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THE INFLUENCE OF TWO-PHONON-EXCITON INTERACTION ON THE CREATION OF LOCALIZED EXCITATIONS IN THE MOLECULAR CHAIN

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The influence of two-phonon-exciton interaction on the soliton characteristics in molecular chain with the cubic anharmonicity is investigated. It is shown that the soliton stability is enhanced by this interaction, especially in the region of the velocities close to the velocity of longitudinal sound waves.

1. Introduction

The possibility of the creation of localized collective nonlinear excitations (solitons) in the one-dimensional molecular structures with nearest neighbours harmonic interaction was demonstrated in several theoretical works[1–5]. These excitations arise as the consequence of the exciton-phonon interaction. The properties of these excitations are described in detail in the papers mentioned, so we shall only mention the fact that in the limiting case when the soliton velocity approaches to the longitudinal sound velocity v_0 $(v \to v_0)$, soliton energy diverges, and the distance between neighbouring molecules tends to zero. This implies that the harmonic approximation has a meaning only in the region $v \ll v_0$, while for $v \lesssim v_0$ one must take into account anharmonic corrections.

The results of these works are generalized in the Ref. 6 for the case of cubic nearest neighbours interaction (cubic anharmonism). It was shown that the energy of autolocalized excitations in this case remains finite even for $v = v_0$.

It is our aim in this paper to continue this work by introducing the effects of two-phonon-exciton interaction into the equations of motion, because their contribution to the soliton characteristics can not be neglected for large amplitudes of molecular vibrations which occur for $v \approx v_0$.

The paper is organized as follows. The chapter 2 contains the derivation of the equation of motion of an exciton in the continuum approximation, with harmonic and cubic anharmonic nearest neighbours interaction, as well as two-phonon contributions to the exciton-phonon interaction included. The general solution of solitonic type is determined. In the third chapter we discuss the effects of two-phonon-exciton interaction on the autolocalization of the exitations in the limiting cases $v \ll v_0$ and $v = v_0$.

2. *Hamiltonian density and the equations of motion*

The Hamiltonian function of a molecular chain, in the presence of an intramolecular excitation (exciton) in the chain, with single-phonon and two-phonon exciton-phonon interaction included, has the form

$$H = \sum_n [\psi_n^* [(e_0 - D_0)\psi_n - D_a(\psi_{n-1} + \psi_{n+1})] + \frac{1}{2} M \dot{U}_n^2 + M v_0^2 U(\varrho_n) +$$

$$+ \frac{\chi}{a} |\psi_n|^2 (U_{n+1} - U_{n-1}) + \frac{\chi_1}{a}(\psi_n^* \psi_{n+1} + \psi_{n+1}^* \psi_n)(U_{n+1} - U_n) +$$

$$+ \frac{\gamma}{a^2} |\psi_n|^2 (U_{n+1} - U_n)^2 + \frac{\gamma_1}{a^2}(\psi_n^* \psi_{n+1} + \psi_{n+1}^* \psi_n)(U_{n+1} - U_n)^2. \quad (2.1)$$

This expression was derived under the same condition as in the Ref. 6 and we use the same notations except for the new constants χ_1, γ and γ_1. χ_1 is the single phonon-exciton coupling constant which arises from the expansion of energy transfer matrix element in the molecular displacement and which was neglected in the previous work[6] compared to the »strong« exciton-single phonon coupling constant χ. γ and γ_1 describe two-phonon-exciton interaction, and have the same relation as χ and χ_1. The function $\psi_n(t)$ describes the probability of an excitation to be localized at the n-th molecule and is normalized as $\sum_n |\psi_n(t)|^2 = 1$.

