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# Solitons in the system of interacting Frenkel excitons

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**Abstract.** The possible formation of solitons in a Frenkel exciton system, with exciton–exciton interaction only, is studied taking into account the Pauli character of exciton operators. This is realized by choosing the trial function of the system in the form which is the particular representation of the spin coherent state for spin 1/2. Possible solutions include both 'bright' and 'dark' solitons. Strict conditions for the existence and stability of particular types of soliton are formulated by imposing limitations on the values of the system parameters and soliton velocity. The energy–momentum relation for both types of solution is obtained. It is concluded that neither kind of soliton can exist if the effective dynamical interaction is a repulsive one. Recent references concerning 'bright' and 'dark' solitons are critically assessed.

## 1. Introduction

Solitons in one-dimensional excitonic systems have attracted considerable interest in the last twenty-five years, especially in the context of the charge and energy transfer over large distances in molecular chains, and self-induced transparency (SIT) [1–21]. The majority of the studies concerning the transfer phenomena deal with the so-called Davydov model, where solitons should arise on account of the single-'exciton' (vibron, electron, Frenkel exciton) trapping by the induced local distortion of the host lattice. Investigations carried out within the general theory of self-trapping (ST) phenomena [5–7] indicate that the original Davydov proposal, i.e. soliton formation on account of the single-exciton ST, cannot explain intra-molecular energy (amide-I quanta) transfer in biopolymers such as the  $\alpha$ -helix and acetanilide (ACN). That is, according to the available data [3-6], the widths of the exciton bands of these substances are too small as compared to the maximal-phonon-frequency nonadibatic limit, so one should expect the formation of small-polaron band states [5–7] rather than solitons. Nevertheless, recent analysis [8] indicates a possibility for soliton formation even in such systems, but only for higher excitation concentrations, where direct or indirect (phonon-mediated) exciton-exciton interaction significantly changes the conditions for soliton existence [8]. These results concern soliton formation in the system of the Bose quasi-particles: vibrational excitons (vibrons) only. However, in the system of Frenkel excitons, their Pauli character results in an extra (kinematical) exciton-exciton interaction [9-20] which additionally affects soliton properties. Thus, for the correct theoretical description of the exciton–solitons in the molecular crystals, proper accounting for the Pauli statistics of Frenkel excitons, and kinematical and dynamical effects is necessary. Up to now there have been only a few attempts (to the best of our knowledge) to treat these effects [10-17]. It was found that the two types of the exciton

soliton, so-called 'bright' and 'dark' ones, may be formed solely on account of the dynamical and kinematical interaction, even without exciton–phonon or exciton–photon coupling.

However, depending on the theoretical tool used in the particular study, different, sometimes even contradictory, conditions for the existence of a particular type of soliton were quoted. Thus, for example, Kruglov [10, 11] and Primatarowa and co-workers [12], using a procedure consisting in the averaging of the equation of motion for Pauli operators over the appropriate coherent state—Pauli-operator coherent states (POCS) [23, 24]—have found that the presence of the 'bright' or 'dark' soliton corresponds to the increase or decrease of the local exciton density, respectively. According to them, if the magnitude of the exciton density is less than 1/2, a 'bright' soliton arises, while if it lies between 1/2 and 1, a 'dark' soliton arises. In contrast, in previous studies [13,17], the appearance of the particular solution is associated with the nature of the dipole–dipole interaction, i.e. whether one deals with longitudinal (J > 0) or transverse excitons (J < 0), and the exciton–exciton (i.e. is it repulsive or attractive) interaction, and with the values of the energy parameters of the system, but was not related to the magnitude of the exciton density.

The origin of these discrepancies lies primarily in the uncontrolled approximation involved in the particular procedure. Thus, the inaccuracy in the studies [10, 13] could be the consequence of the approach based upon the averaging of the equations of motion for the Pauli operators over the POCS, which is not accurate enough due to the too-rough approximation involved in the evaluation of the expectation values of the exciton operators in terms of the exciton density [10, 12]. On the other hand, the approach based upon the bosonization of the Pauli operators leads to uncontrolled errors in the accounting for the diagonal terms which results in the appearance of an unphysical diagonal kinematical exciton–exciton interaction. As pointed out in [12], this term together with the Glauber coherent state *ansatz* for the trial state, gives a large but unphysical contribution to the equation of motion for the soliton envelope, which affects final results in uncontrolled way.

Note also that while in the case of SIT, due to its clear physical meaning which allows straightforward interpretation, this procedure is justified, this is not the case for the solitons in molecular crystals. That is, the expectation value of the exciton operator in the POCS represents the polarization of the medium induced by the electromagnetic (EM) field. Consequently, the solution of the equation of motion for this variable together with the solution of the corresponding Maxwell equations for the EM field describes the coupled propagation of the polarization and EM wave in the medium. In contrast, for molecular crystals, where the soliton should serve as the carrier of energy, polarization has no obvious meaning and therefore the proper treatment of the exciton-soliton demands its description in terms of more appropriate variables. The natural choice would be that utilized recently in the study of a related phenomenon: the soliton-like propagation of the condensed excitons in Cu<sub>2</sub>O [22], where the multiexciton system was described in terms of the multiparticle wave function having the meaning of the amplitude of the exciton density. Such solitons in the system of  $\mathcal{N}$  interacting excitons represent either the exciton bound state (the exciton drop or 'bright' soliton) or the defect (the exciton bubble, i.e. the region of the locally diluted excitons in the otherwise uniformly distributed excitons (or 'dark' soliton)). Furthermore, although in most of the studies on the solitons in the system of Frenkel excitons, the analysis was founded upon a model Hamiltonian which conserves the total exciton number, there were no attempts (with two exceptions [13, 17]) to find the connection between the soliton properties and the exciton number.

