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Polariton trapping by a soliton near an excitonic resonance

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The nonlinear dynamics of optical pulse propagation is considered in the spectral region above the resonance frequency of a longitudinal exciton (ω_L) where two different polaritons, UP and LP, propagate through a crystal at the same frequency. The Kerr nonlinearity leads to interaction between UP and LP. It is shown that a UP wave packet can propagate through the crystal as a soliton and provide a "potential" within which the LP is trapped. Numerical calculations for CuCl crystal, in the spectral region of the lowest exciton resonance, have been performed to examine the conditions under which this trapping effect can take place.

The concept of polariton solitons has been developed some years ago.[1–3] Since then, a great deal of theoretical work has been done focusing on the formation and propagation of polariton solitons in different physical settings including bulk crystals, nonlinear waveguided structures, and crystal surfaces.[4–9] However, only a little attention has been paid to the impact of other possible types of intrinsic excitations in solids on the formation of polariton solitons. This problem is particularly important in the spectral regions near exciton resonances where spatial dispersion in crystals leads to the existence of additional light waves.[10]

In this paper, we discuss optical pulse propagation in the spectral region above the frequency of a longitudinal exciton ω_L, where two different polaritons—the upper branch polariton (UP) and the lower branch polariton (LP) propagate through a crystal at the same frequency (see Fig. 1). Both types of polaritons have been observed in many crystals.[11]

In the *nonlinear* regime, the interaction between UP and LP, which are generated simultaneously by an initial laser pulse, can lead to a coupled propagation of the UP and LP wave packets through a crystal. As shown below, when third-order nonlinearity is included, the UP wave packet can form a soliton and provide a "potential" within which the LP is trapped.

To describe optical pulse propagation in the spectral region of an exciton resonance, we adopt a semiclassical approach. In this approach, a nonlinear material equation which relates the induced excitonic polarization $\mathbf{P}(z,t)$ and the electric field $\mathbf{E}(z,t)$ (propagating along the z axis) in a nongyrotropic cubic Kerr-type crystal can be written as[9]

$$\left\{\frac{\partial^2}{\partial t^2}+\gamma\frac{\partial}{\partial t}+\omega_T^2-\frac{\hbar\omega_T}{\mu}\frac{\partial^2}{\partial z^2}\right\}\mathbf{P}(z,t)+\chi_0|\mathbf{P}(z,t)|^2\mathbf{P}(z,t)=\alpha\mathbf{E}(z,t)\ , \quad (1)$$

where $\alpha=\varepsilon_0(\omega_L^2-\omega_T^2)/4\pi$, ε_0 is the background dielectric constant of the medium, ω_T and ω_L are the frequencies of transverse and longitudinal excitons, respectively, μ is the exciton effective mass, γ is the damping constant, and χ_0 is a nonlinear coefficient ($\chi_0>0$).

Note that Eq. (1) determines only that part of the polarization which is due to the exciton transition of interest.

For simplicity, we neglect any damping processes in our system and suppress the dissipation term $\gamma(\partial\mathbf{P}(z,t)/\partial t)$ in the following discussion.

Equation (1) together with the wave equation for the propagation of a plane wave packet in an isotropic dispersive medium

$$\frac{\partial^2\mathbf{E}(z,t)}{\partial z^2}-\frac{\varepsilon_0}{c^2}\frac{\partial^2\mathbf{E}(z,t)}{\partial t^2}=\frac{4\pi}{c^2}\frac{\partial^2\mathbf{P}(z,t)}{\partial t^2} \quad (2)$$

gives the complete system of equations describing the

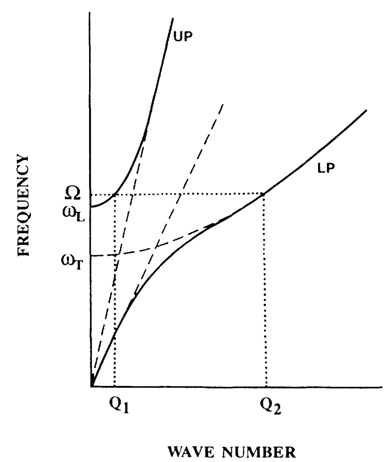

FIG. 1. Qualitative depiction of the exciton-polariton dispersion relation, $\Omega(Q)$ from Eq. (5), in the presence of spatial dispersion. Shown is the upper transverse polariton branch UP and the lower transverse polariton branch LP. ω_T and ω_L are the transverse and longitudinal exciton frequencies, respectively. The straight and curved dashed lines indicate the photonlike and excitonlike asymptotes.