Following the procedure of Ref. 6, we obtain, in the continuum approximation, the Hamiltonian density

$$\mathscr{H} = \frac{1}{a}\left\{E_0 |\psi|^2 - \frac{\hbar^2}{2m}\psi^* \frac{\partial^2 \psi}{\partial x^2} + \frac{1}{2} M \left(\frac{\partial U}{\partial t}\right)^2 + M v_0^2 U(\varrho) - 2\tilde{\chi} |\psi|^2 \varrho + \right.$$

$$\left. + 2\tilde{\gamma} |\psi|^2 \varrho^2 \right\} \quad (2.2)$$

with

$$\varrho = -\frac{\partial U}{\partial x} \tag{2.3}$$

and the normalization condition

$$\frac{1}{a}\int_{-\infty}^{+\infty} |\,\psi(x_1, t)\,|^2 \, dx = 1. \tag{2.4}$$

In (2.2): $E_0 = \Delta - D_0 - 2 D_a$ is the energy of the exciton band bottom, $m = \frac{\hbar^2}{2 D_a a^2}$, $\tilde{\chi} = \chi + \chi_1$ and $\tilde{\gamma} = \gamma + \gamma_1$. Intermolecular interaction potentials are given by

$$U(\varrho) = \frac{1}{2}\varrho^2 + \frac{1}{3}\varkappa\,\varrho^3 \tag{2.5}$$

where $\varkappa \geq 0$ is a dimensionless cubic anharmonicity parameter. One obtains the following equations of motion from the Hamiltonian density (2.2):

$$\mathrm{i}\,\hbar\frac{\partial\psi}{\partial t} = E_0\,\psi - \frac{\hbar^2}{2m}\frac{\partial^2\psi}{\partial x^2} - 2\tilde{\chi}\varrho\psi + 2\tilde{\gamma}\,\varrho^2\,\psi \tag{2.6}$$

$$M\frac{\partial^2 U}{\partial t^2} - M v_0^2 (1 + 2\varkappa\varrho)\frac{\partial^2 U}{\partial x^2} = 2\tilde{\chi}\frac{\partial}{\partial x}|\,\psi\,|^2 - 4\tilde{\gamma}\frac{\partial}{\partial x}(\varrho\,|\,\psi|^2). \tag{2.7}$$

The translational invariance of the system allows us to introduce the following solutions:

$$\psi(x_1, t) = \varphi(\xi)\,\mathrm{e}^{i\,(kx-\omega t)} \tag{2.8}$$

and

$$\varrho(x_1, t) = \varrho(\xi) \tag{2.9}$$

with

$$\xi = \frac{x - vt}{a} \quad (mv = \hbar k). \tag{2.10}$$

Substituting (2.8) and (2.9) into and (2.7) we obtain:

$$\frac{\partial^2\varphi}{\partial\xi^2} + \frac{2\tilde{\chi}}{D_a}\varrho\varphi - \frac{2\varkappa}{D_a}\varrho^2\,\varphi = \alpha\varphi \tag{2.11}$$

$$\varkappa\,\varrho^2 + \varrho\left(1 - s^2 + \frac{4\,\tilde{\gamma}}{M v_0^2}\varphi^2\right) - \frac{2\tilde{\chi}}{M v_0^2}\varphi^2 = 0 \tag{2.12}$$

and

$$\int_{-\infty}^{+\infty} \varphi^2 \, \mathrm{d}\xi = 1 \tag{2.13}$$

where:

$$s = v/v_0 \quad \text{and} \quad a = \frac{1}{D_a}\left(E_0 + \frac{\hbar^2 k^2}{2m} - \hbar\omega\right). \tag{2.14}$$

By expressing φ from equation (2.1), in the first order in two-phonon-exciton coupling constant, and introducing it into (2.12), one obtains the following result

$$\varphi_\xi^2 = \varphi^2 \left\{ a - \frac{2}{3} \nu \frac{\Phi^2}{\Phi + \delta} - \mu \frac{\Phi^3 - 2\delta^3}{\Phi + \delta} \right\} \tag{2.15}$$

with the boundary conditions

$$\varrho(\pm\infty) = 0, \quad \varphi(\pm\infty) = 0, \quad \varphi_\xi(\pm\infty) = 0 \tag{2.16}$$

and the auxiliary function Φ given by

$$\Phi^2 = \varphi^2 + \delta^2, \quad \delta = \frac{1 - s^2}{2} \sqrt{\frac{M v_0^2}{2\tilde{\chi}\varkappa}}, \quad \mu = \frac{4\tilde{\gamma}\tilde{\chi}}{D_a \varkappa M v_0^2},$$

$$\nu = \sqrt{\frac{\tilde{\chi}}{\varkappa^3 M v_0^2}} \left[\frac{\tilde{\gamma}(1 - s^2)(5\sqrt{2} - 4) + 4\tilde{\chi}\varkappa}{2D_a} \right]. \tag{2.17}$$

