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Exciton-polariton solitary waves

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Abstract. Effects of the exciton and polariton dispersions and the nonlinear exciton and photon interactions on the properties of polariton solitons in molecular crystals are investigated. Higher-order terms and phase-modulation (chirp) are taken into account. Bright- and dark-soliton solutions of the resulting modified nonlinear Schrödinger (NLS) equation are presented. Nonlinearity- and dispersion-induced critical points on the polariton dispersion curve are obtained, separating regions with different solutions.

PACS. 71.36.+c Polaritons – 71.35.-y Excitons and related phenomena

Optical solitons have been extensively studied [1,2] due to their importance for fiber-optics communications. Most investigations refer to frequencies far the resonances of the medium. Near resonance, the electromagnetic field couples to the corresponding polar excitations to form mixed states known as polaritons. Polaritons exhibit strong dispersion in the resonance region which changes continuously from photon- to exciton-type on the lower branch and from exciton- to photon-type on the upper branch. The nonlinear interactions in the polariton region originate from both the quasiparticle and the photon subsystems, and together with the dispersion lead to the formation of polariton solitary waves [3–12]. Dispersion effects in the spectra of phonon-polariton solitons have been studied in [8] and for exciton solitons–in [13]. In the present work we investigate the properties of exciton-polariton solitons, taking into account the dynamical (Coulomb) and kinematical (statistical) exciton-exciton interactions as well as the nonresonant (Kerr) nonlinear optical susceptibility of the medium. This leads to higher-order dispersion terms in the underlying nonlinear equations which govern the formation of exciton-polariton solitons.

We start with the Hamiltonian of a system of Frenkel electronic excitons in a molecular crystal interacting with the electromagnetic field [8,13]:

$$H = \omega_0 \sum_n P_n^\dagger P_n - \sum_{n,m} V_{nm} P_n^\dagger P_m - A \sum_{n,m} P_n^\dagger P_m^\dagger P_n P_m - d \sum_n (P_n^\dagger E_n + P_n E_n^*) \tag{1}$$

where ω_0 is the intramolecular excitation energy ($\hbar = 1$) and $P_n^\dagger$ (P_n) are the corresponding creation (annihilation) Pauli operators of an electron-hole pair in the nth molecule. The second term in (1) describes the resonant intermolecular interaction, V_{nm} being the corresponding matrix elements. The term $\sim A$ describes the nonlinear dynamical interaction between excitations on neighbouring molecules which has a quadrupole character. d is the dipole moment matrix element for the exciton transitions and E_n and E_n^* are the complex-conjugate parts of the classical macroscopic electric field.

The Pauli operators obey mixed commutation relations:

$$[P_n, P_m^\dagger] = (1 - 2N_n)\delta_{n,m}, \quad [P_n, P_m] = 0$$
$$P_n^2 = (P_n^\dagger)^2 = 0, \qquad N_n \equiv P_n^\dagger P_n. \tag{2}$$

With the help of (2), the following equation of motion for the operators P_n is obtained:

$$\mathrm{i}\frac{\partial P_n}{\partial t} = \omega_0 P_n - (1 - 2N_n) \sum_m V_{nm} P_m - 2AP_n \sum_m N_m - d(1 - 2N_n)E_n. \tag{3}$$

In comparison with the case of vibrational excitons [8], equation (3) contains two additional nonlinear terms proportional to the local exciton density N_n, which have statistical nature associated with the Pauli commutation relations. The term $\sim N_n V_{nm}$ describes a kinematical repulsion between excitons on neighbouring molecules while the term $\sim N_n d$ describes the dipole moment saturation at high exciton densities.

