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LETTER TO THE EDITOR

Dispersion effects in exciton–polariton solitons

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Abstract

The role of the exciton and polariton dispersion and the Pauli statistics in the properties of exciton–polariton solitons in molecular crystals is investigated. The formation of bright- and dark-soliton solutions in different regions of the spectrum is studied.

The formation and propagation of coherent exciton–photon pulses with constant shape (polariton solitons), considered as self-induced transparency of excitons, were studied in the early 1970s [1, 2]. Since then, extensive investigations of the properties of polariton solitons have been carried out [3–8]. Spatial dispersion effects have been studied in [4–6, 9]. The formation of polariton solitons on crystal surfaces [10] and as a result of complicated interactions [11–15] has been considered. Dispersion effects in the spectra of phonon–polariton solitons have been studied in detail in [9]. In the present letter we investigate specific dispersion and non-linear effects in the properties of exciton–polariton solitons in molecular crystals.

We shall consider the interaction of electromagnetic pulses with Frenkel electronic excitons in cubic molecular crystals with one molecule per unit cell, taking into account only the ground and the first excited state of the molecule. The exciton Hamiltonian can be written as [16, 17]

$$H_{ex} = \hbar\omega_0 \sum_n P_n^\dagger P_n - \sum_{n,m} V_{nm} P_n^\dagger P_m - A \sum_{n,m} P_n^\dagger P_m^\dagger P_n P_m \tag{1}$$

where $\hbar\omega_0$ is the intramolecular excitation energy and $P_n^\dagger$ (P_n) are the corresponding creation (annihilation) Pauli operators of an electron–hole pair in the nth molecule. They obey the commutation relations

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

which combine commutation on neighbouring molecules with anticommutation on one and the same molecule and prohibit the localization of more than one excitation on a single molecule. The second term in (1) describes the resonant intermolecular interaction, where V_{nm} are the corresponding matrix elements. The intramolecular vibrations usually have narrow energy bands ($|V_{nm}| \ll \hbar\omega_0$) and hence, in accordance with the Heitler–London approximation, only terms conserving the total number of the quasiparticles are retained. The term $\sim A$ describes

the non-linear dynamical interaction between excitations on neighbouring molecules. It has a different form as compared to the intramolecular anharmonicity term in the case of vibrational excitations [9], where the two bosons are localized on one and the same molecule. The direct dynamical exciton–exciton interaction has a quadrupole character and it is usually negative and much weaker than the resonant intermolecular interaction ($|A| \ll |V|$). The excitons however may also interact in an indirect way, exchanging other quasiparticles such as lattice phonons, which may result in a considerable increase of the effective non-linear exciton interaction or even a change of its sign.

In the dipole approximation the Hamiltonian of the interaction of the excitons with the electromagnetic field can be described in a semi-classical way as follows:

$$H_{int} = -d \sum_n (P_n^\dagger E_n^+ + P_n E_n^-) \tag{3}$$

where d is the dipole moment matrix element for transitions from the ground state to the excited state of the molecule and E_n^+ (E_n^-) is the positive-frequency (negative-frequency) part of the macroscopic electric field.

The equation of motion for the operators P_n is

$$\mathrm{i}\hbar \frac{\partial P_n}{\partial t} = \hbar\omega_0 P_n - (1 - 2N_n) \sum_m V_{nm} P_m - 2A P_n \sum_m N_m - d(1 - 2N_n) E_n^+. \tag{4}$$

In comparison with the case of vibrational excitons [9], equation (4) contains two additional non-linear terms proportional to the local exciton density N_n, which have statistical nature associated with the Pauli commutation relations (2). The term proportional to V_{nm} describes the so-called kinematical repulsion between excitons on neighbouring molecules. The term proportional to d describes the dipole moment quenching at high exciton densities.

We shall average equation (4) using a wave-function of the form [3]

$$|\Psi(t)\rangle = \prod_n (u_n(t) + v_n(t) P_n^\dagger)|0\rangle \qquad P_n|0\rangle = 0 \qquad |u_n|^2 + |v_n|^2 = 1 \tag{5}$$

which has been successfully applied in the theory of superconductivity to describe macroscopic coherent states. Note that (5) does not contain non-physical states with more than one excitation on a single molecule.