Using initial conditions

$$\varphi(0) = \varphi_0, \quad \varphi_\xi(0) = 0 \tag{2.18}$$

one arrives to

$$a = \frac{2}{3} \nu \frac{\Phi_0^2}{\Phi_0 + \delta} + \mu \frac{\Phi_0^3 - 2\delta^3}{\Phi_0 + \delta}. \tag{2.19}$$

The solution of (2.15) can be written in the form

$$\xi = \pm \sqrt{\frac{3}{2\nu}} \int_{\Phi}^{\Phi_0} C(\Phi) \frac{\mathrm{d}\Phi}{(\Phi - \delta)\sqrt{\Phi_0 - \Phi}} \tag{2.20}$$

where

$$C(\Phi) = \frac{\Phi}{\sqrt{\Phi + \delta}} \left\{ \Phi + \frac{\delta \Phi_0}{\Phi_0 + \delta} + \frac{3\mu}{2\nu}\left(\Phi^2 + \Phi\Phi_0 + \frac{2\delta^3 + \delta\Phi_0^2}{\Phi_0 + \delta} \right) \right\}^{-1/2}$$

and

$$\delta \leq \Phi \leq \Phi_0. \tag{2.21}$$

The behaviour of the function $C(\Phi)$ in the whole range of values for Φ, depends on the parameters δ and $\tilde{\gamma}$. In the limiting case for $v \to v_0$ ($\delta \to 0$), for $\varkappa \neq 0$ and $\tilde{\gamma} \neq 0$, it can be easily seen from (2.17) that $C(\Phi) < 1$. (Equality sign is valid only for the case when $\tilde{\gamma} = 0$, i. e. when one neglects two-phonon-exciton interaction.) In the other limiting case $\delta \to \infty$, the influence of anharmonism and two-phonon processes is negligible (i. e. $\varkappa \to 0$ and $\tilde{\gamma} \to 0$).

This means that the function $C(\Phi)$ varies smoothly in the range

$$\frac{1}{\sqrt{3}} \leq C(\Phi) \leq 1$$

so one could substitute it approximately with a constant $\bar{C}(\delta)$.

In this approximation, the solution (2.20) can be written in the form

$$\Phi = \Phi_0 \frac{1}{\mathrm{ch}^2 b\xi} + \delta\, \mathrm{th}^2 b\, \xi \tag{2.22}$$

where

$$b = \sqrt{\frac{\nu}{6} \frac{\sqrt{\Phi_0 \delta}}{\bar{C}(\delta)}}. \tag{2.23}$$

Normalizing condition (2.13) takes the form

$$\int_{\delta}^{\Phi_0} D(\Phi) \frac{\Phi\, \mathrm{d}\Phi}{\sqrt{\Phi_0 - \Phi}} = \sqrt{\frac{\nu}{6}} \tag{2.24}$$

where

$$D(\Phi) = \frac{1}{\Phi} C(\Phi)(\Phi + \delta). \tag{2.25}$$

The function $D(\Phi)$ behaves in the similar way as $C(\Phi)$. It varies smoothly in the range

$$\frac{2}{\sqrt{3}} \geqslant D(\Phi) \geqslant 1 \quad \text{when} \quad \delta < \Phi \leqslant \sqrt{\varphi_0 + \delta^2}$$

so it can be also substituted by a constant $\bar{D}(\delta)$. In this case, the normalization condition becomes

$$\sqrt{\Phi_0 - \delta}\,(2\Phi_0 + \delta) = \frac{1}{\bar{D}(\delta)} \sqrt{\frac{3\nu}{8}}. \tag{2.26}$$

