Three-wave interaction in semiconductors due to an exciton–biexciton mechanism. Problem of additional boundary conditions and three-frequency solitons

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Some optical manifestations of the low-intensity exciton–biexciton optical Stark effect considered earlier [A. L. Ivanov, L. V. Keldysh, and V. V. Panashchenko, JETP **72**, 359 (1991)] are investigated. This effect involves modification of the exciton–photon–biexciton spectra of a semiconductor and dynamic shifts of exciton and biexciton levels in the presence of a polariton pump wave. The changes in the optical properties of a semiconductor in the presence of a given pump wave are described by a system of macroscopic equations for an electromagnetic field, as well as for exciton and dipole-inactive biexciton polarizations which are components of a probe radiation. These equations are used to consider the relevant Poynting theorem and expressions are derived for the components of the total energy flux of the probe wave. The problem of additional boundary conditions for such systems of macroscopic equations is formulated and analyzed. The proposed set of additional boundary conditions satisfies the requirement of continuity of the total energy flux across the boundary of a crystal. An allowance for the possibility of existence of exciton and biexciton "dead" layers is made in derivation of expressions for the reflection spectra of the probe electromagnetic radiation. A complete system of nonlinear macroscopic equations is analyzed for the case of comparable probe and pump wave intensities and its three-wave soliton solutions are considered. A consistent method is developed for finding soliton solutions in which the polariton effects have to be allowed for exactly. It is shown that the proposed approach and the soliton solutions obtained differ qualitatively from the traditional results on stimulated-Raman-scattering solitons obtained in nonlinear optics.

1. INTRODUCTION

A consistent microscopic theory of the low-threshold optical Stark effect in semiconductors due to the exciton–biexciton interaction was developed by us earlier.[1] This Stark effect is attributed primarily to the long-wavelength dynamic shift of an exciton level in the presence of a polariton pump wave and to the associated modification of the exciton–photon–biexciton spectra of a semiconductor. Such changes in the spectra of elementary excitations result in effective modification, in the exciton resonance region, of the optical characteristics of a semiconductor subjected to pumping. The present paper deals specifically with manifestations of the exciton–biexciton Stark effect.

The exciton–biexciton optical Stark effect may be manifested in an unambiguous manner in a three-wave interaction which in experimental investigations involves a probe wave and a pump wave in the form of two optical pulses generated by external sources. In our case a strong pump wave has a frequency $\omega_{\mathbf{k}}$ within the transparency range near the fundamental absorption edge of a semiconductor and it propagates practically without attenuation in the form of a polariton wave. If a second test wave of frequency ω is regarded as a probe, i.e., if its intensity is considerably less than that of the pump wave so it has no influence on the latter, we can regard the pump wave as determined by an external source. The next two sections of the present paper deal with this case. In the opposite case of a strong mutual influence of two polariton waves the investigated exciton–biexciton interaction mechanism may give rise to three-frequency solitons. A theoretical description of such soliton pulses is presented in Sec. 4.

2. SYSTEM OF MACROSCOPIC EQUATIONS

One of the most natural ways of investigating the characteristic features of the behavior of an exciton–photon–biexciton system in a semiconductor in the presence of a polariton pump wave is a study of changes in the optical properties of a semiconductor with the aid of a probe electromagnetic wave. It is convenient to formulate the problem in a manner traditional in optics, namely by deriving a system of macroscopic equations which can be used, in particular, to describe a transient propagation regime of a probe wave in a crystal. This system of macroscopic equations for the operations representing the electric field $\mathbf{E}(\mathbf{r}, t)$, the exciton polarization $\mathbf{P}(\mathbf{r}, t)$, and the biexciton nonlinear polarization $\mathbf{Q}(\mathbf{r}, t)$ can be derived from an analysis of the appropriate microscopic Hamiltonian and it is of the form:

$$\left[\frac{\varepsilon_g}{c^2}\frac{\partial^2}{\partial t^2}-\Delta_{\mathbf{r}}\right]\mathbf{E}(\mathbf{r},t)=-\frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}\mathbf{P}(\mathbf{r},t), \tag{1a}$$

$$\left[-i\frac{\partial}{\partial t}+\omega_t+\delta^{ex}-\frac{\hbar}{2M}\Delta_{\mathbf{r}}-i\gamma^{ex}\right]\mathbf{P}(\mathbf{r},t)$$

$$=\tfrac{1}{2}\omega_t\tilde{\beta}\mathbf{E}(\mathbf{r},t)-2\tilde{\mathbf{M}}_2^*\mathbf{P}_{\mathbf{k}}(\mathbf{r},t)\mathbf{Q}(\mathbf{r},t), \tag{1b}$$

$$\left[-i\frac{\partial}{\partial t}+\Omega_0^{biex}+\delta^{biex}-\frac{\hbar}{4M}\Delta_{\mathbf{r}}-i\gamma^{biex}\right]\mathbf{Q}(\mathbf{r},t)$$

$$=-2\tilde{\mathbf{M}}_2\mathbf{P}_{\mathbf{k}}(\mathbf{r},t)\mathbf{P}(\mathbf{r},t), \tag{1c}$$

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where $\mathbf{P}_k(\mathbf{r}, t) = \mathbf{P}_k\cos(\omega_k t - \mathbf{kr})$ is the exciton polarization of a coherent polariton pump wave; ε_g is the background permittivity of the crystal; M is the translation mass of an exciton; $\hbar\omega_t$ and $\hbar\Omega_0^{biex}$ are, respectively, the energies of the exciton and biexciton states of the unperturbed semiconductor; γ^{ex} and γ^{biex} are the phenomenological reciprocals of the exciton and biexciton lifetimes; $\tilde{\beta}$ is the dimensionless polariton parameter representing the strength of the exciton–photon interaction. The nature of the interaction matrix element $\widetilde{M}_2$ and of the dynamic shifts δ^{ex} and δ^{biex} of the exciton and biexciton levels was discussed by us in detail earlier.[1] The system (1) is derived making a reasonable assumption of neglect of the spatial dispersion in the expressions for the following quantities:

$$\widetilde{M}_2=\varepsilon\Psi\left(\frac{\mathbf{p}-\mathbf{k}}{2}\right)\left[\frac{1}{2\hbar\omega_t\beta}\right]^{1/2}\approx\varepsilon\Psi(0)\left[\frac{1}{2\hbar\omega_t\beta}\right]^{1/2},$$
$$\delta^{ex}=\varepsilon\left|\Psi\left(\frac{\mathbf{p}-\mathbf{k}}{2}\right)\right|^2|P_0|^2\approx\varepsilon|\Psi(0)|^2|P_\mathbf{k}|^2\frac{1}{2\hbar\omega_t\beta},\qquad(2)$$
$$\delta^{biex}\approx-\delta^{ex}.$$

Here, $\varepsilon<0$ is the characteristic biexciton potential of the exciton–exciton interaction; $\Psi(\mathbf{p})$ is the wave function in the momentum space of the relative motion of excitons in a biexciton. The operators representing the electric field and the exciton and biexciton polarizations are described by the following expressions:

$$\mathbf{E}(\mathbf{r},t)=\sum_\mathbf{p}\left(\frac{2\pi\hbar cp}{V}\right)^{1/2} i\mathbf{e}_1(\alpha_\mathbf{p}(t)e^{i\mathbf{pr}}-\alpha_\mathbf{p}^+(t)e^{-i\mathbf{pr}}),$$
$$\mathbf{P}(\mathbf{r},t)=\sum_\mathbf{p}\left(\frac{\hbar\omega_t\beta}{2V}\right)^{1/2}\mathbf{e}_2(B_\mathbf{p}(t)e^{i\mathbf{pr}}+B_\mathbf{p}^+(t)e^{-i\mathbf{pr}}),\qquad(3)$$
$$\mathbf{Q}(\mathbf{r},t)=\sum_\mathbf{p}\left(\frac{\hbar\omega_t\beta}{2V}\right)^{1/2}\mathbf{e}_3(\widetilde{A}_{\mathbf{p}+\mathbf{k}}(t)e^{i(\mathbf{p}+\mathbf{k})\mathbf{r}}+\widetilde{A}^+_{\mathbf{p}+\mathbf{k}}(t)e^{-i(\mathbf{p}+\mathbf{k})\mathbf{r}}),$$

where the creation and annihilation operators for photons ($\alpha_\mathbf{p}^+$, $\alpha_\mathbf{p}$), excitons ($B_\mathbf{p}^+$, $B_\mathbf{p}$), and biexcitons ($\widetilde{A}_\mathbf{p}^+$, $\widetilde{A}_\mathbf{p}$) are defined in the presence of a coherent pump wave; $\mathbf{e}_i$ represents the polarizations of the electromagnetic, exciton, and biexciton fields; V is the volume of the investigated crystal.