Let us note that although the problem of the existence of the exciton–soliton is a longstanding one and although nowadays there is increasing interest in the investigation of these systems in the present context: discrete moving solitons and nonlinear self-localized modes [20, 21], for the reasons given above there is still a need for revisiting the problem in a way which allows examination of exciton–soliton properties, especially for formulating criteria for their existence, without the above-mentioned inconsistencies. For that purpose we are going to formulate the problem in terms of the soliton density, so that it can be easily related to the exciton number. Particular attention will be paid to the analysis of soliton properties as regards their dependence on the values of physical parameters of the system and the character of the interaction terms, all within the continuum approximation which allows one to establish the soliton energy–momentum relation, which, in spite of a number of studies having been carried out on the subject, has never previously been done. Moreover, since the applicability of the continuum approximation is discussed in detail, we hope that the present analysis will help in the understanding of the limit up to which a strictly continuous soliton can exist and when one should go beyond the continuum approximation and look for excitations which cannot be described within the continuum model.

## 2. The model and method

In what follows we shall consider a system consisting of a one-dimensional molecular chain of period  $R_0$  with one molecule per lattice site, constituted from N molecules and populated by N excitons. It can be described, within the Heitler–London approximation, by the following Hamiltonian [9]:

$$H = \Delta \sum_{n} P_{n}^{+} P_{n} - J \sum_{n} P_{n}^{+} (P_{n+1} + P_{n-1}) + U \sum_{n} P_{n}^{+} P_{n} P_{n+1}^{+} P_{n+1}$$
(1)

where  $P_n^+$  ( $P_n$ ) represents the Pauli operator describing the presence (absence) of an exciton at the *n*th lattice site, while  $\Delta$ , *J* and *U* represent the intra-molecular excitation energy, the resonant dipole–dipole interaction responsible for the exciton transfer and the effective (either direct or phonon-mediated) dynamical exciton–exciton interaction, respectively.

In order to find the moving stationary-soliton solution we use the time-independent variational principle. For that purpose we adopt the semiclassical approximation, and following references [10, 13] we choose the normalized trial state of the system in a form which reflects the Pauli character of the excitations, i.e. we choose the trial state of the system in the form of a Pauli-operator coherent state (POCS):

$$|\Psi\rangle = \prod_{n} (\Phi_n + \Psi_n P_n^+) |0\rangle \qquad |\Phi_n|^2 + |\Psi_n|^2 = 1 \qquad P_n |0\rangle = 0.$$
 (2)

This form was first used in the context of the self-induced transparency (SIT) by Agranovich and Rupasov [21] and by Kruglov [10, 11] and Primatarowa *et al* [12] in the context of the exciton–solitons in molecular crystals. Let us recall that the above choice of trial state is equivalent to the one recently utilized by Konotop and Takeno [18, 19] in the analysis of the related problem of the existence of nonlinear excitations in systems with exchange and dipole– dipole interaction. That is, the POCS (2) represents the particular (s = 1/2) realization of the spin coherent state:

$$|\Psi\rangle = \prod_{n} |\mu_{n}\rangle$$
 where  $|\mu_{n}\rangle = \frac{\exp(\mu_{n}P_{n}^{+})}{\sqrt{1+|\mu_{n}|^{2}}}|0\rangle$ 

which in [18,19] has been used as a trial state. According to [23,24],  $\mu_n = \tan(\theta_n/2)e^{i\phi_n}$ , so  $\Psi_n$ and  $\Phi_n$  can be related to  $\mu_n$  through the explicit expressions  $\Phi_n \equiv \cos \theta_n$  and  $\Psi_n \equiv \sin \theta_n e^{i\phi_n}$  $(0 < \phi_n < 2\pi \text{ and } 0 < \theta_n < \pi)$ .

Due to the constraint that the total exciton number  $\hat{\mathcal{N}} = \sum_n P_n^+ P_n$  is a constant of the motion, the trial state should be normalized as follows:

$$\langle \Psi | \sum_{n} P_{n}^{+} P_{n} | \Psi \rangle = \sum_{n} |\Psi_{n}|^{2} = \mathcal{N}.$$

In what follows we shall describe the soliton in terms of  $\Psi_n$ , which according to the relation  $\langle \Psi | P_n^+ P_n | \Psi \rangle = |\Psi_n|^2$  has the meaning of the amplitude of the exciton density. The connection with the results of the papers [10–12], where the soliton solutions of the equation of motion for the 'polarization' were considered, can be found easily due to the relations  $p_n = \langle \Psi | P_n | \Psi \rangle \equiv \Phi_n^* \Psi_n$  and  $|p_n|^2 = |\Psi_n|^2 (1 - |\Psi_n|^2)$ .

We shall find an equation for the exciton amplitudes  $\Psi_n$  by minimizing the ground-state energy of the system imposing the constraint that the operators of the total exciton number

$$\hat{\mathcal{N}} = \sum_{n} P_n^+ P_n$$

and momentum

$$\hat{P}_{ex} = \frac{\hbar}{2iR_0} \sum_{n} P_n^+ (P_{n+1} - P_{n-1})$$

are integrals of motion. (The expression for the momentum used here follows directly from the evolution equation for the position operator  $R = \sum_{n} R_0 n P_n^+ P_n$ , i.e.  $P = m_{exc} \dot{R}$ .)

In other words, one should minimize the following functional:

 $\mathcal{H} = \langle \Psi | H - \lambda \hat{\mathcal{N}} - v \hat{P}_{ex} | \Psi \rangle$ 

where  $\lambda$  and v represent Lagrange multipliers whose physical meaning will be explained later on. The explicit expression for this functional reads

$$\mathcal{H} = (\Delta - \lambda) \sum_{n} |\Psi|^{2} - J \sum_{n} \Phi_{n} \Psi_{n}^{*} (\Psi_{n+1} \Phi_{n+1}^{*} + \Psi_{n-1} \Phi_{n-1}^{*}) + U \sum_{n} |\Psi_{n}|^{2} |\Psi_{n+1}|^{2} - \frac{v\hbar}{2iR_{0}} \sum_{n} \Phi_{n} \Psi_{n}^{*} (\Phi_{n+1} \Psi_{n+1}^{*} - \Phi_{n-1} \Psi_{n-1}^{*}).$$
(3)

