Selftrapping and Quantum Fluctuations in the Discrete Nonlinear Schrödinger Equation*

C.A. Bustamante and M. I. Molina

Facultad de Ciencias, Departamento de Física, Universidad de Chile
Casilla 653, Las Palmeras 3425, Santiago, Chile.
molina@abello.dic.uchile.cl

Abstract

We study the robustness of the selftrapping phenomenon exhibited by the Discrete Nonlinear Schrödinger (DNLS) equation against the effects of nonadiabaticity and quantum fluctuations in a two–site system (dimer). To test for nonadiabatic effects (in a semiclassical context), we consider the dynamics of an electron (or excitation) in a dimer system and coupled to the vibrational degrees of freedom, modeled here as classical Einstein oscillators of mass $M$. For relaxed (coherent state) oscillators initial condition, the DNLS selftrapping transition persists for a wide range of $M$ spanning 5 decades. When undisplaced initial conditions are used, the selftrapping transition is destroyed for masses greater than $M \sim 0.02$. To test for the effects of quantum fluctuations, we performed a first-principles numerical calculation for the fully quantum version of the above system: the two–site Holstein model. We computed the long-time averaged probability for finding the electron at the initial site as a function of the asymmetry and nonlinearity parameters, defined in terms of the electron-phonon coupling strength and the oscillator frequency. Substantial departures from the usual DNLS system are found: A complex landscape in asymmetry–nonlinearity phase space, which is crisscrossed with narrow “channels”, where the average electronic probability on the initial site remains very close to 1/2, being substantially larger outside. In the adiabatic case, there are also low–probability “pockets” where the average electronic probability is substantially smaller than 1/2.

PACS: 71.38.+i, 63.20.kr, 63.20.pw

*Invited talk to the International Workshop on Disordered Systems with Correlated Disorder, Universidad de Tarapacá, Arica, August 31-September 5, 1998
1 Introduction

The Discrete Nonlinear Schrödinger (DNLS) equation is a paradigmatic equation that describes properties of chemical, optical or condensed matter systems where selftrapping mechanisms are present. It can describe the dynamics of a set of anharmonic oscillators, the power exchange between nonlinear optical couplers or the motion of a quantum mechanical particle propagating in a discrete medium while interacting strongly with vibrations. In this last case, for an electron (or excitation) propagating in a discrete one-dimensional medium, DNLS has the form

\[
i \frac{dC_n}{dt} = V(C_{n+1} + C_{n-1}) - \chi |C_n|^2 C_n
\]

where \(C_n(t)\) is the probability amplitude of finding the electron on site \(n\) at time \(t\), \(V\) is the nearest-neighbors hopping term and \(\chi\) is the nonlinearity parameter, proportional to the square of the (strong) electron-phonon coupling on site \(n\). The DNLS equation can be “derived” as the antiadiabatic limit of the problem of a quantum excitation propagating in a lattice and coupled to the vibrational degrees of freedom of each site, modelled as a set of classical Einstein oscillators

\[
i \frac{dC_n}{dt} = V(C_{n+1} + C_{n-1}) + \alpha u_n C_n
\]

\[M \ddot{u}_n + k u_n = -\alpha |C_n|^2
\]

After the transients, the oscillator amplitude \(u_n(t)\) is given by \(u_n = (1/\sqrt{M k}) \int_0^t dt' \sin(\omega(t-t')|C_n(t')|^2).\) If we assume the oscillator time scale much shorter than the electronic time scale for hopping, then we have approximately, \(u_n \approx (\alpha/k)|C_n(t)|^2.\) Inserting this into (2), leads to the DNLS equation (1), with \(\chi = \alpha^2/k.\) Perhaps the most striking feature of DNLS is that it leads to selftrapping: for nonlinearity parameter values exceeding certain threshold (whose exact value depends on the details of the initial electron state), a portion of the electron remains bound to a particular site(s), while the rest propagates away in a ballistic manner. The effect is most noticeable when completely localized initial conditions are used.

The ability to partially trap an electron, suggests a possible mechanism for tuning the electrical conductivity. It is then clear that the robustness of the selftrapping phenomenon exhibited by the DNLS equation, against perturbing effects commonly found in realistic situations must be ascertained in order to implement possible applications. In this work we focus on two effects: finite inertia and quantum fluctuations. In order to keep the discussion simple, we will work on the smallest nontrivial DNLS unit: the nonlinear dimer.

