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SOLITONS AND BETHE ZONE SPLITTING

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We analyze the possibility o the soliton presence in the systems with Beth splitting of exciton zones and treat their properties by the method of eqquivalent Hamiltonian.

Introduction

Bethe splitting of exciton zones occurs when the multi-level excitation scheme can be applied to the molecules /1,2/. In case of w levels, there appear in principle w different types of excitons. We shall analyze here the influence of souch splitting to the solitons in molecular chains.

The Hamiltonian of the "frozen" exciton system corresponding to w-level scheme of molecular excitations in the harmonical approximation /3,4/ has the form:

$$H_o = \sum_{n\alpha} \Delta_\alpha B^+_{n\alpha} B_{n\alpha} + \sum_{nm\alpha} D^{\alpha\alpha}_{nm} B^+_{n\alpha} B_{n\alpha} - \sum_{nm\alpha} R^{\alpha\alpha}_{nm} B^+_{n\alpha} B_{m\alpha} +$$

$$+ \sum_{nm\alpha\alpha'} D^{\alpha\alpha'}_{nm} B^+_{n\alpha} B_{n\alpha'} - \sum_{nm\alpha\alpha'} R^{\alpha\alpha'}_{nm} B^+_{nm} B_{m\alpha'} \quad ; \ \alpha,\alpha' \in (1,2,\ldots w) \quad (1)$$

$B^+_{n\alpha}$ are Bose-operators creating the excitation of the type α on n-th site, Δ_α is the energy of the excitation of α-level of the isolated molecule, D and R are matrix elements of the dipole-dipole interaction.

Equivalent Hamiltonian theory

The Hamiltonian (1) is usually put in the form:

$$H_o = \sum_{nm\alpha\alpha'} \left[B^+_{n1} \ B^+_{n2} \ \cdots \ B^+_{nw} \right] \cdot \begin{pmatrix} M^{11}_{nm} & \cdots & M^{1w}_{nm} \\ \vdots & \ddots & \vdots \\ M^{w1}_{nm} & \cdots & M^{ww}_{nm} \end{pmatrix} \cdot \begin{pmatrix} B_{m1} \\ \vdots \\ B_{mw} \end{pmatrix} \quad (2)$$

where the matrix elements of $\hat{M}$ are given by

$$M^{\alpha\alpha'}_{nm} = \Delta_\alpha \delta_{\alpha\alpha'} \delta_{nm} + D^{\alpha\alpha'}_{nm} - R^{\alpha\alpha'}_{nm} \quad (3)$$

and the given form is diagonalized by an unitary transformation with the matrix $\hat{U}$ of the same rank as $\hat{M}$. This procedure encounters a number of mathematical obstacles, so we shall apply an approximate approach in which the Hamiltonian (1) is substituted by an effective Hamiltonian diagonal over the set of molecular indeces α.

The general Hamiltonian of the system appearing in the soliton theory includes also phonons and exciton-phonon interaction:

$$H_{tot} = H + H_o \tag{4}$$

where

$$H_p = -\frac{1}{2} \sum_n \left[\frac{p_n^2}{M} + Q \cdot \left(u_n - u_{n-1}\right)^2 \right] \quad ; \quad H_o \longrightarrow H \tag{5}$$

M is the mass of the molecule, Q - force constant and $p_n = M \cdot \dot{u}_n$ is the momentum o the molecule.

The transition to the effective Hamiltonian is realized through the unitary transformation

$$\begin{aligned} H_{eq} &= e^{-i\hat{\lambda}} \hat{H}_{tot} e^{i\hat{\lambda}} = \sum_{n=0} \frac{(-i)^n}{n!} \left[\hat{\lambda}, \left[\hat{\lambda}, \dots \left[\hat{\lambda}, \hat{H}_{tot}\right] \dots\right] \approx \\ &\approx \hat{H}_{tot} - i \cdot \left[\hat{\lambda}, \hat{H}_{tot}\right] - \frac{1}{2} \left[\hat{\lambda}, \left[\hat{\lambda}, \hat{H}_{tot}\right]\right] \end{aligned} \tag{6}$$

where

$$\hat{\lambda} = \sum_{fg\mu\nu} \left[\hat{X}^{\mu\nu}_{fg} B^{+}_{f\mu} B_{f\nu} + \hat{Y}_{fg} B^{+}_{f\mu} B_{g\nu} \right] \quad ; \quad X^{\mu\mu} = Y^{\mu\mu} = 0 \tag{7}$$