In order to find the equation for  $\Psi_n$ , we demand stationarity of the above functional, i.e.  $\partial \mathcal{H}/\partial \Psi_n^* = 0$ . Note that due to the above-established connection between  $\Psi_n$  and  $\Phi_n$ , variations over these parameters are not independent, so  $\partial \mathcal{H}/\partial \Phi_n^* = 0$  is superfluous. Thus it follows that before performing the minimization of the above functional over  $\Psi_n$ , one should eliminate  $\Phi_n$  from the expression for  $\mathcal{H}$ . In such a way, using the relation  $\Phi_n = \sqrt{1 - |\Psi_n|^2}$ , and going over to the continuum approximation, we arrive at

$$\mathcal{H} = (\Delta - \lambda - 2J) \int \frac{dx}{R_0} |\Psi(x)|^2 + JR_0^2 \int \frac{dx}{R_0} |[\Psi(x)\sqrt{1 - |\Psi(x)|^2}]_x|^2 + \frac{i\hbar v}{2} \int \frac{dx}{R_0} \left[ \Psi^*(x)\sqrt{1 - |\Psi(x)|^2} \frac{\partial}{\partial x} (\Psi(x)\sqrt{1 - |\Psi(x)|^2}) - \text{c.c.} \right] + (2J + U) \int \frac{dx}{R_0} |\Psi(x)|^4.$$
(4)

Note that due to the fact that  $\Phi$ -terms are always coupled to the corresponding  $\Psi$ -terms in the above expressions, the sign and even the phase can always be 'absorbed' in the definition of  $\Psi$ ; we have thus opted for the simplest choice, i.e. the positive sign with no further phase.

Clearly, all further results strictly concern the continuum case. The applicability or validity of such an approximation will be discussed in the next section with respect to the particular type of the solution. Here the dispersion in the nonlinear term has been neglected. This is justified since  $|\Psi(x)|^2 < 1$ , so the term is proportional to the product of the exciton density and

its derivative. Accordingly, we may expand  $\sqrt{1 - |\Psi(x)|^2}$  in powers of the small 'parameter'  $|\Psi(x)|^2$ . In addition, due to the smallness of that parameter, all the terms of higher order in the exciton density and its derivatives  $(|\Psi(x)|^2 |\Psi_x(x)|^2 \text{ etc})$  may be neglected, and we obtain

$$\mathcal{H} \approx (\Delta - \lambda - 2J) \int \frac{\mathrm{d}x}{R_0} |\Psi(x)|^2 + JR_0^2 \int \frac{\mathrm{d}x}{R_0} |\Psi_x(x)|^2 (1 - |\Psi(x)|^2) + (2J + U) \int \frac{\mathrm{d}x}{R_0} |\Psi(x)|^4 + \frac{i\hbar v}{2} \int \frac{\mathrm{d}x}{R_0} (1 - |\Psi(x)|^2) (\Psi^*(x)\Psi_x(x) - \mathrm{c.c.}).$$
(5)

This can be recast into a more convenient form by choosing  $\Psi(x) = \psi(x)e^{ikx}$  where  $\psi(x)$  in the general case is a complex function. Substituting this form for the exciton amplitude into the last equation and minimizing the functional so obtained over k, we obtain  $k = \hbar v/(2JR_0^2) \equiv m_{ex}v/\hbar$ , while expression (5) now reads

$$\mathcal{H} = \tilde{\Delta} \int \frac{\mathrm{d}x}{R_0} |\psi(x)|^2 + JR_0^2 \int \frac{\mathrm{d}x}{R_0} |\psi_x(x)|^2 (1 - |\psi(x)|^2) + G(v) \int \frac{\mathrm{d}x}{R_0} |\psi(x)|^4 \tag{6}$$

where  $G(v) = U + 2J + m_{ex}v^2/2$  and  $\tilde{\Delta} = \Delta - 2J - \lambda - m_{ex}v^2/2$ . This procedure is fully equivalent to the usual one where k is determined from the equation for the envelope function  $\psi(x)$  by requiring the terms multiplied by the imaginary unit to vanish. Here, consistently with the above-adopted approximation, the term containing the product of the exciton density and its derivative should be neglected. In such a way, we arrive at the nonlinear Schrödinger equation (NSE) whose solutions (and their properties) are well known [25, 26]. The resulting equation for the soliton amplitude reads

$$\psi_{xx}(x) - \frac{\tilde{\Delta}}{JR_0^2}\psi(x) - \frac{2G(v)}{JR_0^2}\psi^3(x) = 0.$$
(7)

Here we restrict ourselves to the particular case where  $\psi(x)$  is taken to be real. Depending on the sign of the parameters G(v) and J, it may have two types of soliton solution: the bell-shaped or 'bright' soliton if JG(v) < 0 and the kink-like or 'dark' soliton in the opposite case.

## 3. Analysis of the solutions

#### 3.1. The 'bright' soliton

The first integral of this equation, conforming to the 'bell'-shaped boundary conditions  $(\psi(\pm \infty) = 0, \psi_x(\pm \infty) = 0 \text{ and } \psi(x_0) = \psi_0 \neq 0$  where  $x_0$  represents the centre of mass of the soliton), is given as follows:

$$\psi_x^2(x) = V(\psi(x)) \equiv \frac{G(v)}{JR_0^2} \psi^2(x) \left(\frac{\Delta}{G(v)} + \psi^2(x)\right).$$
(8)

 $\psi_0$  denotes the soliton amplitude (the magnitude of the exciton density) and corresponds to the zeros of the 'potential'  $V(\psi)$ , i.e.  $V(\psi_0) = 0$ . Consequently it is given as  $\psi_0^2 = -\tilde{\Delta}/G(v)$ , and the last equation attains the form

$$\psi_x^2(x) = -\frac{G(v)}{JR_0^2}\psi^2(x)(\psi_0^2 - \psi^2(x)).$$
(9)

Integrating this equation, we find the normalized bell-shaped soliton solution:

$$\psi(x) = \mathcal{N}_{\sqrt{\frac{\mu}{2}}} \operatorname{sech} \frac{(x - x_0)}{l}$$
(10)

where  $l = R_0/(\mu N)$  denotes the soliton width while  $\mu = -G(v)/(2J)$ . From the normalization condition we find

$$\psi_0 = \mathcal{N}\sqrt{-\frac{G(v)}{4J}}.$$

Combining this expression for  $\psi_0$  with the one given in terms of G(v) and  $\tilde{\Delta}$ , we may determine the Lagrange multiplier  $\lambda$ :

$$\lambda = \Delta - 2J - \frac{m_{ex}v^2}{2} - \frac{G^2(v)N^2}{4J}.$$
(11)

As is known [26], the stable solution, i.e. the one minimizing functional (8), corresponds to the case J > 0 and G(v) < 0 ( $G(v) \equiv -|G(v)|$ ). Thus the stability condition for this solution is

$$U + 2J + \frac{m_{ex}v^2}{2} < 0. (12)$$

According to this condition, soliton existence, in the system of Frenkel excitons, demands an attractive and large enough  $(U = -|U| < 0 \text{ and } |U| > 2J + m_{ex}v^2/2)$  exciton–exciton interaction.