We shall consider the physical meaning and some characteristics of the system of macroscopic equations given above. First of all, we note that in the case of an anisotropic crystal and an arbitrary scattering geometry the equations in the system (1) are of tensor nature which in the coordinate system of the principal optic axes is governed by the tensor $\tilde{\beta}=\|\beta_i\delta_{ij}\|$ of the oscillator strengths of the exciton–photon transition and by the corresponding scattering tensor $\widetilde{\mathbf{M}}_2=\|\widetilde{M}_{2,ijk}\|$. However, we shall make the problem more specific by considering semiconductor CdS as the example and we shall assume the most interesting—from the point of view of manifestation of the investigated exciton—biexciton Stark effect—scattering geometry: $\mathbf{p}\|\mathbf{k}\|\mathbf{c}$, where $\mathbf{c}$ denotes the c axis; we shall assume that the pump and probe waves have opposite circular polarizations. It is in this case that the expressions given by the system (2) are valid and then the general tensor system of equations (1) may be limited to just three scalar equations (1a)–(1c) for the selected fields.

The equations of the system itself have a clear physical meaning. The first represents the usual Maxwell wave equation for the electric-field component of a probe electromagnetic wave in a semiconductor and the source of this wave is linked in a self-consistent manner to the appropriate exciton polarization. The second equation of the system describes propagation of an exciton polarization wave in our crystal and the linear source of this wave is governed by the process of exciton creation by the probe electromagnetic wave, whereas the nonlinear source of such polarization is related to the forced decay of biexcitons because of the interaction with the pump wave excitons. Finally, the last equation of the system (1) represents the propagation of the dipole-inactive biexciton polarization, the appearance of which is related to the nonlinear process of the direct Coulomb pairing of a probe wave exciton and a pump wave exciton to form a virtual biexciton.

One should point out that when the exciton–biexciton coupling vanishes, for example, in the absence of a pump wave, the first two equations of the investigated system form the usual closed system of macroscopic equations for the exciton polaritons,[2,3] except that the second is derived in the resonance approximation for the exciton–photon interaction. It should be stressed that a consistent approach to the derivation of a system of macroscopic equations from first principles makes it possible to describe in fact all the parameters of the system (1) in terms of microscopic characteristics of the semiconductor. An example of the latter conclusion is represented by the relationships in the system (2) and by the expressions derived in Ref. 1 for the parameters ω_t and $\tilde{\beta}$.

In the case of a given polariton pump wave ($\mathbf{k}$) the system of macroscopic equations (1) becomes linear in terms of the field operators $\mathbf{E}$, $\mathbf{P}$, and $\mathbf{Q}$. The permittivity of the semiconductor in the presence of the pump wave

$$\varepsilon(\mathbf{p},\omega)=\varepsilon_g+4\pi\chi(\mathbf{p},\omega)=\varepsilon_g$$
$$+2\pi\beta\omega_t\left(\Omega_0^{biex}+\delta^{biex}+\frac{\hbar(\mathbf{p}+\mathbf{k})^2}{4M}-\omega-\omega_\mathbf{k}-i\gamma^{biex}\right)\Big/$$
$$\left[\left(\omega_t+\delta^{ex}+\frac{\hbar p^2}{2M}-\omega-i\gamma^{ex}\right)\right.$$
$$\left.\times\left(\Omega_0^{biex}+\delta^{biex}+\frac{\hbar(\mathbf{p}+\mathbf{k})^2}{4M}-\omega-\omega_\mathbf{k}-i\gamma^{biex}\right)-|\widetilde{M}_2P_\mathbf{k}|^2\right]\qquad(4)$$

is independent of the statistics of the probe electromagnetic radiation[2] and is strongly nonlocal.

Intrinsic excitations in our crystal consist of photon, exciton, and biexciton components and are governed by the dispersion equation for transverse waves

$$p^2=\frac{\omega^2}{c^2}\varepsilon(\mathbf{p},\omega),\qquad(5)$$

which in particular represents also the exciton–biexciton optical Stark effect.

Using our macroscopic equations, we can formulate the appropriate Poynting energy theorem for the investigated system. In subsequent applications we will be particularly interested in the energy flux in the semiconductor, which is generally a sum of the energy fluxes of the probe and pump waves, and of a mixed flux. This mixed flux vanishes as a result of averaging over a time interval $T\gg|\omega-\omega_\mathbf{k}|^{-1}$ because of the difference between the frequencies of the probe and pump waves. We can therefore consider separately the energy flux associated with the pump wave and the energy flux of the probe radiation, the expression for which can be obtained as follows in the investigated case of a constant pump wave. The Maxwell equations can yield the familiar relationship

$$\frac{c}{4\pi}\operatorname{div}[\mathbf{EH}]+\frac{1}{4\pi}\left(\varepsilon_g\mathbf{E}\frac{\partial\mathbf{E}}{\partial t}+\mathbf{H}\frac{\partial\mathbf{H}}{\partial t}+4\pi\mathbf{E}\frac{\partial\mathbf{P}}{\partial t}\right)=0, \quad (6)$$

which must be represented in the form of an equation with a clear physical meaning:

$$\frac{\partial w}{\partial t}+\operatorname{div}\mathbf{S}+\vartheta=0. \quad (7)$$

Here **H** is the magnetic induction vector; w is the volume energy density in the medium; **S** is the energy flux vector; ϑ is governed by the parameters γ^{ex} and γ^{biex} representing the energy dissipation processes. With this in mind, following an approach similar to that developed in Ref. 5 for the conventional polariton problem, we shall substitute in $4\pi\mathbf{E}(\partial\mathbf{P}/\partial t)$ an expression for **E** found from Eq. (1b). Then, using the third equation (1c) of our macroscopic system, we find that some transformations yield the following expression for the probe radiation energy flux:

$$\mathbf{S}=\mathbf{S}_{el}+\mathbf{S}_{ex}+\mathbf{S}_{biex},$$

$$\mathbf{S}_{el}=\frac{c}{16\pi}\{[\mathbf{E}_0{}^*\mathbf{H}_0]+\text{H.c.}\}, \quad (8a)$$

$$\mathbf{S}_{ex}=-\frac{\hbar}{4\omega_t\beta M}\{[\dot{\mathbf{P}}_0{}^*\operatorname{rot}\mathbf{P}_0]+\dot{\mathbf{P}}_0{}^*\operatorname{div}\mathbf{P}_0+\text{H.c.}\}, \quad (8b)$$

$$\mathbf{S}_{biex}=-\frac{\hbar}{8\omega_t\beta M}\{[\dot{\mathbf{Q}}_0{}^*\operatorname{rot}\mathbf{Q}_0]+\dot{\mathbf{Q}}_0{}^*\operatorname{div}\mathbf{Q}_0+\text{H.c.}\}, \quad (8c)$$

where $\mathbf{E}_0$, $\mathbf{H}_0$, $\mathbf{P}_0$, and $\mathbf{Q}_0$ are the positive-frequency parts of the corresponding fields. Therefore, the total energy flux **S** of the probe radiation in the semiconductor in the presence of the polariton pump wave has three components: the electromagnetic flux $\mathbf{S}_{el}$ governed by the usual Poynting vector, as well as the exciton $\mathbf{S}_{ex}$ and biexciton $\mathbf{S}_{biex}$ energy fluxes associated with the translational motion of excitons and biexcitons, respectively. Characteristically, the expression for the biexciton flux differs from the exciton flux only by the substitutions $\mathbf{P}_0\to\mathbf{Q}_0$ and $M\to 2M$, which reflects the general physical nature of our quasiparticles. It should be noted that in the case of the excitation of only transverse excitons (as assumed here) the terms with the divergence vanish in the expressions for the fluxes and may be ignored.

