EFFECTS OF NOISE AND NONLOCAL INTERACTIONS IN NONLINEAR DYNAMICS OF MOLECULAR SYSTEMS

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We show that the NLS systems with multiplicative noise, nonlinear damping and nonlocal dispersion exhibit a variety of interesting effects which may be useful for modelling the dynamical behavior of one- and two-dimensional systems.

1 INTRODUCTION

We consider the nonlinear Schrödinger (NLS) system with cubic nonlinearity. This system models optical media, molecular thin films in the continuum limit, deep water waves and many other physical systems which exhibit weak nonlinearity and strong dispersion. The effects we shall discuss are observed in our earlier works \([1, 3, 6]\) and cited therein.

The motivation for studying the 2-D NLS equation with thermal fluctuations is our intention to model efficient energy transfer in Scheibe-aggregates. Our starting point is a two-dimensional Davydov model with nonlinear coupling between the exciton and phonon system, and white noise in the phonon system. In Section 2 we derive a single equation for the exciton system with multiplicative colored noise and a nonlinear damping term. In the continuum limit the collective coordinate approach indicates that an energy balance between energy input (from the noise term) and dissipation can be established. Thus, this model may describe the state of thermal equilibrium in the molecular aggregate. The coherent exciton moving on the aggregate \([10, 11]\) is...
modelled by the ground state solution to the 2-D NLS equation, and the lifetime has been related to the collapse time of the ground state \([4]\). For sufficiently strong nonlinearity the thermal fluctuations will slow down the collapse. As a result this lifetime increases with the variance of the fluctuations, i.e. the temperature.

In Section 3 the role of long-range dispersive interaction in the one-dimensional molecular system is investigated. Dispersive interactions of two types (the power and the exponential dependences of the interaction intensity on the distance) are studied. If the interaction decreases with the distance slowly, there is an interval of bistability where two stable stationary states: narrow, pinned states and broad, mobile states exist at each value of the excitation energy. For cubic nonlinearity the bistability of the solitons occurs already for dipole-dipole dispersive interaction. We demonstrate a possibility of the controlled switching between pinned and mobile states applying a spatially symmetric perturbation in the form of a parametric kick. The mechanism could be important for controlling energy storage and transport in molecular systems.

2 NOISE AND DAMPING

Following the derivation given in Ref. \([4]\), we start by assuming that the coupled exciton-phonon system can be described by the following pair of equations

\[
i\hbar \dot{\psi}_n + \sum_{n'} J_{nn'} \psi_{n'} + \chi u_n \psi_n = 0 , \quad (1)
\]

\[
M \ddot{u}_n + M \lambda \dot{u}_n + M \omega_0^2 u_n - \chi |\psi_n|^2 = \eta_n(t) . \quad (2)
\]

Here \(\psi_n\) is the amplitude of the exciton wave function corresponding to site \(n\) and \(u_n\) represents the elastic degree of freedom at site \(n\). Furthermore, \(-J_{nn'}\) is the dipole-dipole interaction energy, \(\chi\) is the exciton-phonon coupling constant, \(M\) is the molecular mass, \(\lambda\) is the damping coefficient, \(\omega_0\) is the Einstein frequency of each oscillator, and \(\eta_n(t)\) is an external force acting on the phonon system. To describe the interaction of the phonon system with a thermal reservoir at temperature \(T\), \(\eta_n(t)\) is assumed to be Gaussian white noise with zero mean and the autocorrelation function

\[
\langle \eta_n(t) \eta_{n'}(t') \rangle = 2M \lambda k_B T \delta(t - t') \delta_{nn'} , \quad (3)
\]
in accordance with the fluctuation-dissipation theorem ensuring thermal equilibrium.

In order to derive a single equation for the dynamics of the exciton system, we start by writing the solution to Eq. (2) in the integral form. Neglecting all exponentially decaying transient terms and making the additional assumption that \( \psi_n \) varies slowly in space and that only nearest-neighbor coupling \( J \) is of importance, we obtain \[ 3 \] in the continuum approximation for the continuous exciton field \( \psi(x, y, t) \equiv \sqrt{V/Je^{-4iJt/\hbar}} \psi_n(t) \) the equation of motion

\[
i\psi_t + \nabla^2 \psi + |\psi|^2 \psi - \Lambda \psi(|\psi|^2)_t + \sigma \psi = 0, \tag{4}
\]

where \( x \) and \( y \) are scaled on the distance \( \ell \) between nearest neighbors, a noise density \( \sigma(x, y, t) \) is not white, but strongly colored \[ 1 \], and time was transformed into the dimensionless variable: \( Jt/\hbar \rightarrow t \). The nonlinear damping parameter \( \Lambda = \frac{\lambda J}{\hbar \omega_0^2} \).