Substituting the above-obtained soliton solution into the expressions for the soliton energy  $(E_{sol} = \langle \Psi | H | \Psi \rangle)$  and momentum  $(P_{sol} = \langle \Psi | \hat{P}_{ex} | \Psi \rangle)$ , we have

$$E_{sol} = (\Delta - 2J)\mathcal{N} + \frac{m_{ex}v^2}{2}\mathcal{N} - \frac{\mathcal{N}^3}{12J} \left[ G^2(v) + 2|G(v)|m_{ex}v^2 \right]$$
(13)

and

$$P_{sol} = m_{ex} v \mathcal{N} \left( 1 - \frac{|G(v)|}{6J} \mathcal{N}^2 \right).$$
(14)

Let us now examine the relation between  $E_{sol}$  and  $P_{sol}$ . According to the explicit expression for G(v), one can solve (14) as a cubic equation in terms of v. It turns out that there exists a single real solution  $v = v(P_{sol})$ , but under the condition  $\mathcal{N}^2 < 6J/(|U| - 2J)$ . After straightforward but tedious algebraic manipulations, we obtain

$$E_{sol}(P_{sol}) = (\Delta - 2J)\mathcal{N} + \frac{\mathcal{E}^2}{4J\mathcal{N}} \left[ \sinh^2(2\tilde{P}) - 2\sinh^2(\tilde{P}) \right] - \frac{(U - 2J)^2}{12J}\mathcal{N}^3$$
(15)

where

$$\mathcal{E} = \frac{2}{3} [2J(\mathcal{N}^2 + 3) - \mathcal{N}^2 |U|] \qquad \tilde{P} = \frac{1}{3} \operatorname{arcsinh} \left( \frac{6J}{\sqrt{m_{ex} \mathcal{E}^3}} P_{sol} \right).$$

From the above relations it is easy to prove that v represents the soliton velocity since the following relation holds:  $\partial E_{sol}/\partial P_{sol} = v$ , where v denotes the unique solution of the above-mentioned cubic equation, i.e.  $v = v(P_{sol})$ . Note also that the same conclusion follows straightforwardly from equations (13) and (14):

$$\frac{\partial E_{sol}}{\partial P_{sol}} = \frac{\partial E_{sol}/\partial v}{\partial P_{sol}/\partial v} \equiv v.$$

Finally one may find the soliton effective mass from the definition  $m_{sol} = (\partial P_{sol}/\partial v)_{v \to 0}$ , so we have

$$m_{sol} = m_{ex} \mathcal{N} \left[ 1 - \frac{(|U| - 2J)}{6J} \mathcal{N}^2 \right].$$
 (16)

Clearly, due to the above-mentioned condition  $N^2 < 6J/(|U| - 2J)$ , the soliton mass should be positive.

Relation (12) implies that, under the most favourable condition (v = 0), soliton existence demands |U| > 2J, so the formation of 'bright' multiexciton–solitons in the pure excitonic system is not very probable. That is, dynamical exciton–exciton interaction is usually attractive and has a quadrupolar character, and consequently is much weaker than the dipole–dipole transfer term ( $|U| \ll J$ ) [9], so the condition for the soliton formation is not satisfied. Therefore, soliton formation demands an additional interaction which may compensate for the kinematical term. A common mechanism causing the corresponding effects is the exciton– phonon interaction [20], which, irrespectively of the mechanism of soliton formation [8], causes the renormalization of the exciton–exciton interaction; hence U should be replaced by  $U_{eff} = U - \varepsilon$ , where  $\varepsilon \sim E_B$  measures an additional, phonon-mediated, exciton–exciton interaction. ( $E_B$  denotes the so-called small-polaron binding energy.)

Besides the above-formulated condition for soliton existence and stability, the validity of the continuum approximation  $(l \gg R_0)$  and the restriction to the low-exciton-density  $(|\Psi(x)|^2 < 1)$  limit impose two more conditions:

$$\mathcal{N}\frac{|U| - 2J - m_{ex}v^2/2}{2J} \ll 1 \qquad \frac{\mathcal{N}^2}{2}\frac{|U| - 2J - m_{ex}v^2/2}{2J} < 1.$$
(17)

From the second one we obtain  $l \gg NR_0/2$  which, however, does not mean that, for the given set of system parameters, the soliton width increases with the rise of N. It simply means that the simultaneous satisfaction of both conditions in (17) indicates, due to the 1/N dependence of the soliton width, that soliton formation in systems populated with higher numbers of excitons Nis possible in systems with lower dynamical interaction |U|. In this respect, our understanding of the nature of the dependence of the soliton parameters on N substantially differs from that of Kislukha [17] who saw these conditions (i.e. (17)) as indicating the soliton spreading with the rise of the exciton population. On the contrary, increase of N causes shrinking of the soliton. Soliton motion has a twofold role in its properties. That is, while on one hand it violates soliton stability and, according to (14), above some critical velocity

$$v_c = \sqrt{\frac{2}{m_{ex}}}(|U| - 2J)$$

the soliton becomes unstable, on the other hand, due to the decrease of the parameter G(v), it ensures better applicability of the continuum approximation, i.e. increase of the soliton speed causes spreading of the soliton width.