The equation of motion of the averaged exciton amplitude $\langle P_n \rangle \equiv \alpha_n(t)$ in the low-density limit ($\langle N_n \rangle \simeq |\alpha_n|^2$) is:

$$\mathrm{i}\frac{\partial \alpha_n}{\partial t} = \omega_0 \alpha_n - (1 - 2|\alpha_n|^2) \sum_m V_{nm} \alpha_m - 2A\alpha_n \sum_m |\alpha_m|^2 - d(1 - 2|\alpha_n|^2)E_n. \tag{4}$$

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Equation (4) is to be complimented with Maxwell's wave equation, which provides a second (phenomenological) relation between the electric field and the induced polarization:

$$\left(\frac{\partial^2}{\partial x^2}-\frac{\partial^2}{\partial t^2}\right)E(x,t)=\\ 4\pi\frac{\partial^2}{\partial t^2}\left(\frac{d}{a^3}\alpha(x,t)+\chi^{(3)}|E(x,t)|^2E(x,t)\right). \quad (5)$$

The first term in the right-hand side of (5) describes the linear polarization associated with the exciton transitions (a is the lattice constant) and the second term – the nonresonant (Kerr-type) nonlinear polarization associated with other excitations with frequencies far from the exciton region (their contribution to the linear polarization can be accounted for by renormalizing the velocity of light which we assume to be $c = 1$). (4) and (5) is the set which describes the properties of coupled nonlinear excitons and photons.

We shall seek solutions of (4) and (5) in the standard form:

$$\alpha(x,t)=\mathrm{e}^{\mathrm{i}(kx-\omega t)}\varphi(x,t)\\ E(x,t)=\mathrm{e}^{\mathrm{i}(kx-\omega t)}\mathcal{E}(x,t) \quad (6)$$

where k and ω are the central wave-vector and frequency of the carrier wave. Using the nearest-neighbour approximation and taking the continuum limit in (4) we obtain:

$$\mathrm{i}\frac{\partial\varphi}{\partial t}=(\omega_k-\omega)\varphi-\mathrm{i}v_k(1-2|\varphi|^2)\frac{\partial\varphi}{\partial x}-b_ka^2\frac{\partial^2\varphi}{\partial x^2}\\ -4(A-b_k)|\varphi|^2\varphi-\mathrm{i}\frac{v_ka^2}{6}\frac{\partial^3\varphi}{\partial x^3}-d(1-2|\varphi|^2)\mathcal{E} \quad (7)$$

$$[(\omega^2-k^2)+2\mathrm{i}(k\frac{\partial}{\partial x}+\omega\frac{\partial}{\partial t})+(\frac{\partial^2}{\partial x^2}-\frac{\partial^2}{\partial t^2})]\mathcal{E}=\\ 4\pi(-\omega^2-2\mathrm{i}\omega\frac{\partial}{\partial t}+\frac{\partial^2}{\partial t^2})(\frac{d}{a^3}\varphi+\chi^{(3)}|\mathcal{E}|^2\mathcal{E}). \quad (8)$$

ω_k and v_k are the energy and the group velocity of the noninteracting excitons, and b_k is the group-velocity dispersion (GVD) parameter:

$$\omega_k=\omega_0-2b_k,\quad b_k=V\cos ak,\quad v_k=2Va\sin ak. \quad (9)$$

The coefficients (9) take account of the exciton dispersion throughout the whole Brillouin zone. In equation (7) we have kept the nonlinear dispersion term $\sim|\varphi|^2\partial\varphi/\partial x$ and the third-order linear dispersion term $\sim\partial^3\varphi/\partial x^3$. The first one stems from the nonlocal character of the kinematical exciton-exciton interaction and it is similar to the self-steepening term in the generalized NLS equation in optics [1,2]. The second one has the same order of magnitude and it is particularly important for short pulses near the zero-GVD points.

We shall consider $\varphi(x,t)$ and $\mathcal{E}(x,t)$ to be slowly-varying complex functions of the running variable $\xi=x-vt$ ($|\partial\varphi/\partial x|\ll k\varphi,\quad|\partial\varphi/\partial t|\ll\omega\varphi$ and v is the velocity of the solitary wave), thus taking account of both amplitude and phase modulation (chirp). Expressing the electric field from (7) as a function of the polarization, using the small-amplitude approximation $(1-2|\varphi|^2)^{-1}\simeq1+2|\varphi|^2$, substituting it in (8) and keeping first-order nonlinear dispersion and third-order linear dispersion terms, we end up with a modified NLS equation for the polarization:

$$D\varphi-M\varphi''-\Gamma|\varphi|^2\varphi-\mathrm{i}(S-T|\varphi|^2)\varphi'\\ +\mathrm{i}Q\varphi^2\varphi*'-\mathrm{i}R\varphi'''=0 \quad (10)$$