With the help of (2) and (5), the following decoupling relations can be established:

$$\begin{aligned} &\langle N_n P_m \rangle = \langle N_n \rangle \langle P_m \rangle \\ &\langle N_n \rangle = (1/2)(1 \mp \sqrt{1 - 4|\langle P_n \rangle|^2}). \end{aligned} \tag{6}$$

The two signs in (6) correspond to different exciton densities. The upper sign holds for $0 \leqslant \langle N_n \rangle \leqslant 1/2$ and the lower sign for $1/2 \leqslant \langle N_n \rangle \leqslant 1$. Equation (6) reflects the Pauli character of the excitations and describes the deviations of the average local density from the squared modulus of the averaged exciton operators. In the low-density limit ($\langle N_n \rangle \ll 1$) it yields

$$\langle N_n \rangle \approx |\langle P_n \rangle|^2 \tag{7}$$

which is the usual decoupling relation when the average is taken over Glauber's coherent states [18]. As they involve states with arbitrary occupation numbers, they are applicable to vibrational excitons (bosons) and to low-density electronic excitons which can be approximated by bosons. For higher exciton densities a wave-function of the type (5) should be used, which does not contain non-physical states with occupation numbers $n > 1$.

As (6) shows, the average exciton density $\langle N_n \rangle$ can deviate dramatically from $|\langle P_n \rangle|^2$, and in the high-density limit ($\langle N_n \rangle \sim 1$, $|\langle P_n \rangle|^2 \ll 1$) is given by

$$\langle N_n \rangle \approx 1 - |\langle P_n \rangle|^2. \tag{8}$$

An inverse behaviour of the density of excitations in the wave-function associated with $\langle P_n \rangle$ is observed in this case.

Averaging (4) with the help of (5) and (6) gives

$$i\hbar \frac{\partial \langle P_n \rangle}{\partial t} = \hbar\omega_0 \langle P_n \rangle - (1 - 2\langle N_n \rangle) \sum_m V_{nm} \langle P_m \rangle - 2A \langle P_n \rangle \sum_m \langle N_m \rangle - d(1 - 2\langle N_n \rangle) E_n^+. \tag{9}$$

In what follows we consider the low-density limit (7) and using the notation

$$\langle P_n \rangle \equiv \alpha_n(t) \tag{10}$$

equation (9) becomes

$$i\hbar \frac{\partial \alpha_n}{\partial t} = \hbar\omega_0 \alpha_n - (1 - 2|\alpha_n|^2) \sum_m V_{nm} \alpha_m - 2A\alpha_n \sum_m |\alpha_m|^2 - d(1 - 2|\alpha_n|^2) E_n^+. \tag{11}$$

We would like to point out that the wave-function (5) and the decoupling relations (6) allow the description of the dynamics of the electronic excitons by just equation (11), eliminating the need for a second equation for the exciton number as in e.g. references [3] and [6].

Equation (11) together with Maxwell's wave equation:

$$\left(\frac{\partial^2}{\partial x^2} - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right) E^+(x,t) = \frac{4\pi d}{c^2 a^3} \frac{\partial^2 \alpha(x,t)}{\partial t^2} \tag{12}$$

constitutes the set which describes the properties of coupled non-linear excitons and photons (c is the velocity of light and a the lattice constant).

Proceeding as in [9], we shall seek solutions of (11) and (12) in the form

$$\begin{aligned} \alpha(x,t) &= e^{i(kx - \omega t)} \varphi(x,t) \\ E^+(x,t) &= e^{i(kx - \omega t)} \mathcal{E}(x,t) \end{aligned} \tag{13}$$

where k and ω are the wave vector and the frequency of the carrier wave and $\varphi(x,t)$ and $\mathcal{E}(x,t)$ are real slowly varying functions. Using the semi-discrete and the nearest-neighbour approximations, which model the exciton dispersion over the whole Brillouin zone, we obtain

$$i\hbar \frac{\partial \varphi}{\partial t} = (\epsilon_k - \hbar\omega)\varphi - i\hbar v_k \frac{\partial \varphi}{\partial x} - V \cos ak \frac{\partial^2 \varphi}{\partial x^2} - 4(A - V \cos ak)\varphi^3 - d(1 - 2\varphi^2)\mathcal{E} \tag{14}$$

$$\begin{aligned} &\left[\left(\frac{\omega^2}{c^2} - k^2 \right) + 2i \left(k \frac{\partial}{\partial x} + \frac{\omega}{c^2} \frac{\partial}{\partial t} \right) + \left(\frac{\partial^2}{\partial x^2} - \frac{1}{c^2} \frac{\partial^2}{\partial t^2} \right) \right] \mathcal{E} \\ &\qquad = \frac{4\pi d}{c^2 a^3} \left(-\omega^2 - 2i\omega \frac{\partial}{\partial t} + \frac{\partial^2}{\partial t^2} \right) \varphi. \end{aligned} \tag{15}$$