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dispersive and nonlinear properties of a solid in the frequency range investigated.

To solve the system of Eqs. (1) and (2) for the polarization, we take Fourier transforms in space and time to obtain

$$F(k,\omega)\mathbf{P}(k,\omega)=-\chi_0\mathbf{P}^{\mathrm{NL}}(k,\omega)\ , \tag{3}$$

where

$$F(k,\omega)=\omega_T^2-\omega^2+\frac{\hbar\omega_T k^2}{\mu}-\frac{4\pi}{c^2}\frac{\alpha\omega^2}{(k^2-\varepsilon_0\omega^2/c^2)}\ , \tag{4a}$$

$$\mathbf{P}(k,\omega)=\frac{1}{(2\pi)^2}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\mathbf{P}(z,t)\times\exp[-i(kz-\omega t)]dz\,dt\ , \tag{4b}$$

$$\mathbf{P}^{\mathrm{NL}}(k,\omega)=\frac{1}{(2\pi)^2}\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\mathbf{P}(z,t)|\mathbf{P}(z,t)|^2\times\exp[-i(kz-\omega t)]dz\,dt\ . \tag{4c}$$

We assume that the nonlinearity is weak, so that the values of k and ω at which Eq. (3) must be solved are close to those in the linear case:

$$F(k=Q,\omega=\Omega)=0\ . \tag{5}$$

Equation (5) determines the dispersion relations for both polariton branches UP and LP in the linear crystal (see Fig. 1): $Q_1=Q_1(\Omega)$ and $Q_2=Q_2(\Omega)$. Note that below the subscript 1 refers to UP and the subscript 2 refers to LP.

In the vicinity of Ω and Q_j $(j=1,2)$, the function $F(k,\omega)$ can be explained in a Taylor series up to the second-order terms (the second-order approximation of dispersion theory):

$$\begin{aligned}F(k,\omega)=&(\omega-\Omega)\left[\frac{\partial F}{\partial\omega}\right]_{\substack{\omega=\Omega\\k=Q_j}}+(k-Q_j)\left[\frac{\partial F}{\partial k}\right]_{\substack{\omega=\Omega\\k=Q_j}}\\&+\frac{1}{2}(\omega-\Omega)^2\left[\frac{\partial^2F}{\partial\omega^2}\right]_{\substack{\omega=\Omega\\k=Q_j}}\\&+(\omega-\Omega)(k-Q_j)\left[\frac{\partial^2F}{\partial\omega\,\partial k}\right]_{\substack{\omega=\Omega\\k=Q_j}}\\&+\frac{1}{2}(k-Q_j)^2\left[\frac{\partial^2F}{\partial k^2}\right]_{\substack{\omega=\Omega\\k=Q_j}}\ .\end{aligned}\tag{6}$$

We introduce the designations

$$U_j=-\left[\frac{\partial F}{\partial k}\right]_{\substack{\omega=\Omega\\k=Q_j}}\Bigg/\left[\frac{\partial F}{\partial\omega}\right]_{\substack{\omega=\Omega\\k=Q_j}}\ , \tag{7a}$$

$$\begin{aligned}k_j''=\left[\frac{\partial^2k}{\partial\omega^2}\right]_{\substack{\omega=\Omega\\k=Q_j}}=-\Bigg\{&\left[\frac{\partial^2F}{\partial\omega^2}\right]_{\substack{\omega=\Omega\\k=Q_j}}\left[\frac{\partial F}{\partial k}\right]^2_{\substack{\omega=\Omega\\k=Q_j}}-2\left[\frac{\partial^2F}{\partial\omega\,\partial k}\right]_{\substack{\omega=\Omega\\k=Q_j}}\left[\frac{\partial F}{\partial k}\right]_{\substack{\omega=\Omega\\k=Q_j}}\left[\frac{\partial F}{\partial\omega}\right]_{\substack{\omega=\Omega\\k=Q_j}}\\&+\left[\frac{\partial^2F}{\partial k^2}\right]_{\substack{\omega=\Omega\\k=Q_j}}\left[\frac{\partial F}{\partial\omega}\right]^2_{\substack{\omega=\Omega\\k=Q_j}}\Bigg\}\Bigg/\left[\frac{\partial F}{\partial k}\right]^3_{\substack{\omega=\Omega\\k=Q_j}}\ ,\end{aligned}\tag{7b}$$

where U_j and k_j'' characterize the group velocity and the group dispersion of a polariton, respectively.