It can easily be shown that in spite of the presence of the nonlinear damping and multiplicative noise terms in Eq. (4), the norm, defined as

\[
N = \int \int |\psi(x, y, t)|^2 dx dy \tag{5}
\]

will still be a conserved quantity.

To investigate the influence on the collapse process of the damping and noise terms in Eq. (4), we will use the method of collective coordinates. To this end, we will make some simplifying assumptions. We will assume isotropy, which effectively reduces the problem to one space dimension with the radial coordinate \( r = \sqrt{x^2 + y^2} \). We also assume that the noise \( \sigma \) can be approximated by radially isotropic Gaussian white noise. The validity of the approximation was discussed in Ref. \[ 1 \]. Finally, we assume that the collapse process can be described in terms of collective coordinates using the following self-similar trial function for the exciton wave function \( \psi(r, t) \)

\[
\psi(r, t) = A(t) \text{sech} \left( \frac{r}{B(t)} \right) e^{i\alpha(t)r^2}. \tag{6}
\]

This trial function, with three real time-dependent parameters \( A, B, \) and \( \alpha \) determining the amplitude, width, resp. phase of the wave function, was used in Refs. \[ 12, 2 \] to investigate the case when \( \Lambda = 0 \) in Eq. (4). The choice of
this particular type of trial function can be motivated by regarding it as a
generalization of the approximate ground state solution to the ordinary 2-D
NLS found in Ref. [5]. From the definition (5) of the norm, we immediately
obtain the relation between amplitude and width $A(t) \sim \sqrt{N}/B(t)$. In
analogy with the treatment for the undamped case in Ref. [2], we find that
it is possible to arrive to the following differential equation for the width $B$
of the exciton wave function

$$\ddot{B} = \frac{\Delta}{B^3} - \frac{\Gamma \dot{B}}{B^4} + \frac{h(t)}{B^2},$$

where the constants $\Delta$ and $\Gamma$ are some functions of $N$ and $\Lambda$. Note that
$\Delta$ and $\Gamma$ depend on the initial conditions via $N$, and that while $\Delta$ can be
either positive or negative, $\Gamma$ is always positive. In the absence of noise and
damping the collapse will occur if and only if $\Delta < 0$. The white noise $h(t)$
has the autocorrelation

$$\langle h(t)h(t') \rangle = 2D\delta(t - t'),$$

where $D$ is the dimensionless noise variance.

Our numerical calculations [3] show that for $D < D_{\text{crit}} \approx 0.15$, the effect
of the noise is to delay the pseudo-collapse in terms of the ensemble average
of the width, in analogy with the similar result obtained in Ref. [12] for the
undamped case. For $D > D_{\text{crit}}$, we observe a non-monotonic behavior of
$\langle B(t) \rangle$. Initially, the average width will decrease in a similar way as when
$D < D_{\text{crit}}$, but after some time $\langle B(t) \rangle$ will reach a minimum value and
diverge as $t \to \infty$. This is due to the fact that for $D > D_{\text{crit}}$, the noise is
strong enough to destroy the pseudo-collapse and cause dispersion for some
of the systems in the ensemble. As $t \to \infty$ the dominating contribution
to $\langle B(t) \rangle$ will come from the dispersing systems for which $B \to \infty$, and
consequently $\langle B(t) \rangle$ will diverge for $D > D_{\text{crit}}$. The minimum value of
$\langle B(t) \rangle$ will increase towards $B(0)$ as $D$ increases.

3 NONLOCAL INTERACTIONS

In the main part of the previous studies of the discrete NLS models the
dispersive interaction was assumed to be short-ranged and a nearest-neighbor
approximation was used. However, there exist physical situations that def-
initely can not be described in the framework of this approximation. The
DNA molecule contains charged groups, with long-range Coulomb interaction $(1/r)$ between them. The excitation transfer in molecular crystals and the vibron energy transport in biopolymers are due to transition dipole-dipole interaction with $1/r^3$ dependence on the distance, $r$. The nonlocal (long-range) dispersive interaction in these systems provides the existence of additional length-scale: the radius of the dispersive interaction. We will show that it leads to the bifurcative properties of the system due to both the competition between nonlinearity and dispersion, and the interplay of long-range interactions and lattice discreteness.