## 3.2. The 'dark' soliton

Equation (9) has a kink-soliton solution [27] which may be found by imposing the following boundary conditions:  $\psi(x_0) = 0$ ,  $\psi(\pm \infty) = \pm \tilde{\psi}_0$  and  $\psi_x(\pm \infty) = \psi_{xx} = 0$ . Here  $\tilde{\psi}_0$ represents the 'vacuum' solution corresponding to the minima of the 'potential'  $V(\psi(x))$ . Consequently it is explicitly given as  $\tilde{\psi}_0^2 = -\tilde{\Delta}/(2G(v))$ . Integrating (9) with the above boundary conditions, we obtain

$$\psi(x) = \tilde{\psi}_0 \tanh \frac{(x - x_0)}{l} \tag{18}$$

with *l* representing the soliton width:

$$l = \sqrt{\frac{JR_0^2}{\tilde{\psi}_0^2 G(v)}}.$$

The above solution is known as a 'dark' soliton, since its squared modulus

$$\psi^{2}(x) = \tilde{\psi}_{0}^{2} \left[ 1 - \operatorname{sech}^{2} \frac{(x - x_{0})}{l} \right]$$
(19)

describes the region with the locally diluted exciton density—the 'hole' against the background of the uniformly distributed particles with the density  $\psi_0^2$ .

In this case, due to the divergence of the integral

$$\int_{-\infty}^{\infty} \frac{\mathrm{d}x}{R_0} |\Psi(x)|^2 = \int_{-\infty}^{\infty} \frac{\mathrm{d}x}{R_0} \psi^2(x)$$

the soliton amplitude  $(\tilde{\psi}_0)$  cannot be found as simply as in the case of the bright soliton. Furthermore, analogous divergences also appear in the calculation of the integrals of the soliton momentum and energy. In order to avoid these difficulties, we shall follow a standard procedure introduced in the theory of dark solitons [28] and which simply consists in the subtracting of the corresponding 'vacuum' contributions from these integrals. Thus the normalization condition now reads

$$\tilde{\mathcal{N}} = \int_{-\infty}^{\infty} \frac{\mathrm{d}x}{R_0} \, (\psi^2(x) - \tilde{\psi}_0^2). \tag{20}$$

This, obviously negative, quantity should be understood as a 'defect' in the uniform state, i.e. the number of particles which are missing from the 'hole'. After some calculation we obtain

$$\tilde{\mathcal{N}} = -2\tilde{\psi}_0 \sqrt{\frac{J}{G(v)}}$$

from which one may find the soliton amplitude, Lagrange multiplier  $\lambda$  and soliton width as follows:

$$\tilde{\psi}_0 = -\frac{\tilde{\mathcal{N}}}{2}\sqrt{\frac{G(v)}{J}} \qquad \lambda = \Delta - 2J - \frac{m_{ex}v^2}{2} + \frac{G^2(v)\tilde{\mathcal{N}}^2}{2J} \qquad l = \frac{2J}{G(v)}\frac{R_0}{|\tilde{\mathcal{N}}|}.$$
 (21)

Using appropriate—analogous to the above—renormalization of the soliton momentum and energy, we find

$$P_{sol} = m_{ex} v \tilde{\mathcal{N}} \left( 1 - \frac{G(v)}{3J} \tilde{\mathcal{N}}^2 \right)$$
(22)

and

$$E_{sol} = (\Delta - 2J)\tilde{\mathcal{N}} + \frac{m_{ex}v^2}{2}\tilde{\mathcal{N}} + \frac{\tilde{\mathcal{N}}^3}{6J} \left[ G^2(v) - 2G(v)m_{ex}v^2 \right].$$
 (23)

Like in the case of the bright soliton, one can easily express v, by solving equation (23) as a cubic equation for v, as  $v = v(P_{sol})$ , which enables the establishing of the following explicit energy-momentum relation:

$$E_{sol}(P_{sol}) = (\Delta - 2J)\tilde{\mathcal{N}} - \frac{\mathcal{E}^2}{J\tilde{\mathcal{N}}}\cos^2\left(\frac{\pi}{3} + \tilde{P}\right)\cos\left(\frac{2\pi}{3} + 2\tilde{P}\right) + \frac{(U+2J)^2}{6J}\tilde{\mathcal{N}}^3$$
(24)

where

$$\mathcal{E} = \frac{2}{3} [J(3 - 2\tilde{\mathcal{N}}^2) - \tilde{\mathcal{N}}^2 U] \qquad \tilde{P} = \frac{1}{3} \arccos\left(\frac{3J}{\sqrt{m_{ex}|\mathcal{E}|^3}} P_{sol}\right).$$

Here again, v has the meaning of the soliton velocity ( $v = \partial E_{sol} / \partial P_{sol}$ ), while the soliton effective mass becomes

$$m_{sol} = m_{ex} \tilde{\mathcal{N}} \left[ 1 - \frac{(U+2J)}{3J} \tilde{\mathcal{N}}^2 \right].$$
<sup>(25)</sup>

Obviously, due to the 'negativity' of  $\tilde{\mathcal{N}}$ , the soliton mass (as well as its momentum and energy) is also negative. From the formal point of view this is the consequence of our choice of

the normalization condition as defined by (20). These 'negativities' cannot be simply avoided. Thus if one tried to get rid of these difficulties by, for example, choosing the normalization condition as follows:

$$\tilde{\mathcal{N}} = \int_{-\infty}^{\infty} \frac{\mathrm{d}x}{R_0} \; (\tilde{\psi_0}^2 - \psi^2(x))$$

the energy–momentum relation would no longer hold (i.e.  $v \neq \partial E_{sol}/\partial P_{sol}$ ). Furthermore, the alternative normalization condition

$$\tilde{\mathcal{N}} = \int_{-L}^{L} \frac{\mathrm{d}x}{R_0} \,\psi^2(x)$$

(where 2*L* represents the length of the system) also leads to the violation of the  $E_{sol}-P_{sol}$  relation as pointed out in references [27,29]. We consider this relation to be the essential one, and this dictated our choice of the normalization condition.

From the physical point of view, the 'negativity' of these parameters should not be surprising in view of the above-mentioned interpretation of a dark soliton: the hole in the otherwise homogeneous particle-density–exciton 'condensate'.