3. ADDITIONAL BOUNDARY CONDITIONS OF A NEW TYPE AND REFLECTION SPECTRA OF A PROBE ELECTROMAGNETIC WAVE

Experimental studies of the exciton–biexciton optical Stark effect may involve observation of changes in the reflection spectra of probe electromagnetic radiation from the boundary of a semiconductor in the presence of a polariton pump wave. A possible scattering geometry for the case of normal incidence (investigated later) of a probe wave on the surface of a crystal is shown in Fig. 1.

It is known[3,6,7] that the spatially nonlocal nature of the permittivity $\varepsilon(\mathbf{p},\omega)$ is in this case related to the finite exciton and biexciton masses, and it may give rise to "additional" waves in the medium. The dispersion equation (1) considered here is of the sixth degree in the wave vector **p** and, therefore, there are generally six different branches of the solutions $p_i=p_i(\omega)$ representing eigenwaves of the semiconductor in the presence of a polariton pump wave. A probe electromagnetic wave with a given frequency ω excites three different damped waves of the same frequency inside the crystal. The last three roots of the dispersion equation are nonphysical because they correspond to spatial amplification of the probe wave. Therefore, we are faced with the problem of additional boundary conditions of a new type, different from the usual polariton problem where only one additional wave is associated with the spatial dispersion of excitons. One should stress here the novel and general nature of this problem in the cases of pump-induced changes in the spectra of elementary excitations in semiconductors.[8,9]

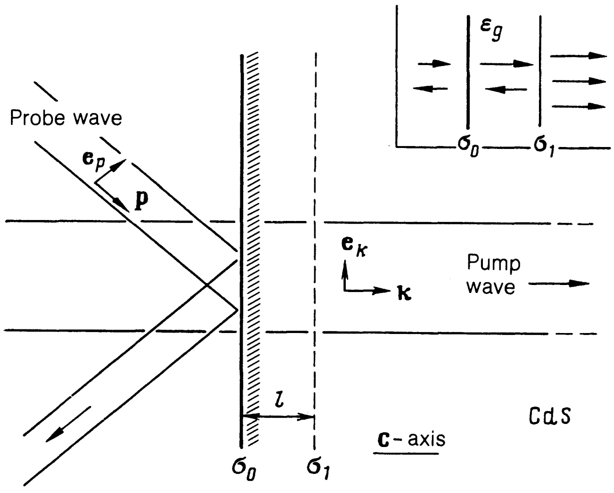

FIG. 1. Possible scattering geometry for the observation of changes in the dispersion characteristics of the semiconductor CdS in the presence of a pump wave, using the spectra of reflection of probe radiation. The inset shows the formation of the transmitted and reflected waves in the case of normal incidence of the probe radiation on the surface of a crystal.

We shall analyze the problem of the additional boundary conditions for the system of macroscopic equations (1) using a phenomenological approach based on qualitative microscopic considerations and in agreement with the natural requirement of continuity of the energy flux across the boundary of a crystal. The specifics of the problem are related to the behavior of the dipole-inactive biexciton polarization on the surface σ_0 of the investigated semiconductor. In other words, the two Maxwellian boundary conditions of continuity of the tangential components of the electric and magnetic fields

$$\mathbf{E}_\tau^{(I)}=\mathbf{E}_\tau^{(II)}, \quad \mathbf{H}_\tau^{(I)}=\mathbf{H}_\tau^{(II)} \quad (9)$$

should be supplemented by two further conditions describing the behavior of the exciton and biexciton polarizations on the boundary. If we assume that the crystal boundary represents an infinitely high potential barrier for excitons and biexcitons, we find that vanishing of the wave functions of excitons and biexcitons at the boundary leads in a natural way to the Pekar form of the additional boundary conditions for the exciton and biexciton polarizations:

$$\mathbf{P}(\mathbf{r},t)|_{\sigma_0}=0, \quad \mathbf{Q}(\mathbf{r},t)|_{\sigma_0}=0. \quad (10)$$

An analysis of our system of macroscopic equations subject to the boundary conditions (9) and (10) solves completely the problem of finding the reflection coefficient of a probe electromagnetic wave incident on the boundary of a semiconductor in the presence of a polariton pump wave (inset in Fig. 1):

$$R(\omega)=\left|\frac{X-Y}{X+Y}\right|^2, \tag{11}$$

where the variables X and Y are related to the wave vectors $p_i(\omega)$, $p_2(\omega)$, and $p_3(\omega)$, which are found from the dispersion equation (5) and which represent three waves excited in the crystal by the probe radiation of frequency ω:

$$X(\omega)=(p_1+p_2)(p_2+p_3)(p_3+p_1),$$

$$Y(\omega)=\frac{c}{\omega}\Big\{(p_1p_2p_3(p_1+p_2+p_3)+\frac{\varepsilon_g\omega^2}{c^2}(p_1^2+p_2^2+p_3^2+p_1p_2+p_2p_3+p_1p_3)-\frac{\varepsilon_g^2\omega^4}{c^4}\left(1+\frac{\omega_t\Omega_c^2Mc^2}{\omega^4\hbar\varepsilon_g}\right)\Big\}, \tag{12}$$

whereas the polariton parameter Ω_c is defined in terms of the longitudinal–transverse polariton splitting ω_{lt} or in terms of the dimensionless parameter $\tilde{\beta}$:

$$\Omega_c^2=2\omega_t\omega_{lt}=\frac{4\pi\beta}{\varepsilon_g}\omega_t^2. \tag{13}$$

It is known that experimental investigations of the conventional exciton reflection spectra frequently reveal a low-temperature spike, which is a sharp peak in the region of the exciton reflection minimum. A theoretical description of this effect is usually based on the existence of an exciton-free "dead" layer.[10] The spike appears because of the interference between a wave reflected from the σ_0 surface of a crystal and the wave reflected from the boundary σ_1 between the exciton-free layer and the rest of the semiconductor. In our case after an allowance for such a dead layer of thickness l with a background permittivity ε_g, we find that the expression for the reflection coefficient of a probe wave becomes

$$R_1(\omega)=\left|\frac{1-n+\tilde{R}(1+n)\exp(2i(\omega/c)nl)}{1+n+\tilde{R}(1-n)\exp(2i(\omega/c)nl)}\right|^2, \tag{14}$$

where $n=\varepsilon_g^{1/2}$,

$$\tilde{R}=\frac{nX-Y}{nX+Y}, \tag{15}$$

and X and Y are defined in accordance with Eq. (12).

