents the red-shift from renormalization of the vacuum state of the semiconductor in the presence of the pump wave, as examined previously. The second term which roughly characterizes the level shift due to induced splitting of the terms at the resonance frequency $\Omega_0^{biex} - \omega_k$ may have either a positive or negative sign depending on the sign of the denominator. However for the more interesting case (from the experimental viewpoint),

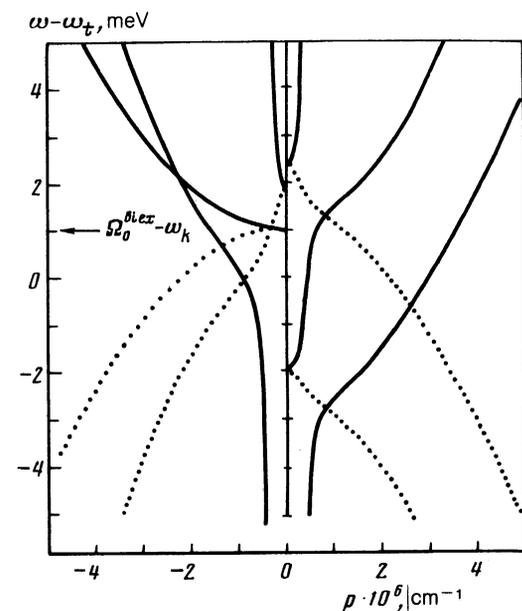


FIG. 3.

when the resonance exciton-biexciton splitting frequency lies above the exciton level ω_i , the second term in Eq. (51) corresponds, like δ_p^{ex} , to a red-shift of the exciton level.

The dispersion equation (17) was obtained in an approximation of an isolated biexciton ground level, which is valid in the frequency range $|\Omega_{\mathbf{p}+\mathbf{k}}^{\text{biex}} + \delta_{\mathbf{p}+\mathbf{k}}^{\text{biex}} - \omega_{\mathbf{k}} - \omega| < \varepsilon$. It follows that the effective exciton shift δ_p^{ex} is primarily determined by the second term in Eq. (51), i.e., it occurs due to the effect of pump-induced splitting on the position of the exciton level. However this claim requires some additional analysis.

At the same time we have so far considered the case $|P_0|^2 a_{\text{biex}}^3 \ll 1$ of moderate pump wave intensities. Formally this derives from the use in the expansion (9) of the unperturbed biexciton wave functions $\{\Psi_j\}$ determined by the corresponding Schrödinger equation (6) or, in other words, a replacement in the diagram equations (see Fig. 2) of the exact vertex function Γ by the function Γ_0 which does not contain pump lines. In the general case, it is possible to derive a solution to the problem within the framework of the exciton quasiparticle approximation for rather high pump wave intensities $|P_0|^2 a_{\text{ex}}^3 \ll 1 \ll |P_0|^2 a_{\text{biex}}^3$, as well. Using precisely the same method of introducing the independent quasiparticle operators for the general Hamiltonian (2) with the particular macropopulated mode \mathbf{k} as before, we obtain the system of equations

$$\begin{aligned} [\alpha_{2\mathbf{p}}, \hat{H}] &= (\omega_{\mathbf{p}}^{\text{ph}} - \omega_{\mathbf{k}}) \alpha_{2\mathbf{p}} + i \frac{\Omega_c}{2} B_{2\mathbf{p}} = \omega \alpha_{2\mathbf{p}}, \\ [B_{2\mathbf{p}}, \hat{H}] &= (\omega_{\mathbf{p}}^{\text{ex}} + |P_0|^2 VW(0) - \omega_{\mathbf{k}}) B_{2\mathbf{p}} - i \frac{\Omega_c}{2} \alpha_{2\mathbf{p}} \\ &+ P_0 \cdot V^{1/2} \sum_1 W \left(1 + \frac{\mathbf{k}-\mathbf{p}}{2} \right) B_{11+(\mathbf{p}+\mathbf{k})/2} B_{2-1+(\mathbf{p}+\mathbf{k})/2} = \omega B_{2\mathbf{p}}, \\ [A_{\mathbf{p}+\mathbf{k}}, \hat{H}] &= \sum_1 \left\{ \left[(\omega_{-1+(\mathbf{p}+\mathbf{k})/2}^{\text{ex}} + \omega_{1+(\mathbf{p}+\mathbf{k})/2}^{\text{ex}} \right. \right. \\ &\quad \left. \left. - 2\omega_{\mathbf{k}} + |P_0|^2 V \left[W(0) + W_{11}(0) \right. \right. \right. \\ &\quad \left. \left. \left. + W_{11} \left(1 + \frac{\mathbf{p}-\mathbf{k}}{2} \right) \right] \right] \tilde{\Psi}(1) + \sum_{1'} W(1-1') \tilde{\Psi}(1') \right\} \frac{1}{V^{1/2}} \\ &\times \left[B_{11+(\mathbf{p}+\mathbf{k})/2} B_{2-1+(\mathbf{p}+\mathbf{k})/2} + P_0 \sum_{1'} W \left(1' + \frac{\mathbf{p}-\mathbf{k}}{2} \right) \tilde{\Psi}(1') B_{21} \right] \\ &= \omega A_{\mathbf{p}+\mathbf{k}} = \frac{\omega}{V^{1/2}} \sum_1 \tilde{\Psi}(1) B_{11+(\mathbf{p}+\mathbf{k})/2} B_{2-1+(\mathbf{p}+\mathbf{k})/2}, \end{aligned} \quad (52)$$