The soliton width can be calculated from (2.22) using the expression

$$D^2 \xi = \langle \xi^2 \rangle - \langle \xi \rangle^2 \tag{2.27}$$

where

$$\langle \xi \rangle = \int_{-\infty}^{+\infty} \xi \varphi^2 \, d\xi$$

$$\langle \xi^2 \rangle = \int_{-\infty}^{+\infty} \xi^2 \varphi^2 \, d\xi$$

which gives

$$D^2 \xi = \frac{8^3}{9} \frac{\bar{C}^6(\delta)}{\nu^3} (2\Phi_0 + \delta)^3 \, [\varphi_0^3 (\pi^2 - 6) - (12 + \pi^2)(\delta - \Phi_0)\, \delta]. \tag{2.28}$$

3. *Discussion of limiting cases $v \ll v_0$ and $v = v_0$*

a) $v \ll v_0$. In the first order in $\varkappa$ and $\tilde{\gamma}$, when $\delta \to \infty$ ($\delta^2 \gg \varphi^2$) from (2.23) and (2.25) one obtains

$$b_\infty \cong \frac{1}{\bar{C}_\infty} \sqrt{\frac{\varphi_0^2 \nu}{12\delta}} \tag{3.1}$$

$$D_\infty = 2\bar{C}_\infty. \tag{3.2}$$

Using the normalizing condition (2.26) and the above relations, we obtain

$$2\varphi_0^2 = b_\infty = \frac{1}{2} \varphi_0 \lim_{\delta \to \infty} \sqrt{\frac{\nu}{\delta}} = \frac{1}{2} \varphi_0 \sqrt{\frac{2\tilde{\chi}}{D_a M v_0^2} \left(\frac{4\tilde{\chi}}{1 - s^2} + \frac{\tilde{\gamma}}{\varkappa} \right)} \tag{3.3}$$

or, finally

$$\varphi_0 = \frac{1}{4} \sqrt{\frac{2\tilde{\chi}}{D_a M v_0^2} \left(\frac{4\tilde{\chi}}{1 - s^2} + \frac{\tilde{\gamma}}{\varkappa} \right)}. \tag{3.4}$$

The soliton solution (2.22) can be written in the form:

$$\varphi^2 = \frac{\varphi_0^2 - 2\delta\,(\delta - \sqrt{\varphi_0^2 + \delta^2})\,\mathrm{sh}^2\, b\xi}{\mathrm{ch}^4\, b\xi} \tag{3.5}$$

which becomes the standard result

$$\varphi_\infty^2 \cong \frac{\varphi_0^2}{\mathrm{ch}^2\, b_\infty \xi} = \frac{\varphi_0^2}{\mathrm{ch}^2\, (2\varphi_0^2 \xi)} = \frac{\varphi_0^2}{\mathrm{ch}^2 \sqrt{a}\, \xi} \tag{3.6}$$

in the first order in $\varkappa$ and $\tilde{\gamma}$ (for $\delta \to \infty$).

The soliton energy in this range of velocities, follows from (2.14), (3.4) and (3.6)

$$\hbar\omega \cong E_0 + a^2 k^2 D_a - \frac{\tilde{\chi}^4}{D_a M^2 v_0^4 (1-s^2)^2} - \frac{\tilde{\chi}^3 \tilde{\gamma}}{2D_a^2 M^2 v_0^4 \varkappa (1-s^2)}. \quad (3.7)$$

Soliton width (2.28) in the approximation linear in $\frac{\tilde{\chi}}{\varkappa}$ becomes (for $\delta \to \infty$)

$$D^2 \xi \cong \frac{(12+\pi^2)}{36}\left(\frac{D_a M v_0^2}{\tilde{\chi}^2}\right)^2 \left[1 - \frac{\tilde{\gamma}(1-s^2)}{8\tilde{\chi}\varkappa}(1-s^2)\right]. \quad (3.8)$$

The relations (3.6), (3.7) and (3.8) become the standard expressions given in Refs. 1—5 in the case when cubic anharmonism and two-phonon processes are neglected ($\varkappa = 0$, $\tilde{\gamma} = 0$).