It should be pointed out that the additional conditions of the Pekar type (10) satisfy the requirement of continuity of the energy flux of the probe radiation across the boundary of a crystal. In fact, in view of the Maxwellian boundary conditions of Eq. (9), the electromagnetic energy flux $\mathbf{S}_{el}$—defined by the Poynting vector (8a)—is continuous at the crystal boundary. Then, the continuity of the total energy flux of the probe radiation is due to the fact that, according to the additional boundary conditions (10), there are no exciton $\mathbf{S}_{ex}$ and biexciton $\mathbf{S}_{biex}$ energy fluxes [Eqs. (8b) and (8c)] at the boundary σ_0 or σ_1, depending on the adopted model, because

$$\mathbf{S}_{ex}|_\sigma\sim\dot{\mathbf{P}}_0|_\sigma\sim\omega\mathbf{P}_0|_\sigma=0,\qquad \mathbf{S}_{biex}|_\sigma\sim\dot{\mathbf{Q}}_0|_\sigma\sim(\omega+\omega_\mathbf{k})\mathbf{Q}_0|_\sigma=0.$$

The expression for the permittivity (4) suggests the occurrence of two resonances and in the case when the pump intensity goes to the limit $I\to 0$, one of these resonances coincides with the position of the exciton term, whereas the other is an exciton–biexciton resonance at a frequency $\Omega^{\text{biex}}-\omega_\mathbf{k}$. It is convenient to distinguish here the case of a double resonance, when the frequency $\Omega^{\text{biex}}-\omega_k$ lies within the region of the unperturbed exciton term, i.e., in the region $\omega_t\pm\omega_{lt}$, from the opposite case of separate resonances.

The numerical calculations reported below and carried out to illustrate the above expressions were made for the specific case of the semiconductor CdS with the following parameters: $\hbar\omega_t=2552$ meV, $\hbar\omega_{lt}=1.9$ meV, $\hbar\Omega_0^{\text{biex}}=5100$ meV, $M=0.9m_0$, $\hbar_\varepsilon=-5$ meV, $\varepsilon_g=8.87$, and $|\Psi(0)|^2=2\cdot 10^{-18}$ cm^3. In the double resonance case (Fig. 2) the modification of the reflection spectra is quite complex and is related to considerable changes which affect all three types of the initial excitations: excitons, photons, and biexcitons. Variation of the frequency $\omega_\mathbf{k}$ of the pump wave can be used to realize the second case, when the exciton reflection spectra and the pump-induced reflection in the exciton–biexciton resonance region have different frequencies. The exciton–biexciton resonance then lies in a photon-like unperturbed polariton dispersion region, which results in considerable attenuation of the relevant reflection line compared with the double resonance. However, in this case the dynamic long-wavelength shift of the exciton level appears in its pure form, i.e., the exciton–biexciton optical Stark effect is observed (Fig. 3).

It should be pointed out that introduction of a dead layer gives rise to a double spike in the reflection spectra, such as that shown for example in Fig. 2 (see also Fig. 5). In the case of the separate resonances this new spike represents the pump-induced reflection line in the region of the exciton–biexciton resonance. In general, both spikes are of the same interference nature and are linked to two spectral points corresponding to nonzero values of the long-wave-

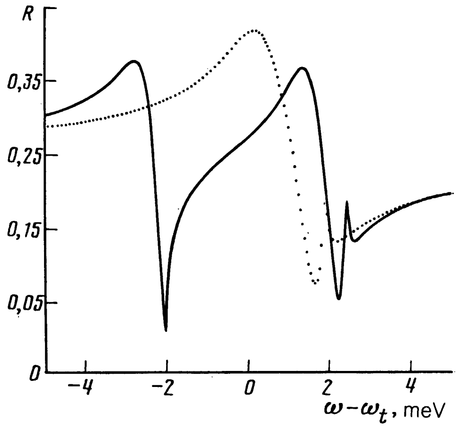

FIG. 2. Modification of the reflection spectra of the probe radiation incident on the semiconductor CdS in the presence of a pump wave of intensity $I=5$ MW/cm^2 (outside the crystal) of frequency $\omega_\mathrm{k}=2547$ meV when the thickness of the exciton-free layer is $l=90$ Å. The case of a double resonance is illustrated. The dotted curve represents the exciton reflection line in the absence of pumping.

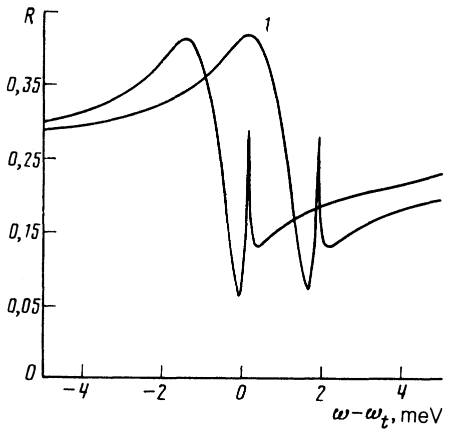

FIG. 3. Shift of the exciton reflection in the case of separate resonances. The pump wave frequency is $\omega_k = 2541$ meV and its intensity is $I = 25$ MW/cm^2. The thickness of the "dead" layer is $l = 90$ Å. Curve 1 represents the unperturbed exciton reflection spectrum.

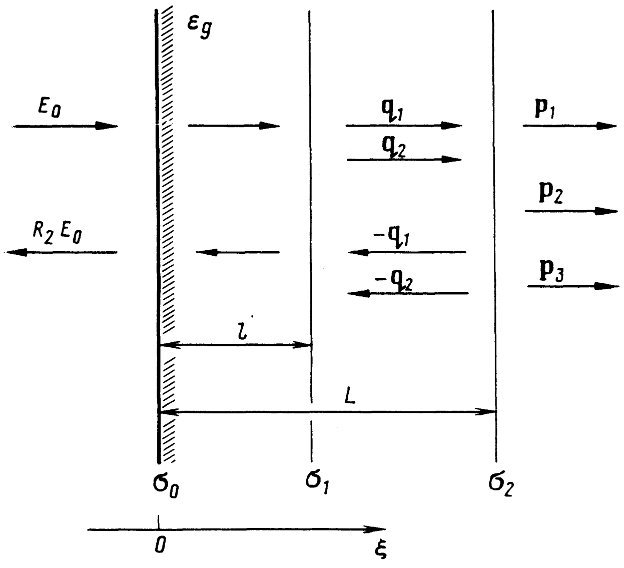

FIG. 4. Formation of the transmitted and reflected waves allowing for the presence of a biexciton-free layer. Two ordinary polariton eigenwaves of a crystal form in the region $\sigma_1 - \sigma_2$.

length limit of the modified dispersion curves of the investigated semiconductor.

We shall discuss in greater detail the problem of the dead layer in the formation of the reflection spectra of the probe radiation. The physical justification for introducing the exciton-free dead layer is the qualitative idea that, because of its finite radius a_{ex}, an exciton cannot approach the boundary of a crystal closer than the distance $l \sim a_{ex}$. In the problem under discussion it is natural to generalize the concept of such a dead layer and introduce similarly an additional biexciton-free layer of thickness $L \sim a_{biex}$, where a_{biex} is the biexciton radius. Figure 4 shows schematically the process of formation of the reflected and transmitted waves in this model. The part of the crystal $\sigma_1 - \sigma_2$ enclosed between the boundaries of the two dead layers is unusual. Here, the incident probe radiation excites two ordinary unperturbed polariton waves and these in turn create, beyond the boundary σ_2, three eigenwaves corresponding to the modified spectrum of the semiconductor in the presence of the pump wave. It should be stressed that crossing of the boundary σ_2 of the biexciton-free layer not only activates the exciton–biexciton mixing, but also induces (by the pump wave) an abrupt long-wavelength shift of the exciton level.

The reflection coefficient of the probe electromagnetic wave can be found by considering the additional conditions on the boundaries σ_1 and σ_2. The problem of the radiation crossing the boundary σ_1 is accounted for fully by one additional boundary condition, which—as before—will be assumed to be the Pekar condition of the absence of the exciton polarization on the surface σ_1, i.e., $\mathbf{P}|_{\sigma_1} = 0$. At the boundary σ_2 the corresponding additional boundary condition applicable to the biexciton polarization $\mathbf{Q}|_{\sigma_2} = 0$ should be supplemented by two further boundary conditions. The latter could be the conditions of continuity of the exciton polarization $\mathbf{P}$ and of its derivative $\partial \mathbf{P}/\partial \xi$ at the boundary σ_2.