which determines both the new dispersion law $\omega = \omega(\mathbf{p}, \omega_{\mathbf{k}}, |P_0|^2)$ and the modified series $\{\tilde{\Psi}_j\}$ of the biexciton wave functions. The use of the unperturbed biexciton wave functions $\{\Psi_j\}$ as such a series naturally leads to the previous results, specifically, the Hamiltonian (12).

In the general case, the complete analysis of Eqs. (52) is a separate problem, even for the case of biexciton ground state selected by virtue of resonance conditions. However, a qualitative analysis of this system may yield a number of conclusions. Specifically, it is possible to obtain an equation analogous to the Schrödinger equation (6) containing the effective potential $\tilde{W} = \tilde{W}(\omega, |P_0|^2)$ in place of the potential W for the modified wave function $\tilde{\Psi}_j(I)$ of the biexciton ground state. Such a transition to the effective potential \tilde{W}

explicitly reflects the screening action of the pump wave excitons \mathbf{k} on the biexciton formation process, which will reduce the attractive potential \tilde{W} as the pump wave intensity grows (the potential \tilde{W} is represented in Fig. 1 by the dashed line). This change in potential in turn results in an effective reduction of both the parameter ε and the matrix element m determining the exciton-biexciton spectral splitting in accordance with Eq. (17). At the same time the red-shift δ_p^{ex} continues to be determined by Eq. (14), which is determined directly by the form of system (52). Moreover, in this case the approximate relation (18), which was written by means of modified wave function $\tilde{\Psi}(1)$, becomes invalid. It is therefore possible to draw a qualitative conclusion that the role of the first term for the effective shift of the exciton level will grow with increasing pump wave intensity in Eq. (51).

The exciton level shift phenomenon described here is dynamical in nature and in this sense is entirely analogous to the exciton optical Stark-effect.²⁻⁶ In the latter case, however, the observation threshold of the blue-shift of the excitation level is determined by the dimensionless parameter $|P_0|^2 a_{\text{ex}}^3$ and is characterized by significant pump wave intensities I . Thus a 1 MeV dynamical shift of the exciton level in a GaAs crystal requires a pump intensity $I \sim 1 \text{ GW/cm}^2$ (Ref. 6).

This exciton and biexciton level shift effect is naturally related to the low-threshold optical Stark-effect, since in this case the observation threshold of such shifts is determined by the parameter $|P_0|^2 a_{\text{biex}}^3 \gg |P_0|^2 a_{\text{ex}}^3$. In this case, a corresponding 1 MeV shift of the exciton level for, for example, a CdS crystal, requires a pump wave intensity $I \sim 1 \text{ MW/cm}^2$.

We note the following in concluding our discussion of the dynamical shift phenomenon of the exciton level. The relatively short-range character of the exciton-exciton interaction which is responsible for the formation of biexcitons as a rule permits us to assume $\Psi((\mathbf{p}-\mathbf{k})/2) \approx \Psi(0)$, i.e., to treat the exciton level shift as a complete shift within the optical range of interest to us, $p \ll 10^6 \text{ cm}^{-1}$. Moreover, in some sense, this discussion concerns the modification of the exciton, photon and biexciton spectra of the semiconductor due to the proposed mechanism in all of phase space $p \leq 1/a_{\text{biex}}$ and not solely in the vicinity of exciton-biexciton resonance $\Omega_{\mathbf{p}+\mathbf{k}}^{\text{biex}} - \omega_{\mathbf{k}}$ usually treated.