ϵ_k and v_k are the energy and the velocity of the non-interacting excitons:

$$\epsilon_k = \hbar\omega_0 - 2V \cos ak \qquad v_k = \frac{2Va^2}{\hbar} \sin ak. \tag{16}$$

Looking for localized solutions in the form of pulses with constant shape, which depend on space and time through the running variable $\xi = x - vt$ (v is the velocity of the solitary wave), and combining (14) and (15), the following non-linear equation can be derived:

$$\chi_k \varphi - M_k \frac{\partial^2 \varphi}{\partial \xi^2} - A_k \varphi^3 - i P_k \frac{\partial \varphi}{\partial \xi} = 0 \tag{17}$$

where

$$\begin{aligned}
&\chi_k = \epsilon_k - \hbar\omega - \frac{\hbar\Omega_0\omega^2}{c^2k^2-\omega^2}\\
&M_k = a^2(V\cos ak + W)\\
&W \equiv [2\hbar(c^2k-\omega v)(v_k - v) + (c^2-v^2)(\epsilon_k - \hbar\omega) - \hbar\Omega_0 v^2]\frac{1}{(c^2k^2-\omega^2)a^2}\\
&P_k = \hbar(v_k - v) + [2(c^2k-\omega v)(\epsilon_k - \hbar\omega) - 2\hbar\Omega_0\omega v]\frac{1}{c^2k^2-\omega^2}\\
&A_k = 4(A - V\cos ak) - \frac{2\hbar\Omega_0\omega^2}{c^2k^2-\omega^2}\\
&\Omega_0 = \frac{4\pi d^2}{\hbar a^3}.
\end{aligned} \tag{18}$$

In deriving (18), the slowly varying envelope approximation

$$|\partial\varphi/\partial x| \ll k\varphi \qquad |\partial\varphi/\partial t| \ll \omega\varphi \tag{19}$$

has been employed and terms up to the second derivative have been kept. The set (17), (18) determine completely the propagation of coupled exciton–photon solitary waves. Note that in contrast to the case in [9], the non-linear coefficient A_k is a complicated function of k and ω. The first term $4A$ describes the dynamical exciton interaction which has a Coulomb origin. The second and third terms come from the Pauli statistics of electronic excitons. The second term $-4V\cos ak$ corresponds to a kinematical exciton repulsion, while the third term $\sim\Omega_0$ describes an additional exciton–polariton interaction which has a dipole character. On the lower polariton branch ($kc > \omega$) it corresponds to repulsion, while on the upper branch ($kc < \omega$) it yields exciton attraction.

The type of the soliton solutions of equation (17) depends on the sign of the quantity M_k/A_k. Positive values yield bright-soliton solutions:

$$\varphi(x,t) = \varphi_0 \operatorname{sech}\frac{x-vt}{L} \tag{20}$$

$$\hbar\omega = \epsilon_k - \frac{A_k\varphi_0^2}{2} - \frac{\hbar\Omega_0\omega^2}{c^2k^2-\omega^2} \qquad L^2 = \frac{2M_k}{A_k\varphi_0^2} \tag{21}$$

while negative values yield dark-soliton solutions:

$$\varphi(x,t) = \varphi_1 \tanh\frac{x-vt}{L} \tag{22}$$

$$\hbar\omega = \epsilon_k - A_k\varphi_1^2 - \frac{\hbar\Omega_0\omega^2}{c^2k^2-\omega^2} \qquad L^2 = -\frac{2M_k}{A_k\varphi_1^2} \tag{23}$$

where $2L$ is the width of the non-linear formations.