The above solution minimizes the functional if J > 0 and G(v) > 0, so the stability condition now reads

$$U + 2J + \frac{m_{ex}v^2}{2} > 0. (26)$$

On the other hand, the applicability of the continuum approximation  $l \gg R_0$  and the restriction to the low-exciton-density limit impose

$$|\tilde{\mathcal{N}}| \frac{U + 2J + m_{ex}v^2/2}{2J} \ll 1 \qquad \frac{\tilde{\mathcal{N}}^2}{2} \frac{U + 2J + m_{ex}v^2/2}{2J} < 1.$$
(27)

The motion of the 'dark' solitons has quite the opposite tendency to that of the 'bright' solitons: increase of the soliton velocity supports its stability followed, immediately, by its shrinking.

From the above results it follows that, like in the case for 'bright' solitons, existence of 'dark' ones demands an attractive exciton–exciton interaction (U < 0). That is, although the stability condition is satisfied even for the repulsive inter-exciton interaction, the continuum approximation may be satisfied only through the compensation of the effective kinematic term  $(2J + m_{ex}v^2/2)$  by the—high enough—attractive dynamical exciton–exciton interaction. Due to the smallness of the direct dynamical term  $(|U| \ll J)$ , the exciton–phonon interaction should be the mechanism which, in the system of interacting Frenkel excitons, will ensure soliton formation.

## 4. Discussion

Here we wish to analyse in detail the basic ingredients of our approach: a time-independent variational method based upon the semiclassical and continuum approximations. We note that our results, obtained within the time-independent variational method, are no less general than the previous ones obtained by the time-dependent variational procedure [10–12, 15–17]. That is, the choice of the time-dependent soliton solution in the form where the soliton envelope depends on time only through the coordinate in the moving frame:  $f(x - vt)e^{i(kx-\omega t)}$ , is practically equivalent to the time-independent variational procedure with explicit accounting for the momentum and particle number (norm) conservation [26]. Explicit time dependence is important only for the analysis of the soliton stability, i.e. linear stability with respect to small perturbations ( $\Psi \rightarrow \Psi_{sol} + \delta \Psi$ ;  $\delta \Psi \ll \Psi_{sol}$ ), or for the examination of the soliton dynamics

under the influence of external perturbations. Since our analysis relies upon equation (9), which is the stationary limit of the NSE, for the linear stability of the above-obtained solutions we may adopt the criteria formulated for the 'bright' and 'dark' solitons of the NSE. Accordingly, we may state that our 'bright'-soliton solution is stable with respect to small perturbations [26], while for the 'dark' soliton, the soliton stability condition demands  $\partial P_{sol}/\partial v < 0$  [28] which leads to

$$\tilde{\mathcal{N}}^2 \frac{U + 2J + 5m_{ex}v^2/2}{3J} < 1.$$
<sup>(28)</sup>

Here, we use the term 'stability' to denote so-called 'longitudinal stability' (i.e. the small perturbation is a function of the coordinate in the direction of the soliton motion,  $\delta \Psi = \delta \Psi(x)$ ). With respect to so-called transverse perturbations, both 'bright' and 'dark' solitons are unstable [25,26].

As regards the validity of the semiclassical approximation, introduced through the trial state *ansatz*, we note that our trial function chosen as a POCS is not the eigenstate of the Hamiltonian, exciton momentum and particle number operator, and therefore in the soliton solutions we find that there appear fluctuations of these quantities. The magnitudes of these fluctuations determine the applicability of the semiclassical approximation involved in the present treatment. To estimate the validity of the semiclassical approach, we shall calculate the mean square variance of these operators. For the mean square variance of the exciton number

$$(\delta \mathcal{N})^2 = \langle \Psi | (\hat{\mathcal{N}} - \mathcal{N})^2 | \Psi \rangle \equiv \langle \hat{\mathcal{N}}^2 \rangle - \mathcal{N}^2$$

we have

$$(\delta \mathcal{N})^2 = \langle \Psi | \sum_m P_m^+ P_m - \sum_{n \neq m} P_n^+ P_m^+ P_n P_m - \mathcal{N}^2 | \Psi \rangle \equiv \mathcal{N} - \sum_n |\psi_n|^4.$$
(29)

Similarly, for the momentum and energy fluctuation we obtain

$$(\delta \hat{P}_{ex})^{2} = \frac{\hbar^{2}}{4R_{0}^{2}} \Biggl\{ \sum_{m} \langle \Psi | 2P_{m}^{+}P_{m} - (P_{m+1}^{+}P_{m-1} + \text{h.c.}) + 2P_{m}^{+}P_{m}(P_{m+1}^{+}P_{m-1} + P_{m-1}^{+}P_{m+1} - P_{m-1}^{+}P_{m-1}^{+}) |\Psi\rangle \Biggr\}.$$
(30)

$$(\delta \mathcal{H})^{2} = J^{2} \left\{ \sum_{m} \langle \Psi | 2P_{m}^{+}P_{m} + (P_{m+1}^{+}P_{m-1} + h.c.) + 2P_{m}^{+}P_{m}(P_{m+1}^{+}P_{m-1} + P_{m-1}^{+}P_{m+1} + P_{m+1}^{+}P_{m+1}P_{m-1}^{+}P_{m-1}^{+}) | \Psi \rangle \right\} + UJ \sum_{m} \langle \Psi | P_{m-1}^{+}P_{m+1}^{+}P_{m}P_{m+1} + P_{m-1}^{+}P_{m+1}^{+}P_{m}P_{m-1} + h.c. | \Psi \rangle + U^{2} \sum_{m} \langle \Psi | P_{m-1}^{+}P_{m-1}P_{m}^{+}P_{m}P_{m+1}^{+}P_{m+1} | \Psi \rangle.$$
(31)

Using the definition of the POCS and after going to the continuum limit, the mean square variance of the momentum fluctuation becomes

$$(\delta \hat{P}_{ex})^2 \approx \frac{\hbar^2}{2R_0^2} \int \frac{\mathrm{d}x}{R_0} \left(1 - 2|\psi|^2\right) \{2R_0^2 \left[(1 - |\psi|^2)|\psi_x|^2 - (1/2)((|\psi|^2)_x)^2 - |\psi|^4\right]\}.$$
 (32)