We seek a solution of Eq. (3) in the form

$$\mathbf{P}(z,t)=\widetilde{\mathbf{P}}_1(z,t)\exp[i(Q_1z-\Omega t)]+\widetilde{\mathbf{P}}_2(z,t)\exp[i(Q_2z-\Omega t)]\ , \tag{8}$$

where Ω is a carrier frequency, $Q_1(\Omega)$ and $Q_2(\Omega)$ are determined by Eq. (5), and the complex amplitudes $\widetilde{\mathbf{P}}_1(z,t)$ and $\widetilde{\mathbf{P}}_2(z,t)$ are slowly varying functions.

Using Eqs. (6)–(8) and the same technique as in Ref. 9 to solve Eq. (3), we obtain the following system of coupled equations:

$$i\left[\frac{\partial\widetilde{\mathbf{P}}_1}{\partial z}+\frac{1}{U_1}\frac{\partial\widetilde{\mathbf{P}}_1}{\partial t}\right]-\frac{k_1''}{2}\frac{\partial^2\widetilde{\mathbf{P}}_1}{\partial t^2}+\mathbf{O}_1(z,t)+\chi_1|\mathbf{P}_1+\mathbf{P}_2|^2\widetilde{\mathbf{P}}_1=0\ , \tag{9a}$$

$$i\left[\frac{\partial\widetilde{\mathbf{P}}_2}{\partial z}+\frac{1}{U_2}\frac{\partial\widetilde{\mathbf{P}}_2}{\partial t}\right]-\frac{k_2''}{2}\frac{\partial^2\widetilde{\mathbf{P}}_2}{\partial t^2}+\mathbf{O}_2(z,t)+\chi_2|\mathbf{P}_1+\mathbf{P}_2|^2\widetilde{\mathbf{P}}_2=0\ , \tag{9b}$$

where

$$\mathbf{O}_j(z,t)=W_j\left[\frac{\partial^2\widetilde{\mathbf{P}}_j}{\partial z^2}-\frac{1}{U_j^2}\frac{\partial^2\widetilde{\mathbf{P}}_j}{\partial t^2}\right]+V_j\left[\frac{\partial^2\widetilde{\mathbf{P}}_j}{\partial z\,\partial t}+\frac{1}{U_j}\frac{\partial^2\widetilde{\mathbf{P}}_j}{\partial t^2}\right]\ , \tag{10a}$$

$$W_j=\left[\frac{\partial^2F}{\partial k^2}\right]_{\substack{\omega=\Omega\\k=Q_j}}\Bigg/2\left[\frac{\partial F}{\partial k}\right]_{\substack{\omega=\Omega\\k=Q_j}},$$

$$V_j=-\left[\frac{\partial^2F}{\partial k\,\partial\omega}\right]_{\substack{\omega=\Omega\\k=Q_j}}\Bigg/\left[\frac{\partial F}{\partial k}\right]_{\substack{\omega=\Omega\\k=Q_j}},$$

$$\chi_j=\chi_0\Bigg/\left[\frac{\partial F}{\partial k}\right]_{\substack{\omega=\Omega\\k=Q_j}}\ . \tag{10b}$$

To solve the system (9) we assume that one of the fields, namely, $\mathbf{P}_1(z,t)$, is strong and the other field, $\mathbf{P}_2(z,t)$, is weak. This is the generally observed case in the transmitted light pulse experiments in the spectral region above ω_L.[12] Then, we can put $|\mathbf{P}_1+\mathbf{P}_2|^2 \cong |\mathbf{P}_1|^2$ in Eqs. (9a) and (9b). In the moving coordinate system ($\xi=z$, $\eta=t-z/U_1$), the system (9) becomes

$$i\frac{\partial \tilde{\mathbf{P}}_1}{\partial \xi}+\frac{|k_1''|}{2}\frac{\partial^2 \tilde{\mathbf{P}}_1}{2\eta^2}+\mathbf{O}_1(\xi,\eta)+\chi_1|\tilde{\mathbf{P}}_1|^2\tilde{\mathbf{P}}_1=0\ , \tag{11a}$$

$$i\frac{\partial \tilde{\mathbf{P}}_2}{\partial \xi}+\frac{|k_2''|}{2}\frac{\partial^2 \tilde{\mathbf{P}}_2}{\partial\eta^2}+i\delta\frac{\partial \tilde{\mathbf{P}}_2}{\partial \eta}+\mathbf{O}_2(\xi,\eta)+\chi_2|\tilde{\mathbf{P}}_1|^2\tilde{\mathbf{P}}_2=0\ , \tag{11b}$$