From the microscopic point of view, such boundary conditions are linked to the requirement of continuity of the exciton wave function and of its spatial derivative at the point $\xi = L$ where there is a finite jump in the exciton potential. Therefore, in the case of the two-layer model proposed here, the additional boundary conditions are

$$\mathbf{P}(\mathbf{r}, t)|_{\sigma_1}=0, \quad \mathbf{Q}(\mathbf{r}, t)|_{\sigma_2}=0,$$
$$\mathbf{P}(\mathbf{r}, t)|_{\sigma_2-\delta}=\mathbf{P}(\mathbf{r}, t)|_{\sigma_2+\delta}, \quad \left.\frac{\partial \mathbf{P}}{\partial \xi}\right|_{\sigma_2-\delta} = \left.\frac{\partial \mathbf{P}}{\partial \xi}\right|_{\sigma_2+\delta}, \quad \delta \to 0. \tag{16}$$

It should be stressed that the additional boundary conditions (16) satisfy the requirement of continuity of the energy flux of the probe radiation across the boundaries σ_1 and σ_2. At the boundary σ_2 not only the electromagnetic energy flux $\mathbf{S}_{el}$ is continuous, but it applies also to the exciton flux $\mathbf{S}_{ex}$. The continuity of the latter follows directly from Eq. (8b) for the exciton energy flux and from the proposed exciton additional boundary conditions at the surface σ_2.

After introduction of the initial boundary conditions (16) and fairly cumbersome calculations, we obtain an expression for the reflection coefficient $R_2(\omega)$ of the probe electromagnetic wave incident on the boundary of a crystal. The reflection is still given by (14), where in the expression (15) for the reflection coefficient $\tilde{R}$ we have to replace the parameters X and Y with the following quantities:

$$\tilde{X}=(q_1^2-q_2^2)(1+\alpha_{11}+\alpha_{22}+\alpha_{11}\alpha_{22}-\alpha_{12}\alpha_{21}),$$
$$\tilde{Y}=\frac{c}{\omega}[(q_1-q_2)(q_1q_2+\lambda^2)(1+\alpha_{12}\alpha_{21}-\alpha_{11}\alpha_{22}) + (q_1+q_2)(q_1q_2-\lambda^2)(\alpha_{11}-\alpha_{22}) + 2q_2(q_2^2-\lambda^2)\alpha_{12}-2q_1(q_1^2-\lambda^2)\alpha_{21}]. \tag{17}$$

In this case the matrix of the coefficients α_{ij} is given by

$$\alpha_{11}=-\frac{f(p_1, p_2, p_3, -q_1, q_2)}{f(p_1, p_2, p_3, q_1, q_2)}\exp(2iq_1\tilde{L}),$$
$$\alpha_{12}=-\frac{f(p_1, p_2, p_3, q_1, -q_1)}{f(p_1, p_2, p_3, q_1, q_2)}\exp(i(q_1+q_2)\tilde{L}), \tag{18}$$
$$\alpha_{21}=\alpha_{12}(q_1\to q_2,\ q_2\to q_1), \quad \alpha_{22}=\alpha_{11}(q_1\to q_2,\ q_2\to q_1),$$
$$\tilde{L}=L-l,$$

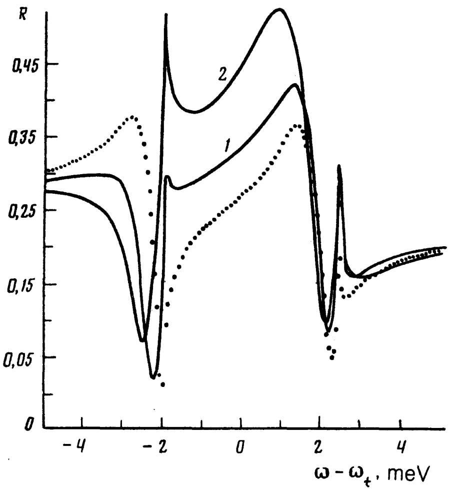

FIG. 5. Influence of the thickness of the biexciton-free layer on the form of the reflection spectra of the probe radiation in the presence of a pump wave of frequency $\omega_k = 2547$ meV and of intensity $I = 5$ MW/cm^2 when the thickness of the biexciton-free layer is $l = 90$ Å. Curve 1 corresponds to $L = 180$ Å and curve 2 corresponds to $L = 250$ Å. The dotted curve represents the reflection spectrum calculated using the one-layer model (Fig. 2).

where in turn the function f is related to the wave vectors q_1 and q_2 of the polariton waves excited by the probe wave frequency ω in the $\sigma_1 - \sigma_2$ layer and to the corresponding wave vectors p_1, p_2, and p_3 of the three waves propagating in the bulk of the semiconductor:

$$f(p_1, p_2, p_3, q_1, q_2) = (q_1 - q_2)$$
$$\times\Big\{(p_1+q_1)(p_2+q_1)(p_3+q_1)(p_1+q_2)(p_2+q_2)$$
$$\times(p_3+q_2) - \frac{2M}{\hbar}\tilde{\delta}[(q_1q_2+\lambda^2)(p_1p_2+p_1p_3+p_2p_3-q_1q_2)$$
$$+(q_1+q_2)[p_1p_2p_3+\lambda^2(p_1+p_2+p_3+q_1+q_2)]]\Big\}. \quad (19)$$

The following additional notation is used in Eqs. (17)–(19):

$$\lambda^2 = \frac{\varepsilon_g\omega^2}{c^2}, \quad \tilde{\delta} = \delta^{ex} - i(\gamma^{ex} - \gamma_0^{ex}), \quad (20)$$

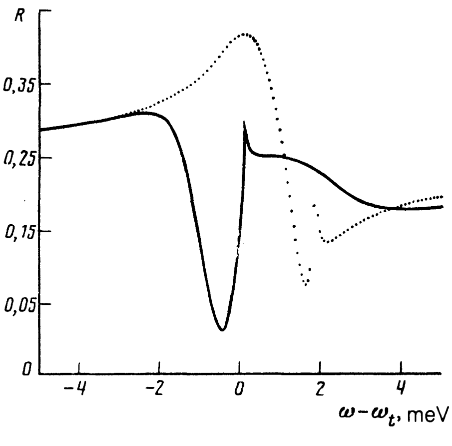

FIG. 6. Reflection spectrum of the probe radiation in the case of separate resonances. The dotted curve represents the exciton reflection in the absence of pumping. The continuous curve is the reflection spectrum of Fig. 3 after allowance for the biexciton-free layer of thickness $l = 200$ Å.

where γ_0^{ex} is the exciton attenuation in the $\sigma_1 - \sigma_2$ layer. It should be pointed out that the expression obtained for the reflection coefficient $R_2(\omega)$ is symmetric under transpositions of the wave vectors q_1 and q_2 or p_1, p_2, and p_3.

Figure 5 shows the reflection spectra of a probe electromagnetic wave obtained for different thicknesses L of the biexciton-free layer. The differences in the profiles of the reflection lines compared with the one-layer model considered earlier are quite large, so that we can consider an additional mechanism associated with the nature of the additional boundary conditions and modifying the reflection spectra of a semiconductor in the presence of a polariton pump wave. This is manifested clearly also in the case of the separate resonances (Fig. 6).

4. POSSIBILITY OF FORMATION OF POLARITON SOLITONS DUE TO THE EXCITON–BIEXCITON INTERACTION

We have analyzed so far the case when a probe electromagnetic wave is so weak that it does not influence a polariton pump wave regarded as governed by an external source. However, it is very interesting to consider the nature of the propagation of two coherent polariton pulses of comparable intensity when they interact resonantly due to the exciton–biexciton mechanism. In this case we can expect formation of three coupled solitons in a semiconductor: they are two polariton solitons and one biexciton soliton, all propagating at the same velocity v_s, without dispersion spreading. The problem is topical not only because of the increasing interest in such nonlinear waves in crystal optics,[11,13] but also because of the special manifestation of the investigated exciton–biexciton nonlinearity mechanism. An important feature of the approach adopted below is a correct allowance for the polariton effects, i.e., for the proximity of the carrier frequencies ω and ω_k of coherent electromagnetic pulses incident on a crystal to an exciton resonance at a frequency ω_t.