The relation (3.7) indicates that in the presence of two phonon-exciton interaction the rest energy of the soliton decreases by an amount of

$$|\Delta E_0| = \frac{\tilde{\chi}^3 \tilde{\gamma}}{2D_a M^2 v_0^2 \varkappa}. \quad (3.9)$$

As for the soliton dimension, (3.8) shows that cubic anharmonism and two--phonon-exciton interaction enhance the localization, with the increase of the parameter $\tilde{\gamma}/\varkappa$.

b) $v = v_0$ ($\delta = 0$). Taking (2.20) and (2.21) as the starting point and setting $\delta = 0$, one obtains the expression for the soliton amplitude for $v = v_0$

$$\varphi = \frac{2(\tilde{a}\varphi_0^2 + \varphi_0)}{1 + (1 + \tilde{a}\varphi_0)\,\mathrm{ch}\sqrt{a}\,\xi} \quad (3.10)$$

where

$$\tilde{a} = \frac{3}{\sqrt{2}}\frac{\tilde{\chi}}{\sqrt{\tilde{\chi} M v_0^2 \varkappa}}, \qquad a = \frac{2}{3}\tilde{a}\, v_0 \varphi_0^2 + \frac{2v_0}{3}\varphi_0,$$

$$v_0 = 2\sqrt{2}\frac{\tilde{\chi}^{3/2}}{D_a\sqrt{M v_0^2 \varkappa}}. \quad (3.11)$$

Normalizing condition in the approximation linear in $\tilde{a}$ is

$$a(16 + 32\tilde{a}\varphi_0) \cong 64\varphi_0^4 (1 + 4\tilde{a}\varphi_0). \quad (3.12)$$

The last equation can be solved by the iteration method. The first approximation gives

$$\varphi_0^{(1)} = \varphi_0^{(0)} - \frac{1}{3}\tilde{a}\,\varphi^{(0)2}; \; \varphi_0^{(0)} = \sqrt[3]{\frac{v_0}{6}} \quad (3.13)$$

and

$$a^{(1)} = \frac{2}{3} \nu_0 \varphi_0^{(0)} \left(1 + \frac{2}{3} \tilde{a} \varphi_0^{(0)}\right). \tag{3.14}$$

Soliton energy (2.14) for $v = v_0$ is

$$\hbar\omega = E_0 + a^2 k^2 D_a - a^{(1)} D_a = \tag{3.15}$$

$$= E_0 + a^2 k^2 D_a - \frac{4 \cdot 2^{3/2}}{3^{4/3}} \frac{\tilde{\chi}^2}{D_a^{1/3} (\varkappa M v_0^2)^{2/3}} \left(1 + \frac{4^{1/3}}{3^{1/3}} \frac{\tilde{\gamma}}{D_a^{1/3} (\varkappa M v_0^2)^{2/3}}\right)$$

and the soliton width in the first order in $\varkappa$ is

$$D^2 \xi = \sqrt{2} (\pi^2 - 6) \frac{D_a \sqrt{M v_0^2 \varkappa}}{\tilde{\chi}^{3/2}} \left[1 - \frac{3}{10\sqrt{2}} \left(\frac{3\sqrt{2}}{16}\right)^{1/3} \frac{\tilde{\gamma}}{D_a^{1/2} (\varkappa M v_0^2)^{5/6}}\right]. \tag{3.16}$$

Once again, one can conclude: the cubic anharmonism and two-phonon processes enhance localization.