where

$$\mathbf{O}_1(\xi,\eta)=W_1\frac{\partial^2 \tilde{\mathbf{P}}_1}{\partial \xi^2}+R_1\frac{\partial^2 \tilde{\mathbf{P}}_1}{\partial \xi\,\partial\eta}\ , \tag{12a}$$

$$\mathbf{O}_2(\xi,\eta)=W_2\frac{\partial^2 \tilde{\mathbf{P}}_2}{\partial \xi^2}+R_2\left[\delta\frac{\partial^2 \tilde{\mathbf{P}}_2}{\partial \eta^2}+\frac{\partial^2 \tilde{\mathbf{P}}_2}{\partial \xi\,\partial\eta}\right]\ , \tag{12b}$$

$\delta=U_2^{-1}-U_1^{-1}$ is the group velocity mismatch, $R_j=V_j-2W_j/U_j$, and we take into account that in the spectral region of interest $k_1''<0$, $k_2''<0$ and $\chi_1>0$, $\chi_2>0$.

Equation (11a) has a soliton solution in the form (see Ref. 9)

$$\tilde{\mathbf{P}}_1(\xi,\eta)=\varphi_0 \operatorname{sech}(\eta/\tau)\exp[i(\nu_1\xi+\beta_1\eta)]\ , \tag{13}$$

where the soliton duration τ, and its amplitude φ_0 are related as

$$|k_1''|/\tau^2=\chi_1\varphi_0^2\ . \tag{14}$$

The expressions for ν_1 and β_1 are given in Ref. 9, Eqs. (18a) and (18b).

Seeking a solution of Eq. (11b) in the form

$$\tilde{\mathbf{P}}_2(\xi,\eta)=\varphi_2(\eta)\exp\left[i(\nu_2\xi+\beta_2\eta)\right]\ , \tag{15}$$

and substituting Eq. (13) into Eq. (11b), we finally obtain the linear Schrödinger equation for $\varphi_2(\eta)$:

$$\frac{\partial^2\varphi_2(\eta)}{\partial\eta^2}+\frac{2}{(|k_2''|+2R_2\delta)}\left[E+\frac{\chi_2(\varphi_0)^2}{ch^2(\eta/\tau)}\right]\varphi_2(\eta)=0\ , \tag{16}$$

where

$$E=-\left[\left[\frac{|k_2''|}{2}+R_2\delta\right]\beta_2^2+(\delta+R_2\nu_2)\beta_2+W_2\nu_2^2+\nu_2\right]\ , \tag{17a}$$

$$\beta_2=-\frac{(\delta+R_2\nu_2)}{|k_2''|+2R_2\delta}\ . \tag{17b}$$

The spectrum of solutions of Eq. (16) is continuous at positive E values and discrete at negative E values. We are interested in bound solutions of Eqs. (11a) and (11b), as these represent a LP polariton "captured" by an UP polariton, therefore we consider the discrete spectrum which has the form[13]

$$E_n=-\frac{(|k_2''|+2R_2\delta)}{8\tau^2}[-(1+2n)+\sqrt{1+8G}\,]^2\ , \tag{18}$$

where

$$n=0,1,\ldots<S\ , \tag{19a}$$

$$S=\tfrac{1}{2}[-1+\sqrt{1+8G}\,]\ , \tag{19b}$$

$$G=\frac{|k_1''|}{(|k_2''|+2R_2\delta)}\left[\frac{\partial F}{\partial k}\right]_{\substack{\omega=\Omega\\ k=Q_1}}\Bigg/\left[\frac{\partial F}{\partial k}\right]_{\substack{\omega=\Omega\\ k=Q_2}}\ . \tag{19c}$$

The discrete energy levels of Eq. (18) can be considered as the localized states of the LP in the potential well created by the UP soliton. It is interesting that the number of discrete energy levels, S, is independent from the parameters of the UP soliton; in fact, S is determined only by the dispersive characteristics of the medium. The values of $S(\Omega)$ have been calculated numerically for CuCl crystal, in the spectral region of the lowest exciton resonance. The material parameters required for calculations were taken from Ref. 12. Results are presented in Fig. 2.