This paper is organized as follows: In section 2 we review the DNLS equation for the symmetric nonlinear dimer and its selftrapping phenomenology. We also consider a generalized nonlinear dimer. In section 3, we introduce finite oscillator inertia and examine its consequences on selftrapping. In section 4 we examine the fully quantum mechanical problem, given by the two-site Holstein model. We further specialize to the symmetric (section 4.1) and general (section 4.2) cases focussing on selftrapping properties. We end with a brief discussion of the preliminary results found so far.
2 DNLS dimer

In the case of two sites, system (1) reduces to

\[
\begin{align*}
\frac{idC_1}{dt} &= V C_2 - \chi |C_1|^2 C_1 \\
\frac{idC_2}{dt} &= V C_1 - \chi |C_2|^2 C_2
\end{align*}
\] (4)

For completely localized initial conditions $|C_1(0)|^2 = 1$, an exact time-dependent solution for $C_1(t)$ and $C_2(t)$ can be found\cite{5}. The solution exhibits a *selftrapping transition*: the probability at the initial site $|C_1(t)|^2$, oscillates in a periodic fashion for $\chi < 4V$, with a period that increases with nonlinearity. At the critical value $\chi = 4V$ the period becomes infinite and for $\chi > 4V$ the particles selftraps. The long time-averaged probability $<|C_1|^2>$ obeys\cite{6}

\[
< |C_1|^2 > = \begin{cases} 
1/2 & \chi < 4V \\
1/2 \left( 1 + \frac{\pi}{2K(4V/\chi)} \right) & \chi > 4V
\end{cases}
\] (5)

where $K$ is the complete elliptic integral of the first kind. Another closely related system, the *nondegenerate* nonlinear dimer, considers an additional site energy difference in (4) and can be similarly solved\cite{7} in terms of jacobian elliptic functions. It is found that there is a transition line in parameter space that separates the localized from nonlocalized states. The difference with the previous (degenerate) case is that now the selftrapped state can be accessed without crossing the transition line.

We can pose the most general nonlinear dimer by incorporating, besides a site energy mismatch, different nonlinearity parameters:

\[
\begin{align*}
\frac{idC_1}{dt} &= \delta C_1 + V C_2 - \chi_1 |C_1|^2 C_1 \\
\frac{idC_2}{dt} &= V C_1 - \chi_2 |C_2|^2 C_2
\end{align*}
\] (6)

The transformation $\mathcal{C}_i = C_i \exp\left[ (1/2)(\chi_1 - \chi_2)|C_2|^2 \right]$ along with the normalization condition $|C_1|^2 + |C_2|^2 = 1$ maps (6) into

\[
\begin{align*}
\frac{id\mathcal{C}_1}{dt} &= \Delta \mathcal{C}_1 + V \mathcal{C}_2 - \chi |\mathcal{C}_1|^2 \mathcal{C}_1 \\
\frac{id\mathcal{C}_2}{dt} &= V \mathcal{C}_1 - \chi |\mathcal{C}_2|^2 \mathcal{C}_2
\end{align*}
\] (7)

where $\chi = (1/2)(\chi_1 + \chi_2)$ and $\Delta = \delta - (1/2)(\chi_1 - \chi_2)$. Therefore, if we are only interested in site occupation probabilities, the general nonlinear dimer is equivalent to the one studied earlier by Tsironis\cite{7}. In Fig. 1 we show a probability density phase diagram for $<P>_T$ in $\Delta-\chi$ space. The transition line, which starts at $\chi = 3.09$, $\Delta = -0.383$, is clearly visible.

3 Semiclassical dimer: finite inertia effects

As we have mentioned, the DNLS equation is obtained in the limit of a negligible oscillator inertia in (3), where the oscillator adjust instantaneously to the electronic presence. We
now examine the robustness of the selftrapping phenomenon when finite oscillator inertia is taken into account.

While in the limits of both small and large mass, analytical work is possible via perturbation theory, we numerically integrate the dynamical equations for the full dimer system, equations (2) and (3) with \( n = 1, 2 \). The electron is placed initially completely on site 1: \( C_1(0) = 1 \). We explore two possible oscillator initial conditions: relaxed type, where oscillator 1 starts from rest with a displacement determined assuming a complete oscillator relaxation \( u_1(0) = (\alpha/k)|C_1(0)|^2 = \alpha/k, \dot{u}_1(0) = 0 \), and undisplaced, where \( u_1(0) = 0 = \dot{u}_1(0) \). For oscillator 2, \( u_2(0) = 0 = \dot{u}_2(0) \) in both cases. The coupled equations are solved using a fourth-order Runge-Kutta scheme, whose precision is monitored through the conservation of probability (norm).

We examine the effect of nonlinearity on the time-averaged probability at the initially occupied site \( \langle P \rangle_T = \frac{1}{T} \int_0^T |C_1(t)|^2 dt \) (with \( T \gg 1/V, \sqrt{M/k} \)), as an indicator for the occurrence of selftrapping. Results are shown in Fig. 2. We note that, for relaxed initial conditions, the finite inertia of the oscillators plays practically no role in the existence and location of the selftrapping transition, for at least five decades in \( M \). There is a slight shift in the critical nonlinearity towards smaller values, as \( M \) is increased. On the contrary, for undisplaced initial conditions, the selftrapping transition is obliterated when \( M \) exceeds 0.02 approximately. Since relaxed initial conditions are the “natural” ones when the oscillators have small mass, we conclude that the DNLS selftrapping transition is robust against finite inertia effects. Similar conclusions have been reached in the case of a number of finite vibrational impurities embedded in a linear chain.