The velocity of the solitons, determined from the condition $P_k = 0$, is

$$v = \frac{2c^2k(\epsilon_k - \hbar\omega) + (c^2k^2-\omega^2)\hbar v_k}{2\omega(\epsilon_k - \hbar\omega + \hbar\Omega_0) + \hbar(c^2k^2-\omega^2)}. \tag{24}$$

A simple form of the non-linear interaction coefficient A_k can be obtained if we neglect the non-linear correction to the soliton frequency in (21) and use the linear polariton dispersion relation instead:

$$\frac{\hbar\Omega_0\omega^2}{c^2k^2-\omega^2} = \epsilon_k - \hbar\omega \tag{25}$$

which corresponds to $\chi_k = 0$. Using (18), (25) and (16) we get

$$A_k = 4A - 2\hbar(\omega_0 - \omega) = 4(A - V) - 2\hbar(\omega_T - \omega) \tag{26}$$

where $\omega_T = \omega_0 - 2V/\hbar$ is the transverse exciton frequency at $k = 0$. The non-linear coefficient A_k changes sign at $\omega_A = \omega_0 - 2A/\hbar$, being negative for $\omega < \omega_A$ and positive for $\omega > \omega_A$. The cut-off frequency yields a critical point k_1 on the upper branch which falls in the resonance polariton region (figure 1) and a critical point k_3 on the lower branch which is near the middle of the Brillouin zone.

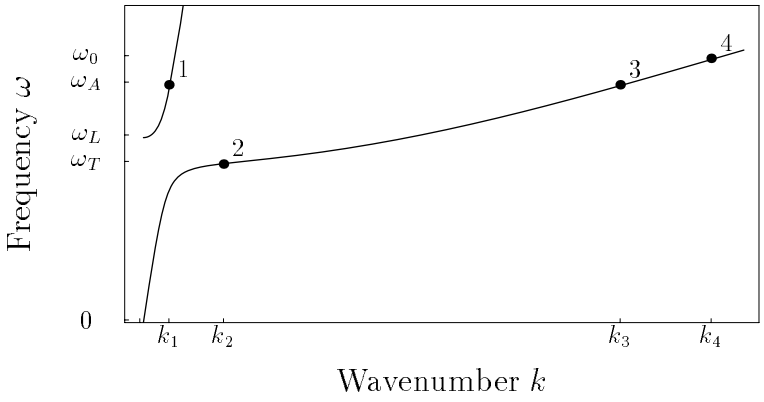

Figure 1. Critical points on the exciton–polariton dispersion curves which separate bright-soliton from dark-soliton solutions. $\Omega_0 = 0.1\omega_0$, $V = 0.2\hbar\omega_0$ and $A = 0.05\hbar\omega_0$. $\omega_T = \omega_0 - 2V/\hbar$, $\omega_L = \omega_T + \Omega_0$ and $\omega_A = \omega_0 - 2A/\hbar$.

The dispersion coefficient M_k plays the role of an inverse polariton effective mass and includes the effects of both the polariton-type and the exciton-type dispersion. The polariton-type dispersion dominates on the photon-like parts of the spectrum and in the resonance region ($|W| \gg |V|$). In these regions M_k is positive on the upper branch ($k < \omega/c$) and negative on the lower branch ($k > \omega/c$). $|W|$ decreases on the lower branch with the increase of the wavenumber and the dispersion coefficient M_k vanishes at a critical point k_2 which reflects the change from a polariton-type to an exciton-type dispersion. M_k vanishes again at the middle of the Brillouin zone at $k_4 = \pi/2a$, where the exciton effective mass changes sign.

The regions corresponding to bright- and dark-soliton solutions are shown in figure 1. On the upper branch, dark solitons exist for $k < k_1$ and bright solitons for $k > k_1$. On the lower branch four regions with solutions of different types are formed: bright solitons exist in the regions $0 < k < k_2$ and $k_3 < k < k_4$; and dark solitons for $k_2 < k < k_3$ and $k > k_4$. Thus in the low-density limit, bright solitons are formed in the photon-like parts of the spectrum, which is consistent with the results of [2]. In the high-density limit, according to (8) the solution types are reversed and dark solitons are formed in the photon-like parts of the spectrum. This is in agreement with the results of [8] for the case where the initial population inversion exceeds a threshold value and where the presence of damping limits the spread of the polariton dispersion curve near the light-line [6, 9].

We would like to point out that the changes of the solution type at k_1 and k_2 are in the resonance region and could have practical applications in optical switching devices.

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