In some approximation the equation of motion is the nonlocal discrete NLS equation of the form

$$\frac{i}{dt}\psi_n + \sum_{m \neq n} J_{n-m}(\psi_m - \psi_n) + |\psi_n|^2 \psi_n = 0,$$

where the long-range dispersive coupling is taken to be either exponentially, $J_n = J e^{-\beta |n|}$, or algebraically, $J_n = J |n|^{-s}$, decreasing with the distance $n$ between lattice sites. The parameters $\beta$ and $s$ are introduced to cover different physical situations from the nearest-neighbor approximation ($\beta \to \infty$, $s \to \infty$) to the quadrupole-quadrupole ($s = 5$) and dipole-dipole ($s = 3$) interactions. The equation (9) conserves the number of excitations $N = \sum_n |\psi_n|^2$.

We are interested in stationary solutions of Eq. (9) of the form $\psi_n(t) = \phi_n \exp(i\Lambda t)$ with a real shape function $\phi_n$ and a frequency $\Lambda$. This gives the governing equation for $\phi_n$.

$$\Lambda \phi_n = \sum_{m \neq n} J_{n-m}(\phi_m - \phi_n) + \phi_n^3.$$  \hspace{1cm} (10)

Figure 1 shows the dependence $N(\Lambda)$ obtained from direct numerical solution of Eq. (10) for algebraically decaying $J_{n-m}$. A monotonic function is obtained only for $s > s_{cr}$. For $2 < s < s_{cr}$ the dependence becomes nonmonotonic (of $N$-type) with a local maximum and a local minimum. These extrema coalesce at $s = s_{cr} \approx 3.03$. For $s < 2$ the local maximum disappears. The dependence $N(\Lambda)$ obtained analytically using the variational approach is in a good qualitative agreement with the dependence obtained numerically (see 1). Thus the main features of all discrete NLS models with dispersive interaction $J_{n-m}$ decreasing faster than $|n-m|^{-s_{cr}}$ coincide qualitatively with
Figure 1: Number of excitations, $N$, versus frequency, $\Lambda$, found numerically from Eq. (10) for $s = \infty$ (full), 4 (dotted), 3 (short-dashed), 2.5 (long-dashed), 2 (short-long-dashed), 1.9 (dashed-dotted).

the features obtained in the nearest-neighbor approximation where only one stationary state exists for any number of excitations, $N$. However in the case of long-range nonlocal NLS equation (9), i.e. for $2 < s < s_{cr}$, there exist for each $N$ in the interval $[N_l(s), N_u(s)]$ three stationary states with frequencies $\Lambda_1(N) < \Lambda_2(N) < \Lambda_3(N)$. In particular, this means that in the case of dipole-dipole interaction ($s = 3$) multiple solutions exist. It is noteworthy that similar results are also obtained for the dispersive interaction of the exponentially decaying form. In this case the bistability takes place for $\beta \leq 1.67$. According to the theorem which was proven in [9], the necessary and sufficient stability criterion for the stationary states is $dN/d\Lambda > 0$. Therefore, we can conclude that in the interval $[N_l(s), N_u(s)]$ there are only two linearly stable stationary states: $\Lambda_1(N)$ and $\Lambda_3(N)$. The intermediate state is unstable since $dN/d\Lambda < 0$ at $\Lambda = \Lambda_2$.

The low frequency states are wide and continuum-like while the high fre-
frequency solutions represents intrinsically localized states with a width of a few lattice spacings. It can be shown that the existence of two so different soliton states for one value of the excitation number, $N$, is due to the presence of two different length scales in the system: the usual scale of the NLS model which is related to the competition between nonlinearity and dispersion (expressed in terms of the ratio $N/J$) and the range of the dispersive interaction $\xi$.

Having established the existence of bistable stationary states in the non-local discrete NLS system, a natural question that arises concerns the role of these states in the full dynamics of the model. In particular, it is of interest to investigate the possibility of switching between the stable states under the influence of external perturbations, and to clear up what type of perturbations can be used to control the switching. Switching of this type is important for example in the description of nonlinear transport and storage of energy in biomolecules like the DNA, since a mobile continuum-like excitation can provide action at distance while the switching to a discrete, pinned state can facilitate the structural changes of the DNA [7]. As it was shown recently in [8], switching will occur if the system is perturbed in a way so that an internal, spatially localized and symmetrical mode ('breathing mode') of the stationary state is excited above a threshold value.

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