where

$$D=\omega_k-\omega-\frac{\Omega_0\omega^2}{k^2-\omega^2}$$
$$M=a^2b_k+\frac{2(k-\omega v)(v_k-v)+(1-v^2)(\omega_k-\omega)-\Omega_0v^2}{k^2-\omega^2}$$
$$\Gamma=4A-2(\omega_0-\omega)+\chi\frac{\omega^2(\omega_k-\omega)^3}{k^2-\omega^2}$$
$$S=(v_k-v)+\frac{[2(k-\omega v)(\omega_k-\omega)-2\Omega_0\omega v]}{k^2-\omega^2}$$
$$Q=\{4(2A-\omega_0+\omega)(k-\omega v)\\ +\chi\omega(\omega_k-\omega)^2[2v(\omega_k-\omega)-\omega(v_k-v)]\}/(k^2-\omega^2)$$
$$T=2v+\{8(2A-\omega_0+\omega)(k-\omega v)\\ +2\chi\omega(\omega_k-\omega)^2[2v(\omega_k-\omega)+\omega(v_k-v)]\}/(k^2-\omega^2)$$
$$R=\frac{v_ka^2}{6}-\frac{2(k-\omega v)b_ka^2+(1-v^2)(v_k-v)}{k^2-\omega^2}$$
$$\Omega_0=\frac{4\pi d^2}{a^3},\quad\chi=\frac{4\pi}{d^2}\chi^{(3)}. \quad (11)$$

Equation (10) governs the formation and the properties of chirped exciton-polariton solitons. Note that the coefficients (11) contain terms $\sim(k^2-\omega^2)^{-1}$ which are dominant in the photonlike parts of the spectrum and in the resonance region, where $k\sim\omega$ and they can be neglected in the excitonlike part of the spectrum where $k\gg\omega$. For $T=Q=R=0$ (10) reduces to the standard NLS equation whose solutions have been extensively studied.

Solitary-wave analytical solutions of equation (10) can be looked for in the factorized form:

$$\varphi(\xi)=\rho(\xi)\mathrm{e}^{\mathrm{i}\Phi(\xi)} \quad (12)$$

where $\rho(\xi)$ is the amplitude and $\Phi(\xi)$ is the nonlinear contribution to the phase. Separating the real and the imaginary parts of (10), the following system of coupled nonlinear equations is obtained:

$$(D+M\Phi'^2)\rho-(M-3R\Phi')\rho''-\Gamma\rho^3\\ +[S-(T-Q)\rho^2]\rho\Phi'+3R\rho'\Phi''+R\rho\Phi'''=0 \quad (13)$$

$$M\rho\Phi''+(S+2M\Phi')\rho'-(T+Q)\rho^2\rho'+R\rho'''=0. \quad (14)$$

To solve equations (13, 14) we can approximate the third-derivative ρ''' by the following expression derived from the cubic NLS equation:

$$\rho''' = (D - 3\Gamma\rho^2)\rho'/M. \qquad (15)$$

With the help of (15), the equation for the phase (14) can be integrated to give:

$$\Phi' = -\frac{SM + RD}{2M^2} + \frac{(T+Q)M + 3R\Gamma}{4M^2}\rho^2. \qquad (16)$$

We shall look for solutions with fixed central wavenumber, for which the constant term in (16) must vanish *i.e.*

$$SM + RD = 0. \qquad (17)$$

Equation (17) generalizes the standard equation for the velocity of the polariton solitary waves $S = 0$ by taking into account the third-order dispersion.

Substituting the phase derivatives from (16) into (13), we end up with an equation for the amplitude ρ which contains third- and fifth-order nonlinear terms:

$$D\rho - M\rho'' - G\rho^3 - F\rho^5 = 0 \qquad (18)$$

where

$$G = \Gamma - \frac{3RD}{M^3}[(T+Q)M + 3R\Gamma]$$
$$F = \frac{(3T - 5Q)M + 33R\Gamma}{16M^3}[(T+Q)M + 3R\Gamma]\cdot \qquad (19)$$

(18) is known as the cubic-quintic NLS equation which has been obtained and investigated in some detail in optics [14,15]. The quintic nonlinear term originates exclusively from the higher-order dispersion terms in (10, 13, 14). The latter also contribute to the cubic nonlinear coefficient G. The physics of the exciton-polariton solitary waves is controlled by the coefficients in (18). They are complicated functions of the frequency and the wave-vector and govern the type of the solitary-wave solutions.