leading finally to

$$\begin{aligned} H_{eq} = &\sum_{n\alpha} \tilde{\Delta}_\alpha B^{+}_{n\alpha} B_{n\alpha} - \sum_{n\alpha} \tilde{R}_\alpha \left(B^{+}_{n\alpha} B_{n+1,\alpha} + B^{+}_{n\alpha} B_{n-1,\alpha}\right) + \\ &+ \sum_{n\alpha} \tilde{J}_{\alpha R} \left[B^{+}_{n\alpha} B_{n+1,\alpha} \left(u_{n+1} - u_n\right) + B^{+}_{n\alpha} B_{n-1,\alpha} \left(u_n - u_{n-1}\right)\right] - \\ &- \sum_{n\alpha} \tilde{J}_{\alpha D} B^{+}_{n\alpha} B_{n\alpha} \left(u_{n+1} - u_{n-1}\right) + \sum_{n\alpha} \left(\tilde{g}^{*\alpha\alpha}_{RD} B^{+}_{n\alpha} B_{n-1,\alpha} + g^{\alpha\alpha}_{RD} B^{+}_{n-1} B_{n\alpha}\right) \cdot \\ &\cdot \left(u_{n+1} - u_n\right) + \sum_{n\alpha} \left(\tilde{g}^{*\alpha\alpha}_{RD} B^{+}_{n\alpha} B_{n+1,\alpha} + g^{\alpha\alpha}_{RD} B^{+}_{n+1} B_{n\alpha}\right) \cdot \left(u_n - u_{n-1}\right) + \\ &+ \sum_n \left[-\frac{1}{2M} p_n^2 + \frac{Q}{2} \cdot \left(u_n - u_{n-1}\right)^2 \right] \end{aligned} \tag{8}$$

$$\tilde{\Delta}_\alpha = \Delta_\alpha + 2D^{\alpha\alpha} + 4\sum_{\alpha'} \frac{\left|D^{\alpha\alpha'}\right|^2}{\Delta_\alpha - \Delta_{\alpha'}} \quad ; \quad \tilde{R}_\alpha = R^{\alpha\alpha} + 4\sum_{\alpha'} \frac{R_e\left(R^{\alpha\alpha'} D^{\alpha\alpha'}\right)}{\Delta_\alpha - \Delta_{\alpha'}}$$

$$\tilde{g}_{RD} = \sum_{\alpha'} - \frac{R^{\alpha\alpha'} J^{\alpha\alpha'}_D}{\Delta_\alpha - \Delta_{\alpha'}} \quad ; \quad \tilde{J}_{\alpha R} = J^{\alpha\alpha}_R + 2\sum_{\alpha'} \frac{R_e\left(R^{\alpha\alpha'} J^{\alpha\alpha'}_D + 2J^{\alpha\alpha'}_R D^{\alpha\alpha'}\right)}{\Delta_\alpha - \Delta_{\alpha'}} \quad ;$$

$$J^{\alpha\alpha'}_R = N^{-1} \sum_k k\, R^{\alpha\alpha'}_k \sin ak \quad ; \quad J^{\alpha\alpha'}_D = N^{-1} \sum_k k\, D^{\alpha\alpha'}_k \sin ak$$

In this way, H_{og} is splitt into the sum of w independent Hamiltonians with respect to α and this faciliates the soliton analysis. An important restriction is the demand of the positive efective mass o excitons, which is satisfied for $\tilde{R}_\mu > 0$. This means that the number of various types of solitons which can appear in the system is equal to the number of terms in (8) with $\tilde{R}_\mu > 0$.