The system of the macroscopic equations (1) becomes closed and self-consistent on addition of two equations describing the electric field $\mathbf{E}_k$ and the polarization $\mathbf{P}_k$ of a pump pulse. These two equations resemble Eqs. (1a) and (1b), and they represent a certain equivalence and mutual influence, via the biexciton field, of the pump and probe waves of comparable intensities. We shall consider a specific case and simplify the treatment, as we have done in the analysis of the problem of the additional boundary conditions, by selecting the case $\mathbf{p}\|\mathbf{k}\|\xi$, where $\mathbf{p} = \mathbf{p}(\omega)$ and $\mathbf{k} = \mathbf{k}(\omega_k)$ are the carrier-wave vectors of the polariton pulses and $\xi \| \mathbf{c}$ is the coordinate axis along which these pulses are propagating. Then, limiting our study to the soliton solutions of the system of the selected macroscopic equations, we shall seek them in the form reflecting the requirement of the frequency and wave matching:

$$E(\mathbf{r}, t) = \tilde{E}(\eta)\exp(-i\omega t + ip\xi),$$
$$P(\mathbf{r}, t) = \tilde{P}(\eta)\exp(-i\omega t + ip\xi),$$
$$E_k(\mathbf{r}, t) = \tilde{E}_k(\eta)\exp(-i\omega_k t + ik\xi), \quad (21)$$
$$P_k(\mathbf{r}, t) = \tilde{P}_k(\eta)\exp(-i\omega_k t + ik\xi),$$
$$Q(\mathbf{r}, t) = \tilde{Q}(\eta)\exp(-i(\omega+\omega_k)t + i(p+k)\xi),$$

where $\eta = t - \xi/v_s$ is the associated time and $\widetilde{F}(\eta)$ is the soliton profile of the envelope of the relevant field. In the approximation of slowly varying amplitudes, the original system of five macroscopic equations can be transformed to a closed system of three nonlinear equations for the positive-frequency parts of the envelopes of the exciton and biexciton polarizations:

$$\begin{aligned}
\frac{d}{d\tau}\tilde{P} &= -i\nu_1\tilde{P} - i\beta_1\tilde{P}_k^*\tilde{Q} + i\rho_1(2|\tilde{P}|^2 - |\tilde{P}_k|^2)\tilde{P},\\
\frac{d}{d\tau}\tilde{P}_k &= -i\nu_2\tilde{P}_k - i\beta_2\tilde{P}^*\tilde{Q} + i\rho_2(2|\tilde{P}_k|^2 - |\tilde{P}|^2)\tilde{P}_k,\\
\frac{d}{d\tau}\tilde{Q} &= -ia\tilde{Q} - i\mu\tilde{P}_k\tilde{P} - i\Delta(|\tilde{P}|^2 + |\tilde{P}_k|^2)\tilde{Q}.
\end{aligned} \tag{22}$$

The parameters of the equations are given by the relationship

$$\begin{aligned}
\nu_j &= \frac{a_j s_j - 2\pi\beta\omega_t\omega_j^2}{g_j}, \qquad \beta_j = \frac{\varepsilon\Psi(0)s_j}{[2\hbar\omega_t\beta]^{1/2}g_j},\\
\rho_j &= \frac{\varepsilon|\Psi(0)|^2 s_j}{2\hbar\omega_t\beta g_j}, \qquad a = \left(\Omega_0^{biex} + \frac{\hbar(p+k)^2}{4M} - \omega - \omega_k\right)(\omega_t b)^{-1},\\
\mu &= \frac{\varepsilon\Psi(0)}{[2\hbar\omega_t\beta]^{1/2}\omega_t b}, \qquad \Delta = -\frac{\varepsilon|\Psi(0)|^2}{2\hbar\omega_t^2\beta b},\\
b &= 1 - \frac{\hbar(p+k)}{2Mv_s},
\end{aligned} \tag{23}$$

where the index is $j = 1$ or 2 and

$$\begin{aligned}
&g_j = \omega_t b_j s_j + 4\pi\beta\omega_j\omega_t^2(1 + \omega_j d_j/s_j), \quad d_j = \varepsilon_g\omega_j - c^2 q_j/v_s,\\
&a_j = \omega_t - \omega_j + \hbar q_j^2/2M, \quad s_j = c^2 q_j^2 - \varepsilon_g\omega_j^2, \quad b_j = 1 - \hbar q_j/Mv_s.
\end{aligned} \tag{24}$$

The term with the cubic nonlinearity on the right-hand side of the system (22) represents the nature of the dynamic shifts of the positions of the exciton and biexciton levels because of the mutual attraction of excitons in two different polariton waves and also because of repulsion between the excitons forming each of the waves separately. The following additional notation is used in the expressions given by the system (24): $q_1 = p, q_2 = k, \omega_1 = \omega, \omega_2 = \omega_k$. Moreover, in the system (22) the dimensionless time $\tau = \omega_t \eta$ is adopted.

In contrast to the approach traditional in nonlinear optics,[14,15] the three-wave system of equations obtained above does not contain electromagnetic fields as the variables, but their polarizations. However, the actual form of the equations in the system (22) is natural and related to the circumstance that the polariton waves interact with one another by the polarization components, i.e., by the exciton components. The biexciton wave is dipole-inactive.

There is one additional and most important reason for adopting the polarization system (22). The similarity of the carrier frequencies ω and ω_k of the polariton waves to an exciton resonance ω_t means that incorrect results would be obtained by the usual procedure employed in the description of nonlinear processes with the aid of just the nonlinear lower order electrical susceptibilities, i.e., $\chi^{(2)}$ and $\chi^{(3)}$. This can be illustrated quite well by taking the example of the relationship (4), from which it follows that when the general expression for $\chi = \chi(\omega, \mathbf{p}, |P_k|^2)$ is expanded as a series near an exciton resonance land, this series contains all the nonlinear susceptibilities $\chi^{(n)}$, but we need retain in our analysis just the lowest of them, which in our case is $\chi^{(3)}$.

The use of a system of the polarization equations (22) makes it possible to bypass this difficulty and in fact to allow explicitly, within the framework of the initial microscopic model, for a whole series of the nonlinear susceptibilities, i.e., we can consider the problem rigorously. The envelopes of the electric fields of the polariton pulses are then related to the corresponding exciton polarizations by

$$\begin{aligned}
\tilde{E} &= \left[4\pi\omega_1^2\tilde{P} + 8\pi i\omega_1\omega_t\left(1 + \frac{d_1\omega_1}{s_1}\right)\dot{\tilde{P}}\right]s_1^{-1},\\
\tilde{E}_k &= \left[4\pi\omega_2^2\tilde{P}_k + 8\pi i\omega_2\omega_t\left(1 + \frac{d_2\omega_2}{s_2}\right)\dot{\tilde{P}}_k\right]s_2^{-1}.
\end{aligned} \tag{25}$$

The use of the method of slowly varying amplitudes imposes the following constraint on the values of the soliton velocity v_s and on the duration of the soliton pulses τ_s:

$$\frac{c}{\tau_s\omega_t v_s} \ll 1. \tag{26}$$

Moreover, the system of equations (22) represents the cases of the interaction of two polariton pulses with nondegenerate carrier frequencies $|\omega - \omega_k|^{-1} \ll \tau_s$ in the absence of attenuation $\gamma^{ex} = \gamma^{biex} = 0$.