The specific nature of this low threshold exciton-biexciton optical Stark-effect, like the exciton optical Stark-effect, is that they are both determined by the exciton, e.g., the polarization, component P_0 of the polariton pump wave. In the case where a pump wave of intensity I established by an external source is normally incident on the semiconductor surface, we can obtain the following expression for the characteristic energy parameter:

$$\begin{aligned} \varepsilon \left| \Psi \left(\frac{\mathbf{p}-\mathbf{k}}{2} \right) \right|^2 |P_0|^2 \\ = \frac{n}{(n+1)^2} \frac{\varepsilon_g^{1/2}}{c} \frac{\Omega_c^2 \left| \Psi \left(\frac{\mathbf{p}-\mathbf{k}}{2} \right) \right|^2}{(\omega_i - \omega_{\mathbf{k}})^2} \varepsilon \frac{I}{\hbar \omega_{\mathbf{k}}}, \end{aligned} \quad (53)$$

which determines the features of the modified spectrum in accordance with the dispersion equation (17). Here the polariton refractive index n is found from

$$n = \left[\varepsilon_g \left[1 + \frac{\Omega_c^2}{\omega_i^2 - \omega_{\mathbf{k}}^2} \right] \right]^{1/2}, \quad (54)$$

which is valid for $|\omega_i - \omega_k| \gg \omega_{it}$. The spectral changes here are therefore determined, in accordance with Eq. (53), by two parameters: The exciton-photon interaction strength (the parameter Ω_c) and the exciton-exciton interaction potential ε . However, as demonstrated above, the potential ε is itself responsible for the specific nature of this spectral modification mechanism.

We pause to note another interesting result. The terms $V^{-1/2} M_1(\mathbf{p}, \mathbf{q}) A_{\mathbf{p}}^+ B_{2\mathbf{q}} \alpha_{1\mathbf{p}-\mathbf{q}}$ are not generally present explicitly in the quasiparticle Hamiltonian (8) obtained here, where the matrix element M_1 is determined by Eq. (1) and characterizes the "giant" oscillator strength of the exciton-biexciton transition.¹⁵⁻¹⁷ Nonetheless the Hamiltonian (8) formally contains this matrix element, which is now naturally determined by the form of the biexciton operator (3) and the polariton term of the quasiparticle Hamiltonian. This follows from

$$\langle 0 | A_{\mathbf{p}} | H_M | \alpha_{1\mathbf{p}-\mathbf{q}}^+ B_{2\mathbf{q}}^+ | 0 \rangle = V^{-1/2} M_1(\mathbf{p}, \mathbf{q}), \quad (55)$$

where $|0\rangle$ is the ground state of the quasiparticle system considered here.

The fact that the modification of the semiconductor spectra is not determined by the giant oscillator strength, i.e., the matrix element $M_1(\mathbf{p}, \mathbf{q})$, is due to the following circumstance. The corresponding states $A_{\mathbf{p}}^+ |\mathbf{k}\rangle$ and $\alpha_{1\mathbf{k}}^+ B_{2\mathbf{p}-\mathbf{k}}^+ |\mathbf{k}\rangle$ are not independent, e.g., orthogonal states, in the presence of a coherent polariton pump wave, as was the case for the states $A_{\mathbf{p}}^+ |0\rangle$ and $\alpha_{1\mathbf{k}}^+ B_{2\mathbf{p}-\mathbf{k}}^+ |0\rangle$. Therefore formal introduction of the matrix element $M_1(\mathbf{p}, \mathbf{q})$ for this transition is not valid. Moreover, along these same lines, it follows that even the ordinary two-photon biexciton absorption coefficient is not determined by the matrix element M_1 , but rather by the matrix element examined above

$$M_2(\mathbf{p}, \mathbf{q}) = \varepsilon \Psi(\mathbf{q}-\mathbf{p}/2). \quad (56)$$

The matrix element $M_2(\mathbf{p}, \mathbf{q})$ also characterizes the giant oscillator strength of the exciton-biexciton transition in the generally understood sense,¹⁷ although by an amount $2\varepsilon/\Omega_c$ times smaller than in the case of the matrix element $M_1(\mathbf{p}, \mathbf{q})$. We note that this conclusion evidently eliminates the existing qualitative contradiction between the results of experiment and theory¹⁴ and also provides a physically valid result in the limiting case $\varepsilon \rightarrow 0$, since here the matrix element satisfies $M_1 \rightarrow \infty$ in accordance with Eq. (1), while $M_2 \rightarrow 0$ in accordance with Eq. (56).

In light of these results, some commentary on recently published studies^{24,25} devoted to an analysis of similar issues, seems appropriate. We emphasize that the authors of these studies basically limited their approach to the application of perturbation theory and a "three-level" model: The ground state, the exciton and the biexciton levels, neglecting spatial dispersion. Moreover, the discussion in these articles does not concern the dynamical shift δ_p^{ex} of the exciton level, but rather focuses on ordinary exciton-biexciton splitting.

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