Let us suppose that for some fixed μ, $\tilde{R}_\mu > 0$ and test the properties of the corresponding soliton. Writing down Schrodingers equation for the wave function

$$|\mu\rangle = \sum_f A^\mu_f(t)\, B_{f\mu} \;;\quad \langle\mu|\mu\rangle = 1 \;;\quad \sum_f |A^\mu_f(t)|^2 = 1 \tag{9}$$

and averaging it over coherent phonon states $|C_p\rangle$, the continuum version of the equation obtained is

$$i\hbar \frac{\partial A^\mu}{\partial t} = (C_\mu + \tilde{\Delta}_\mu - 2\tilde{R}_\mu)A^\mu - a^2\tilde{R}_\mu \frac{\partial^2 A^\mu}{\partial x} + 2\chi_\mu A^\mu \frac{\partial \beta}{\partial x} \tag{10}$$

$$\chi_\mu = a\cdot(\tilde{J}_{\mu R} - \tilde{J}_{\mu D} + \mathcal{E}^{\mu\mu}_{RD} + \overset{*}{\mathcal{E}}{}^{\mu\mu}_{RD}) \tag{11}$$

were

$$C_\mu = \frac{1}{2a}\int_{-\infty}^{+\infty} dx\, \left[M\left(\frac{\partial \beta}{\partial x}\right)^2 + Q\, a^2 \left(\frac{\partial \beta}{\partial x}\right)^2 \right] \tag{12}$$

Combined equation for phonon operators, averaged over states $|\mu\rangle|C_p\rangle$ is

$$\frac{\partial^2 \beta}{\partial t^2} = v_0^2 \frac{\partial^2 \beta}{\partial x^2} + \frac{2\chi_\mu}{M}\frac{\partial}{\partial x}(\overset{*}{A}{}^\mu A^\mu) \;;\quad v_0^2 = a^2\frac{Q}{M} \tag{13}$$

and the continuum form of normalizing condition is

$$\int_{-\infty}^{+\infty} dx\, |A^\mu(x,t)|^2 = a \tag{14}$$

Solving the sistem (10) and (13) we obtain the final expressio for the energy and normalized amplitude. Energy of the soliton of μ - type is

$$E_{\mu(k)} = \tilde{\Delta}_\mu - 2\tilde{R}_\mu + a^2k^2\tilde{R}_\mu - \frac{\chi^4_\mu}{\tilde{R}_\mu M^2 v_0^2 (1 - v^2_{\mu k})^2}\left[1 + \frac{2}{3}\,\frac{1+v^2_{\mu k}}{1-v^2_{\mu k}}\right] \tag{15}$$

where

$$\nu_{\mu\kappa} = \frac{v_{\mu\kappa}}{v_0} \quad ; \quad v_{\mu\kappa} = \frac{2\, a^2\, \tilde{R}_\mu}{\hbar}\, \kappa \tag{16}$$

Normalized amplitude has the form

$$A^\mu(x,t) = \frac{a \cdot \Omega_\mu^{1/2}}{2}\; \frac{\exp(iKx - iE_\mu(k)t/\hbar)}{\mathrm{ch}\, \frac{a \cdot \Omega_\mu}{2} \zeta} \tag{17}$$

with

$$\Omega_\mu \equiv \Omega_\mu(\kappa) = \frac{2\, \chi_\mu^2}{a^2 \cdot \tilde{R}_\mu\, M\, v_0^2\, (1 - \nu_{\mu\kappa}^2)} \tag{18}$$

It is characteristic that each type of the solitons μ, has the corresponding type of lattice deformation given by

$$\frac{d\beta_\mu}{d\zeta} = -\frac{1}{2}\; \frac{a^2 \Omega_\mu \chi_\mu}{M_{\cdot} v_0^2\, (1 - \nu_{\mu\kappa}^2)}\; \frac{1}{\mathrm{ch}^2\, \frac{a \cdot \Omega_\mu}{2} \zeta} \tag{19}$$

$$\zeta = x - v_{\mu_k} t \tag{20}$$

It is important to stress that wave-packets can form also for $\tilde{R}_\mu < 0$, but only i $v_{\mu k} > v_0$. It is easy to determine their energy:

$$E'_\mu(\kappa) = \tilde{\Delta}_\mu + 2|\tilde{R}_\mu| - a^2\kappa^2 |\tilde{R}_\mu| + \frac{\chi'^4}{|\tilde{R}_\mu| M^2 v_0^4 (\nu'^2_{\mu\kappa} - 1)^2} \left\{ 1 + \frac{2}{3}\, \frac{\nu'^2_{\mu\kappa} + 1}{\nu'^2_{\mu\kappa} - 1} \right\} \tag{21}$$

One can see that thier energy is higher that the energy of the corresponding excitons, because the term arising from the exciton-phonon interaction has the positive contribution. The existence of these waves is rather doubtfull because they would represent rather unstable excitations.

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