Equation (18) possesses the following bright-soliton solution:

$$\rho(\xi) = \rho_0\sqrt{\frac{1+B}{1+B\cosh(2\xi/L)}}, \quad B > 0 \qquad (20)$$

where

$$\omega = \omega_k - \frac{G\rho_0^2(1+B)}{4} - \frac{\Omega_0\omega^2}{k^2-\omega^2}$$
$$L^2 = \frac{4M}{G\rho_0^2(1+B)} \qquad (21)$$

and

$$B = 1 + \frac{4F\rho_0^2}{3G}\cdot \qquad (22)$$

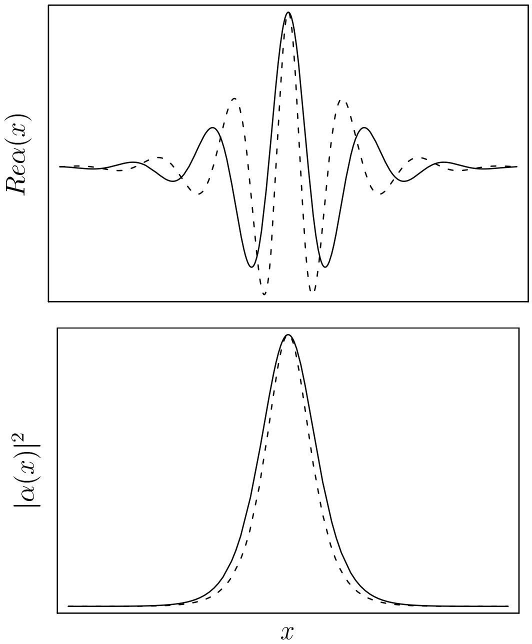

Fig. 1. Bright-soliton solution (20) for $F = 0$ (solid line) and for $F \neq 0$ and $B > 1$ (dashed line).

From (21) it follows that bright-solitons are only possible for $M/G > 0$. For $B = 1$ ($F = 0$) (20) coincides with the corresponding solution of the cubic NLS equation.

The dark-soliton solution of (18) has the form:

$$\rho(\xi) = \rho_1\frac{\sqrt{B}\sinh\xi/L}{\sqrt{1+B\sinh^2\xi/L}}, \quad B > 0 \qquad (23)$$

where

$$\omega = \omega_k - G\rho_1^2\frac{1-3B}{2(2-3B)} - \frac{\Omega_0\omega^2}{k^2-\omega^2}$$
$$L^2 = \frac{2M(2-3B)}{G\rho_1^2} \qquad (24)$$

and

$$B = \frac{3G + 4F\rho_1^2}{3(G + 2F\rho_1^2)}\cdot \qquad (25)$$

For $B = 1$ (23) coincides with the dark-soliton solution of the NLS equation. The relation (24) opens up two possibilities for the existence of dark-soliton solutions (23): $B > 2/3$ implies $M/G < 0$ which is the usual condition for dark-soliton solutions of the cubic NLS equation. For $B < 2/3$ however, dark solitons exist for $M/G > 0$, which is the condition for the existence of bright-solitons too. Thus the regions with bright- and dark-soliton solutions of equation (18) are not completely separated. The analysis of the coefficients (19) shows, that bright and dark solitons can coexist only in very narrow regions near the zeroes of G, where G and F have different signs.

The real part and the squared modulus of the bright-soliton solution (20) (dashed line) are shown in Figure 1.

For $B > 1$ the quintic term leads to a blue shift (chirp) of the central carrier-wave frequency and a decrease of the soliton's width, compared to the solution of the cubic NLS equation (solid line). $B < 1$ yields a red frequency shift and an increase of the soliton's width.

As mentioned above, the coefficients in (18) govern the type of the solitary-wave solutions and their parameters. The type of the soliton solution depends strongly on the sign of the ratio M/G. Except for the zero GVD points $M = 0$, where all approximations are violated, the nonlinear coefficient G differs from Γ by a small parameter and the analysis can be carried out using $G \simeq \Gamma$.