The relation (3.5) for $\tilde{\gamma} = 0$, turns into the corresponding relation from the Ref. 6, where only cubic anharmonism was discussed. Two-phonon-exciton interaction decreases the soliton rest energy for a factor

$$\Gamma = \left(\frac{4}{3}\right)^{1/3} \frac{\tilde{\gamma}}{D_a^{1/3} (\varkappa M v_0^2)^{2/3}} \tag{3.17}$$

which, in the region of large velocities ($v \approx v_0$) is not negligible. One can demonstrate this by analysing the expression (3.17) for some typical values of relevant parameters. We shall estimate the exciton-phonon coupling constant, for this purpose. The coupling constants of the *strong* exciton-phonon interaction χ and γ arise from the expansion of the parameter D_0 (which represents the change of the dipole-dipole interaction of the excited molecule with nearest neighbours) in phonon displacements, while the coupling constants of the *weak* exciton-phonon interaction arise from the expansion of the matrix element D_a (dipole-dipole interaction) in phonon displacements (see Ref. 7, Chap. IV). Starting from the fact that D_0 decreases proportionally to the sixth power of the distance between the molecules $\left(D_0 = \frac{P}{a^6}, \text{ see Ref. 7, p. 160}\right)$, we can write:

$$D_0 (a + \Delta U) = D_0 \frac{a^6}{(a + \Delta U)^6} \approx D_0 - 6D_0 \frac{\Delta U}{a} + 21\, D_0 \left(\frac{\Delta U}{a}\right)^2 \tag{3.18}$$

where ΔU is the change of the distance between the neighbouring molecules. As $\varrho \approx \frac{\Delta U}{a}$, one can easily conclude that for the strong exciton-phonon interaction $\chi = 6D_0$ and $\gamma = 21D_0 \approx 3\chi$.

Taking data from Ref. 6 for the following two sets of parameters:

a) $D_a = 7.8\ \mathrm{cm}^{-1}$, $\chi = 27.3\ \mathrm{cm}^{-1}$, $M = 100\, m_p$, $v_0 = 114 \cdot 10^2$ cm/s,
$\varkappa = 5.2$, $a = 5.4 \cdot 10^{-8}$ cm

b) $D_a = 10\ \mathrm{cm}^{-1}$, $\chi = 60\ \mathrm{cm}^{-1}$, $M = 50\, \mathrm{m}_p$, $v_0 = 97.4 \cdot 10^2$ cm/s,
$a = 5.4 \cdot 10^{-8}$ cm

where m_B is the proton mass, the contribution of two-phonon-exciton interaction is numerically evaluated as:

$$\text{a) } \Gamma \approx 0.2; \quad \text{b) } \Gamma \approx 0.6 \tag{3.19}$$

which is of the same order of magnitude as the contribution of the cubic anharmonism ($\Gamma \approx 1$).

4. Summary

The analysis of the relevant physical characteristics of the soliton (energy, dimension, etc.) for the arbitrary velocity could not be performed, because, as can be seen from (2.22) and (2.26) it demands complicated numerical work to estimate the contribution of two-phonon processes. For this reason, we have analysed these quantities for the limiting cases $v \ll v_0$ and $v = v_0$ in the linear approximation in coupling constant of two-phonon-exciton interaction.

The analysis has shown that this interaction enhances the localization of the excitation and decreases its rest energy in both limiting cases.

In the region of small velocities, the contribution of this interaction, as well as cubic anharmonism is negligible, so in the limiting case $\varkappa = \tilde{\gamma} = 0$, all results coincide with the results of Refs. 1—5.

From the expression (3.15)—(3.17) and numerical results (3.19) it follows that the contribution of two-phonon processes in the region of large velocities ($v \approx v_0$) is of the same order of magnitude as the cubic anharmonism.

Thus, one can conclude that soliton stability at high velocities is improved by the presence of two-phonon-exciton interaction.

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DOPRINOS DVOFONONSKE EKSITON-FONON INTERAKCIJE KREIRANJU LOKALIZOVANIH EKSITACIJA U MOLEKULARNIM LANCIMA

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U radu je razmatran uticaj dvofononske eksiton-fonon interakcije na karakteristike solitona u molekularnim lancima sa kubnim anharmonizmom. Pokazano je da ova interakcija povećava stabilnost solitona u celom intervalu brzine $v < v_0$, gde je v_0 brzina longitudinalnog zvuka.