A restriction on ν_2 follows from the condition $E<0$ [see Eqs. (17a) and (17b)]:

$$\nu_-<\nu_2<\nu_+\ , \tag{20}$$

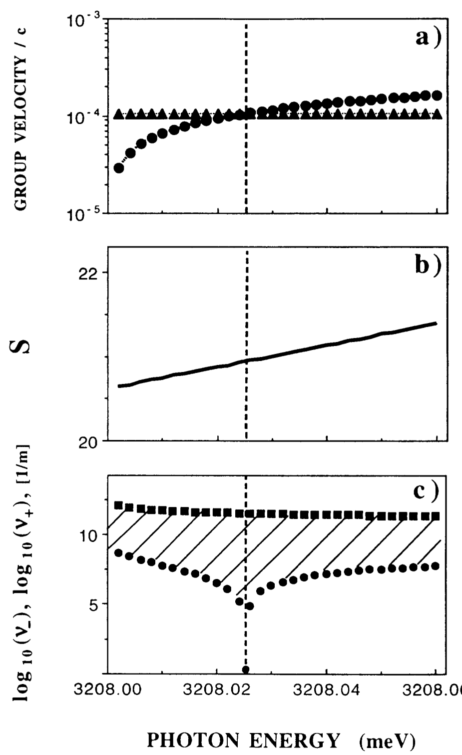

FIG. 2. The spectral region above the lowest exciton resonance in CuCl: $\hbar\omega_T=3.2025$ eV, $\hbar\omega_L=3.2080$ eV, $\mu=2m_e$, $\varepsilon_0=5$ (Ref. 12). As the functions of photon energy we show: (a) the group velocity of UP (●) and LP (▲) normalized by c. The dashed line corresponds to the case $U_1=U_2$; (b) The number of discrete energy levels, S; (c) The values of $\log_{10}(\nu_-)$ (circles) and $\log_{10}(\nu_+)$ (squares). The region where the inequalities $\nu_-<\nu_2<\nu_+$ [see Eq. (21)] are fulfilled is shaded.

where

$$\nu_{\pm}=\frac{[|k_2''|+R_2\delta\pm\sqrt{|k_2''|(|k_2''|+2R_2\delta)}]}{[R_2^2-2W_2(|k_2''|+2R_2\delta)]} . \tag{21}$$

Existence of the bound solutions of Eqs. (11a) and (11b) is most probable in the case when the group velocities of the UP and LP become equal, $U_1(\Omega)=U_2(\Omega)$, and, as a consequence, $\delta=0$. If $U_1\neq U_2$, the conditiion (20) must be satisfied for the discrete spectrum of Eq. (18) to appear. Beyond the region defined by the inequalities (20), E values of Eq. (17a) are positive and the spectrum of solutions of Eq. (16) is continuous.[13] This corresponds to independent propagation of the UP and LP wave packets through a crystal. The spectral range where formation of the UP-LP bound states can take place in CuCl is illustrated in Fig. 2.

It is interesting to note that the possibility of coupled propagation of two optical pulses with different polarizations has been recently predicted for birefringent optical fibers.[14] While linear birefringence leads to splitting of a single injected soliton into two separating solitons with different polarizations and group velocities, this splitting could be eliminated by the Kerr nonlinearity. This phenomenon is called "soliton trapping" and discussed in several papers.[15–17] The influence of one wave on another in the presence of nonlinearity has also been discussed in related contexts.[18,19]

Although the physical situation considered in the present paper is different from the soliton trapping effect in birefringent optical fibers, both our model [see Eqs. (11a) and (11b)] and the soliton trapping problem (see Ref. 17) are related to the Manakov model.[20]

The effect of polariton trapping by a soliton should be observed in time-resolved transmission experiments similar to those which have been reported, for example, in Ref. 12 or, more recently, in Ref. 21. At photon energies just above $\hbar\omega_L$, a laser pulse excites two different polariton wave packets corresponding to UP and LP which propagate through the crysal at slightly different group velocities (see Fig. 4 in Ref. 12). The time delay between the transmitted UP and LP signals has been measured and the corresponding group velocities have been calculated. As the input laser power is increased, the nonlinear propagation regime described above will lead to a decrease in the time delay between the two transmitted pulses. It is possible that at sufficiently high power, only one output signal (with some amplitude modulation) would be observed instead of the two signals observed in the linear regime.

In conclusion, in the frequency region above the frequency ω_L, the UP wave packet can propagate through the crystal as a soliton and provide a "potential" within which the lower branch polariton is trapped. The energy spectrum of the "captured" LP is characterized by discrete energy levels [see Eq. (18)]; these levels correspond to localized states of the LP in the potential well created by the UP soliton.

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