With the help of the linear polariton dispersion relation $D = 0$, the nonlinear coefficient G can be approximated by:

$$G \simeq \Gamma \simeq 2(\omega - \omega_0 + 2A) + \frac{\Omega_0^3 \chi}{(1 - k^2/\omega^2)^4} \cdot \qquad (26)$$

The first term in (26) originates from the exciton-exciton interaction and it is positive for $\omega > \omega_0 - 2A$ and negative for $\omega < \omega_0 - 2A$. It is dominant in the excitonic part of the spectrum (for $k \gg \omega$). The second (Kerr-type) term in (25) is always positive ($\chi > 0$) and due to the strong resonance at $k \simeq \omega$ it is dominant in the photon-like parts of the spectrum. It decreases very rapidly for small deviation from the light-line and the two terms cancel each other in the resonance region at points k_1 and k_2 on the upper and lower polariton branches respectively (Fig. 2). The nonlinear coefficient G has a third zero k_4 in the excitonic part of the spectrum for $\omega \simeq \omega_0 - 2A$.

The GVD coefficient M plays the role of an inverse polariton effective mass and includes the effects of both the polariton- and the exciton-type dispersion. The polariton-type dispersion dominates in the photonlike parts of the spectrum and in the resonance region (Fig. 2). In these regions M is positive on the upper branch ($k < \omega$) and negative on the lower branch ($k > \omega$). $|M|$ decreases on the lower branch with the increase of the wave-number and the GVD coefficient M vanishes at a critical point k_3 which reflects the change from a polariton- to an exciton-type dispersion. M has a second zero in the middle of the Brillouin zone at $k_5 \simeq \pi/2a$, where the exciton effective mass changes sign. Near the zero-GVD points k_3 and k_5, the nonlinear coefficients in (18) diverge and these regions should be excluded from the analysis. This is a result of the approximation (15) which correlates higher-order dispersion and nonlinear terms.

Keeping in mind the signs of G and M, we can determine the regions corresponding to bright- and dark-soliton solutions along the polariton dispersion curve (Fig. 2). On the upper branch, dark solitons exist for $k < k_1$ and bright solitons – for $k > k_1$. On the lower branch five regions with different type of solutions are formed: dark solitons exist in the regions $0 < k < k_2$, $k_3 < k < k_4$ and $k > k_5$; and bright solitons – for $k_2 < k < k_3$ and $k_4 < k < k_5$. The zero-nonlinearity critical point k_4 is shifted from the zero GVD point k_5 due to the presence of the dynamical interaction $\sim A$. For $A = 0$, $k_4 \equiv k_5$ and this yields dark-soliton solutions in the whole exciton-like part of the spectrum ($k > k_3$).

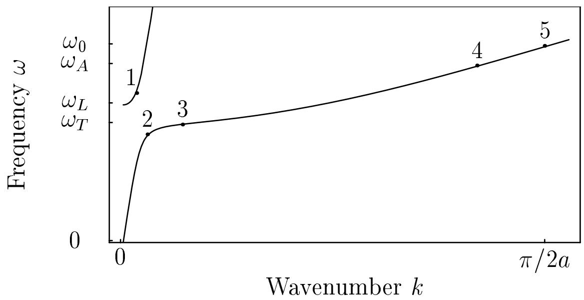

Fig. 2. Critical points on the exciton-polariton dispersion curves which separate bright- from dark-soliton solutions. $\Omega_0 = 0.1\omega_0$, $V = 0.2\omega_0$ and $A = 0.05\omega_0$. $\omega_T = \omega_0 - 2V$, $\omega_L = \omega_T + \Omega_0$ and $\omega_A = \omega_0 - 2A$.

The present investigation shows that the properties of polariton solitary waves far from the excitonic resonances (where polaritons are nearly photons) are governed by the polariton-type dispersion (which has different signs on the two branches) and the Kerr-type optical nonlinearity. This opens up new channels for control of the properties of optical solitons in these regions. The changes of the type of the solution at k_1 and k_2 which are in the resonance region can have practical applications in optical switching devices.

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