Analogously, after some extensive calculation, we obtain the following approximate expression for the energy fluctuations:

$$(\delta\mathcal{H})^{2} \approx 2J^{2} \int \frac{\mathrm{d}x}{R_{0}} (1 - 2|\psi|^{2}) \left\{ 2|\psi|^{2} - |\psi|^{4} - 2R_{0}^{2} \left[ (1 - |\psi|^{2})|\psi_{x}|^{2} - \frac{R_{0}^{2}}{2} ((|\psi|^{2})_{x})^{2} \right] \right\} + UJ \left\{ \int \frac{\mathrm{d}x}{R_{0}} \left[ 4|\psi|^{4} (1 - |\psi|^{2}) - 3R_{0}^{2} ((|\psi|^{2})_{x})^{2} - 2R_{0}^{2} (1 - |\psi|^{2})|\psi|^{2}|\psi_{x}|^{2} + 4R_{0}^{2} |\psi|^{2} ((|\psi|^{2})_{x})^{2} \right] \right\} + U^{2} \int \frac{\mathrm{d}x}{R_{0}} |\psi|^{6}.$$
(33)

These relations are valid equally for 'bright' and 'dark' solitons. Substituting the explicit expression for the particular solutions and keeping the most dominant terms only, we found that the momentum and energy mean square variances are both proportional to  $(\delta N)^2$ , i.e.  $\delta \hat{P}_{ex} / \hat{P}_{ex} \sim \delta N / N$  and  $\delta H / E \sim \delta N / N$ . Obviously the relative mean variance of the exciton number  $\delta N / N$  measures the magnitude of the quantum fluctuations and in such a way practically determines the degree of the validity of the semiclassical approximation. The mean square variance of the particle number is explicitly given by

$$(\delta \mathcal{N})^2 = \begin{cases} \mathcal{N}\left(1 - \frac{\mu \mathcal{N}^2}{3}\right) & \text{`bright' soliton} \\ \mathcal{N}(1 - O(N^{-1})) & \text{`dark' soliton.} \end{cases}$$
(34)

Thus the relative variance of the exciton number approaches  $\delta N/N \leq 1/N^{1/2}$ , which provides the applicability of the semiclassical approximation in the limit of high exciton concentrations. Note, however, that  $(\delta N)^2 > 0$ , so by virtue of equation (29) which may be written as

$$(\delta \mathcal{N})^2 > 0 = \sum_m |\psi_m|^2 (1 - |\psi_m|^2)$$

the number of excitons participating in the soliton formation is limited. This condition is, however, practically identical to the previously underlined demand for the smallness of the magnitude of the exciton density which, as discussed in sections 2 and 3, practically means that  $\psi_0^2 < 1$ , and therefore the previously emphasized conditions for the soliton existence (12), (17), (26) and (27) practically provide also the applicability of the semiclassical concept.

In order to estimate the accuracy of the continuum limit, let us examine the influence of the terms neglected in equation (9). For that purpose we collect all the terms of the next order in the product of the exciton density and its derivative. Adding these corrections to functional (8) we obtain

$$\mathcal{H} = JR_0^2 \int \frac{\mathrm{d}x}{R_0} |\psi_x(x)|^2 + G(v) \int \frac{\mathrm{d}x}{R_0} |\psi(x)|^4 - (5J + 2U) \int \frac{\mathrm{d}x}{R_0} |\psi(x)|^2 |\psi_x(x)|^2 \quad (35)$$

where the term proportional to  $\tilde{\Delta}$ , irrelevant for the further analysis, has been omitted. The equation for the soliton envelope which follows from this functional is

$$JR_0^2\psi_{xx} - (5J+2U)R_0^2\left[(\psi_x(x)|\psi(x)|^2)_x - |\psi_x|^2\psi\right] - 2G(v)|\psi(x)|^2\psi = 0.$$
(36)

Here, we do not need to look for the explicit solution for  $\psi(x)$ , and for the estimation of this term in the soliton properties we can use a simple qualitative approach based upon the virial theorem [26]. Thus we perform the scale change  $x \to \mu x$ . For the 'bright'-soliton solutions, according to norm conservation,  $\psi(x)$  must scale as follows:  $\psi(x) \to \mu^{1/2} \psi(\mu x)$ , and the above functional becomes

$$\mathcal{H}(\mu) = \mu^2 E_k + \mu E_p - \mu^3 E'_p \tag{37}$$

where

$$E_{k} = JR_{0}^{2} \int \frac{\mathrm{d}x}{R_{0}} |\psi_{x}(x)|^{2} \qquad E_{p} = G(v) \int \frac{\mathrm{d}x}{R_{0}} |\psi(x)|^{4}$$

denote the 'kinetic' and 'potential' energy, respectively, while

$$E'_{p} = (5J + 2U) \int \frac{\mathrm{d}x}{R_{0}} |\psi_{x}(x)|^{2} |\psi(x)|^{2}$$

represents the correction arising on account of previously disregarded terms. Stability of the soliton solution demands  $(\partial \mathcal{H}(\mu)/\partial \mu)_{\mu=1} = 0$ ;  $(\partial^2 \mathcal{H}(\mu)/\partial \mu^2)_{\mu=1} > 0$  which lead to:  $2E_k + E_p - 3E'_p = 0$  and  $2E_k - 6E'_p > 0$ , leading to an improved stability condition:  $E_k + E_p < 0$ . With respect to the previously obtained one, where stable solutions exist for J > 0 only, this relation opens up the possibility for

- (i) stabilization of the solutions corresponding to J < 0 and G > 0, and
- (ii) existence of new solutions even in the case where both J and G(v) are negative.