Introducing the real amplitudes x, y, and z and the phases φ, ψ, and ζ of positive-frequency soliton envelopes

$$\tilde{P} = xe^{i\varphi}, \quad \tilde{P}_k = ye^{i\psi}, \quad \tilde{Q} = ze^{i\zeta} \tag{27}$$

we obtain the nonlinear equations

$$\begin{aligned}
&\dot{x} = -\beta_1 yz\sin\theta, \quad \dot{y} = -\beta_2 xz\sin\theta, \quad \dot{z} = \mu xy\sin\theta,\\
&x\dot{\varphi} = -\nu_1 x + \rho_1(2x^2 - y^2)x - \beta_1 yz\cos\theta,\\
&y\dot{\psi} = -\nu_2 y + \rho_2(2y^2 - x^2)y - \beta_2 xz\cos\theta,\\
&z\dot{\zeta} = -az - \Delta(x^2 + y^2)z - \mu xy\cos\theta,
\end{aligned} \tag{28}$$

where $\theta = \varphi + \psi - \zeta$ is the phase matching angle for the interaction between the polarization waves.

The first three amplitude equations of the system (28) have three integrals of motion and of these two are independent:

$$\beta_2 x^2 - \beta_1 y^2 = C_1, \quad \mu x^2 + \beta_1 z^2 = C_2, \quad \mu y^2 + \beta_2 z^2 = C_3. \tag{29}$$

An analysis of the integrals of motion given by Eq. (29) allows us to classify fully all possible soliton solutions the nature of which is governed by the signs of the parameters β_1, β_2, and μ and by the values of the constants C_i. However, we shall consider only the most interesting (from our point of view) class of solutions and use it as an example to illustrate the specific features of the method adopted to investigate the system (28). We shall in fact consider the case when the constant has the value $C_2 = 0$ in the integrals of motion of Eq. (29). It should be pointed out that, in accordance with a definition generally accepted in nonlinear optics,[14] the three-wave soliton solutions are understood to be such three steady-state nonlinear waves that at least of them are solitary, i.e., their amplitudes tend to zero for $\tau \to \pm\infty$. Then, in the case selected for investigation, we find from the integrals of motion (29) that the relationship between the amplitudes of the nonlinear waves is

$$x^2=-\frac{\beta_1}{\beta_2}(y_0^2-y^2), \quad z^2=\frac{\mu}{\beta_2}(y_0^2-y^2), \tag{30}$$

where y_0 is the amplitude of the polarization wave $P_{\mathbf{k}}$ in the limit $\tau \to \pm\infty$. Therefore, a steady-state wave **k** can still be regarded as a pump wave in which the solitary pulses of the polariton wave **p** and of the wave **p** + **k** of the biexciton polarization "burn a hole" representing a dark soliton, which is a pulse representing reduction in the pump intensity.

This situation may be realized experimentally if a coherent electromagnetic pulse of sufficiently long duration is used as the pump wave and a short probe pulse of comparable intensity forms a dark soliton.[16] The relationship described by Eq. (30) is possible for the following signs of the parameters of the system (29): $\beta_1>0, \beta_2<0$. We then have $\mu<0$, which follows directly from Eq. (23) in the case when $\Psi(0)>0$. The frequencies ω and $\omega_{\mathbf{k}}$ of electromagnetic pulses set by an external source, the intensity of the pump wave governed by the value of y_0, and the duration τ_s of the soliton pulses are suitable free parameters of the problem.

Applying the formal procedure of substitution of the variables

$$u=\frac{y}{y_0}\cos\theta, \quad v=\frac{y}{y_0}\sin\theta \tag{31}$$

and using the relationships in Eq. (30), we can separate from the complete system (28), a closed system of two nonlinear equations suitable for the final analysis:

$$\begin{aligned} \dot{u}&=2uv-\tilde{B}v-\varkappa v(u^2+v^2),\\ \dot{v}&=1-3u^2-v^2+\tilde{B}u+\varkappa u(u^2+v^2). \end{aligned} \tag{32}$$

Here, the parameters $\tilde{B}$ and $\varkappa$ are given by

$$\begin{aligned} \tilde{B}&=\frac{a-\nu_1-\nu_2}{y_0|\beta_1\mu|^{1/2}}+\frac{y_0(\Delta+2\rho_1-\rho_2)}{|\beta_1\mu|^{1/2}}\left|\frac{\beta_1}{\beta_2}\right|,\\ \varkappa&=\left[\Delta-\rho_1+2\rho_2-(\Delta+2\rho_1-\rho_2)\left|\frac{\beta_1}{\beta_2}\right|\right]\frac{y_0}{|\beta_1\mu|^{1/2}}, \end{aligned} \tag{33}$$

and we adopt a new time

$$\tau'=y_0|\beta_1\mu|^{1/2}\tau. \tag{34}$$

If we regard u and v as canonically conjugate variables, we can construct the following Hamiltonian for the system of equations (32):

$$\tilde{H}(u, v)=(\tilde{B}/2)(u^2+v^2)+(\varkappa/4)(u^2+v^2)^2-u(u^2+v^2-1), \tag{35}$$

which is an integral of motion. This is precisely why we can find analytically the soliton solutions of the complete system (28). The limiting stationary points of the system (32)

$$u_\infty=(\tilde{B}+\varkappa)/2, \quad v_\infty=\pm[1-(\tilde{B}+\varkappa)^2/4]^{1/2}, \tag{36}$$

representing the behavior of the soliton solutions in the limit $\tau' \to \pm\infty$, allow us to establish an algebraic relationship between the variables u and v in the form

$$\tilde{H}(u,v)=\tilde{H}(u_\infty, v_\infty)=\tilde{B}/2+\varkappa/4. \tag{37}$$

Before we continue finding the profile of the soliton pulses, we shall consider the task of determination of the dispersion relationships for carrier-wave vectors of the polariton solitons. These dispersion relationships follow from the requirement of the absence of phase modulation in the limit $\tau \to \pm\infty$:

$$\dot{\varphi}|_{\tau\to\pm\infty}=\dot{\psi}|_{\tau\to\pm\infty}=\dot{\zeta}|_{\tau\to\pm\infty}=0, \tag{38}$$

as assumed initially in our approach by selecting the solutions in the form of Eq. (21). An analysis of the last three equations of the system (28) including Eq. (30) makes it possible to derive the following two independent equations form the conditions (38):

$$\begin{aligned} &\nu_2-2\rho_2^2y_0=0,\\ &\nu_1+a+y_0^2(\rho_1+\Delta)=0, \end{aligned} \tag{39}$$

which are in fact the required dispersion relationships. Using the first of them, we can determine the carrier wave vector $k=k(\omega_{\mathbf{k}}, I)$ of the pump wave. This dispersion relationship for a dark soliton then represents the usual polariton dispersion law which allows for a short-wavelength shift of the exciton level induced by the pump wave itself. The physical meaning of this result is quite clear: it is related to the fact that this should be the dispersion relationship in the absence of two corresponding solitary polariton and biexciton waves, i.e., when $\tau \to \pm\infty$.

The second of the equations (39) allows us to find the carrier-wave vector $p=p(\omega, \omega_{\mathbf{k}}, v_s, I)$ of a solitary polariton wave. This dispersion relationship contains, in the form of an as yet unknown parameter, the solitary velocity v_s and it generally differs from the usual polarization dispersion law and from the spectral relationship (5) considered earlier. This is the essential difference between our approach and the traditional procedure,[14] where in the investigation of the similar problem of stimulated Raman scattering solitons the dispersion relationships are used justifiably in the form of the unperturbed dispersion laws that apply in the absence of any interaction between the waves. In our case this would have been an incorrect selection of the usual polariton dispersion law and this would have resulted, in particular, to the conditions $\nu_1=\nu_2=0$ that the terms linear in the polarization vanish from the first and third equations of the initial system (21).