Let us now examine these possibilities in detail. First, we shall analyse how these changes affect the solution examined in section 3.1 assuming that the conditions found there, equations (17) and (26), are satisfied. For that purpose we first estimate the changes of the soliton width due to this correction using the direct variational method and treating the soliton solution (10) as a trial state with *l* being the variational parameter. In such a way we find that the modified soliton width, measured in units of the lattice constant ( $\tilde{l} = l/R_0$ ), is given as

$$\tilde{l} = \tilde{l}_0 \, 3|G(v)|(5J-2|U|)\mathcal{N}^2 \Big/ \Big\{ 10J^2 \bigg[ 1 - \sqrt{1 - \frac{6\mathcal{N}^2(5J-2|U|)}{5J\tilde{l}_0}} \bigg] \Big\}.$$
(38)

Assuming that the above-quoted conditions are satisfied, in particular that for applicability of the continuum limit  $\tilde{l} \gg 1$ , one can see that the soliton relative width satisfies

$$\tilde{l} \approx \tilde{l}_0 / \left( 1 + \frac{3N^2(5J - 2|U|)}{10J^2} \right).$$
 (39)

It follows that, in the system of the so-called longitudinal excitons (J > 0) and if the conditions (18) are satisfied, these corrections do not induce any special modification of the soliton solutions. That is, since  $E_k \sim J$  and  $E_p \sim G(v)$ , one can see that the modified stability condition demands attractive exciton–exciton interaction, so it is practically equivalent to the stability condition (12). The only change is the decrease of the soliton width. Stabilization of the bright solitons in the system of transverse excitons (J < 0) and JG(v) > 0 is not a realistic possibility. That is, G(v) > 0 requires  $U - 2|J| - m_{ex}v^2/2 > 0$  which, due to the smallness of the exciton–exciton interaction with respect to the dipole–dipole one, cannot be realized in actual systems even if one takes into account exciton–phonon interaction. That is, effective exciton–exciton interaction arising due to the coupling with the lattice is always attractive, and cannot change the sign of G(v). On the other hand, if G(v) < 0, the modified stability condition is always satisfied while the direct variation leads to the following expression for the relative inverse soliton width:

$$\mu = \frac{5|J|}{3(5|J| - U)\mathcal{N}^2} \left[ 1 + \sqrt{1 + \frac{3|G(v)|(5|J| - 2U)\mathcal{N}^2}{5J^2}} \right].$$
 (40)

Demanding the applicability of the continuum approximation  $\mu \ll 1$ , from this equation we found the following condition:

$$\mathcal{N}^2 \gg \frac{2+G(v)/J}{3(1-U/J)}.$$
 (41)

On the other hand, smallness of the exciton density demands  $\psi_0^2 < 1$ , which together with the expression for  $\mu$  imposes the following limitation on the exciton number:

$$\mathcal{N}^2 < \frac{2J}{G(v)} \left( 1 + \frac{3U}{5J} \right). \tag{42}$$

These conditions cannot be satisfied simultaneously, so these 'bright'-soliton solutions cannot be formed.

A similar procedure may be applied also to the 'dark' solitons but with the proper scaling rule for the 'dark'-soliton solution:  $\psi(x) \rightarrow \psi(\mu x)$ . In this case, the functional  $\mathcal{H}$  scales as follows:

$$\mathcal{H}(\mu) = \mu(E_k - E'_p) + \mu^{-1}E_p.$$
(43)

From the stationarity of  $\mathcal{H}$  we have  $(E_k - E'_p) - E_p = 0$ , while the stability condition gives  $2E_p > 0$  which, combined with the above condition, results in  $E_k - E'_p > 0$ . This however, cannot substantially violate the stability of the previously analysed 'dark' solitons. That is, the above condition may be specified as  $J - (5J + 2U)\psi_0^2 > 0$ , which, due to the smallness of the soliton amplitude, provides stability of the 'dark' solitons even in the presence of these corrections. The only influence of these additional terms is reflected through the change (in fact shrinking) of the soliton width. This can be seen from the fact that the variational parameter  $\mu$ , which may be identified with the soliton inverse relative width, is in this case given by

$$\mu = \sqrt{\frac{E_p}{E_k - E'_p}}$$

which is bigger than in the unperturbed case:  $E'_p = 0$ .

# 5. Concluding remarks

Concluding this paper, let us note that by formulating the theory in terms of the amplitude of the exciton density we are offering, for the present system, a more justified physical picture, with quite obvious meaning, than when describing the system in terms of 'polarization'. Thus our 'bright'-soliton solution corresponds to the  $\mathcal{N}$ -exciton bound state while the 'dark' soliton describes the defect in the homogeneous exciton distribution. In contrast to the results of references [10–12], here we have shown that the whether 'bright' or 'dark' solitons will form in the Frenkel exciton system depends on the sign and the magnitude of the energy parameters of the system but not on the magnitude of the exciton density. In particular, we found that neither 'bright' nor 'dark' solitons can exist if the effective dynamical interaction is a repulsive one. Thus soliton existence in the system considered demands  $U_{eff} < 0$ .

Furthermore, explicit accounting for the conservation of the exciton number through the normalization of the soliton wave function allows soliton parameters (amplitude, width, energy, momentum and effective mass) to be expressed in terms of the exciton number, whose value appears to be, for the given set of system parameters, the main limiting factor as regards soliton existence.

Soliton motion has quite different consequences for its properties to those predicted in [12] within the so-called semicontinuum approximation. Thus, transcribing the results of reference [12] employing the continuum approximation and utilizing the connection between the soliton velocity and wave vector of the carrier wave, it follows that both the nonlinear term and the effective transfer integral decrease as the velocity rises, which means that motion causes the shrinking of the 'bright' soliton, supporting its stability, and violates the stability of the 'dark' soliton, but increases its width.

Finally, let us mention the interesting recent papers of Konotop and Takeno where a related problem has been studied, with account taken of discreteness effects [18, 19]. The subject of these studies was the system of excitons with exchange and dipole–dipole interaction which, under certain circumstances, could be mapped onto our problem. However, most of their studies were performed within the discrete (lattice) model. For that reason, it is difficult to establish a proper correspondence of our results with theirs. However, one can see that they did not attempt to establish a correspondence between the soliton properties and the number of excitons. Moreover, they did not find the explicit expressions for the energy and momenta of the corresponding solutions, so they were not in a position to derive an energy–momentum relationship. For this reason, the nature of these nonlinear solutions (and their possible soliton character) still remains unknown. Of course, we are aware of the problem of defining the momentum in the discrete treatment without resorting to some continuum (or semicontinuum) approximation.

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