We shall consider one further important circumstance. The requirement that the coordinate v_∞ at the stationary points of Eq. (36) should be real leads to the condition $|\tilde{B}+\varkappa|\leqslant 2$, which together with the relationships (39) can be transformed to

$$|a+\Delta y_0^2|\leqslant y_0|\beta_1\mu|^{1/2}. \tag{40}$$

We can easily see that this inequality defines the frequency interval of the existence of the soliton solutions proportional to the amplitude ($y_0 \propto I^{1/2}$) of the pump wave, with the center of the interval given by the condition $a+\Delta y_0^2=0$ and corresponding to the exact resonance of the interaction of the waves when allowance is made for the dynamic shift of the biexciton level.

Using the integral of motion (37), we can now find the soliton solution of the system (32) and then, taking account of the dispersion relationships given by the system (39), we obtain nonlinear solutions of the complete system (28) in their final form. We then find that the intensity profiles of the investigated soliton pulses are

$$y^2=(1-\Phi)y_0^2,\quad x^2=-\Phi\frac{\beta_1}{\beta_2}y_0^2,\quad z^2=\Phi\frac{\mu}{\beta_2}y_0^2,\qquad (41)$$

where

$$\Phi=\frac{4(1-\Lambda^2)}{(\varkappa^2+4\Lambda\varkappa+4)^{1/2}\,\mathrm{ch}(\tau/\tau_s)+\Lambda\varkappa+2},\qquad (42)$$

and the parameter Λ is given by

$$\Lambda=-\frac{(\tilde{B}+\varkappa)}{2}=-\frac{a+\Delta y_0^2}{y_0|\beta_1\mu|^{1/2}}.\qquad (43)$$

The soliton pulse duration τ_s is then given by

$$\tau_s=[2y_0[(1-\Lambda^2)|\beta_1\mu|]^{1/2}]^{-1}.\qquad (44)$$

Therefore, in the frequency interval defined by Eq. (40) and representing the range of existence of our soliton solutions, these solutions have the minimum duration and the maximum amplitude at the central point of the exact resonance where $\Lambda=0$. A shift of the frequency detuning $(\Omega^{\text{biex}}_{\mathbf{p}+\mathbf{k}}-\omega-\omega_{\mathbf{k}})\sim a$ to the end of the interval of Eq. (40) increases the duration of the solitons and reduces their amplitude for fixed values of the parameters $\omega_{\mathbf{k}}$, I, and v_s. At the end point of the frequency interval of the existence of the soliton solutions we have $\Lambda=\pm 1$ and, in accordance with Eqs. (41) and (42), we find that two solitary waves out of the three soliton solutions vanish and the third (pump) wave becomes a steady-state nonlinear wave of constant intensity. Typical graphs showing the profiles of the polariton solitons are presented in Fig. 7. It should be noted that the relationship (44) allows us to derive an expression for the soliton velocity $v_s=v_s(\omega,\ \omega_{\mathbf{k}},\ \tau_s,\ I,\ p,\ k)$. Therefore, for given parameters of the problem ω, $\omega_{\mathbf{k}}$, τ_s, and I we have to consider Eq. (44) and the second equation of (39) as a system of algebraic equations for the variables p and v_s.

It is of interest to consider also the time dependence of the phase-matching angle θ

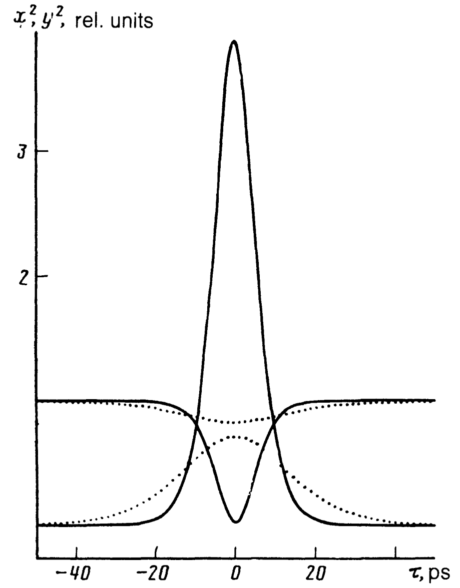

FIG. 7. Intensity profiles of the soliton pulses of the exciton polarization. The calculations were carried out for fixed parameters $\omega_{\mathbf{k}}=2540$ meV, $I=0.2$ MW/cm^2, $v_s=5.54\times10^7$ cm/s. The continuous curves correspond to $\omega=2560$ meV, ($\Lambda=-0.073$, $\varkappa=-0.385$, $\tau_s=3.42$ ps). The dotted curves represent the pulses obtained for $\omega=2560.1$ meV ($\Lambda=0.93$, $\varkappa=-0.415$, $\tau_s=8.94$ ps).

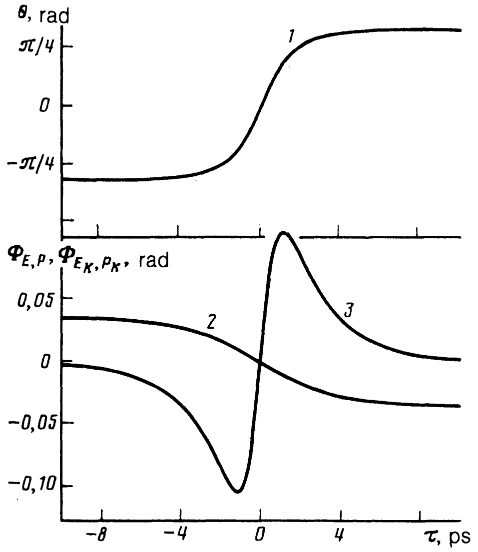

FIG. 8. Time dependence of the phase-matching angle θ (1) and of the phase shifts between the electric field and the polarizations in the case of the "bright" (2) and "dark" (3) solitons, calculated for the following parameters of the soliton pulses: $\tau_s=2.1$ ps, $I=0.2$ MW/cm^2, $\omega_{\mathbf{k}}=2540$ meV, $\omega=2560.1$ meV ($\Lambda=0.51$).

$$\mathrm{tg}\,\theta=\pm\frac{[(1-\Lambda^2)(\varkappa^2+4\Lambda\varkappa+4)]^{1/2}\,\mathrm{sh}(\tau/\tau_s)}{\Lambda(\varkappa^2+4\Lambda\varkappa+4)^{1/2}\,\mathrm{ch}(\tau/\tau_s)+2\Lambda+\varkappa}\qquad (45)$$

and of the phase shift $\phi_{E,\,P}$ or $\phi_{E_{\mathbf{k}},P_{\mathbf{k}}}$ between the electric fields and the polarizations of the appropriate polariton pulse:

$$\mathrm{tg}(\phi_{E,P})=\frac{\dot{x}}{x(2\omega_t/\omega_1+2\omega_t d_1/s_1)^{-1}-x\dot{\varphi}},$$
$$\mathrm{tg}(\phi_{E_{\mathbf{k}},P_{\mathbf{k}}})=\frac{\dot{y}}{y(2\omega_t/\omega_2+2\omega_t d_2/s_2)^{-1}-y\dot{\psi}},\qquad (46)$$

where the quantities x, $x\dot{\varphi}$, y, $y\dot{\psi}$ are governed by the nonlinear quadratures for the envelopes of the soliton pulses given by Eqs. (41) and (42), by the system of equations (26), and by the expression (45) describing the phase-matching angle. Figure 8 shows the time dependences of $\theta=\theta\tau$, $\phi_{E,\,P}(\tau)$ and $\phi_{E_{\mathbf{k}},P_{\mathbf{k}}}(\tau)$.

We shall conclude this analysis by mentioning one further qualitative difference betwen our soliton solutions and the generally accepted results of nonlinear optics. The adopted approach makes it possible to analyze the case of the appearance of a dark soliton at one of the polariton frequencies, whereas it is usually assumed that a dark soliton can form only in the case of a wave with the maximum frequency. In our problem this wave is the dipole-inactive biexciton polarization wave, which obviously cannot be used as the pump wave.

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