4 Quantum dimer

We come now to the most delicate effect: quantum fluctuations. The DNLS equation was “derived” for the coupled electron-phonon system by making two approximations. The oscillators were first treated classically. Later, an antiadiabatic limit was taken to effectively eliminate the vibrational degrees of freedom, which become completely correlated (enslaved) with the electronic amplitude.

In this section, we drop the previous assumptions and perform a fully quantum mechanical calculation for the rather general two-site Holstein model

\[
H = H_{\text{el}} + H_{\text{ph}} + H_{\text{e-ph}} 
\]

with

\[
H_{\text{el}} = \delta C_1^\dagger C_1 - V(C_1^\dagger C_2 + C_2^\dagger C_1) 
\]

\[
H_{\text{ph}} = \omega (a_1^\dagger a_1 + a_2^\dagger a_2) 
\]

\[
H_{\text{e-ph}} = g_1 C_1^\dagger C_1(a_1^\dagger + a_1) + g_2 C_2^\dagger C_2(a_2^\dagger + a_2) 
\]

where \( \delta \) is a site energy asymmetry, \( C_j^\dagger(C_j) \) creates (destroys) an electron on the \( j \)th site, \( a_j^\dagger(a_j) \) is the usual phonon creation (destruction) operator at site \( j \), \( \omega \) is the (Einstein) oscillator frequency, chosen to be the same for both oscillators for simplicity and \( g_j \) is the electron-phonon coupling at site \( j \). The nonlinearity parameter used in DNLS corresponds
here to $\chi_j = g_j^2/\omega$. Previous work on a single Holstein impurity embedded in a finite chain have displayed significant departures from the DNLS behavior\cite{15}. In that case, it was proved that in the antiadiabatic limit, the Hamiltonian becomes equivalent to the one of a static, linear impurity and $<P>_T$ never converges to the DNLS profile.

We decouple one of the phonon fields in (8) using the canonical transformation

$$a = \frac{1}{G}(g_1 a_1 - g_2 a_2 - \frac{g_2^2}{\omega})$$
$$b = \frac{1}{G}(g_2 a_1 + g_1 a_2 + \frac{g_2 g_1}{\omega})$$  \hspace{1cm} (12)

where $G \equiv (g_1^2 + g_2^2)^{1/2}$ is an effective coupling. Using $\Delta \equiv \delta + (g_2^2 - g_1^2)/\omega$ as the asymmetry parameter, the transformed Hamiltonian reads

$$H = (\Delta + G^2/\omega)C_1^\dagger C_1 + V(C_1^\dagger C_2 + C_2^\dagger C_1) + \omega a^\dagger a + G C_1^\dagger C_1 (a + a^\dagger) + (\omega b^\dagger b - g_2^2/\omega)$$  \hspace{1cm} (13)

The Hamiltonian thus simplified allows for an exact numerical diagonalization scheme, where the eigenenergies and quantum amplitudes are computed by expanding in a phonon basis set\cite{10}. With the eigenenergies and eigenfunctions we are ready to compute dynamical observables. As the initial condition for the electron we choose the one that places it completely on one of the sites (“site 1”) and focus on the probability $P(t)$ for finding it there at an arbitrary time later. As before, we shall pay particular attention to the long-time average of $P(t)$, $<P>_T = (1/T) \int_0^T P(t) dt$, as well as $<P>_\infty = \lim_{T \to \infty} <P>_T$. For the phonon part, we use two different initial conditions: undispaced oscillators (i.e., zero phonons present) and oscillators naturally “relaxed” with the electron presence (a coherent state).

### 4.1 Symmetric quantum dimer

Let us consider the special situation $\Delta = 0$. In this case it is possible to prove that $<P>_\infty = 1/2$, independent on the initial conditions\cite{10}. The original Hamiltonian can be separated into two diagonal blocks $H_{\pm}$ with eigenvalues $\{\epsilon_{\pm}\}$ and eigenvectors $\{|\epsilon_{\pm}\rangle\}$. The probability $P(t)$ reads

$$P(t) = (1/2) + \sum_{\epsilon_+,\epsilon_-} C(|\epsilon_{\pm}\rangle) \cos((\epsilon_+ - \epsilon_-)t)$$  \hspace{1cm} (14)

where the coefficients $C(|\epsilon_{\pm}\rangle)$ depend on the initial conditions and on the eigenvectors $|\epsilon_{\pm}\rangle$. Barring any degeneracy, $<P>_\infty = 1/2$, regardless of the initial conditions.

In the rest of this section we will use the coupling parameter $g \equiv G/\sqrt{2}$ since it allows a better comparison with previous works. It corresponds to the coupling per site when $\delta = 0$.

We are particularly interested in the adiabatic ($\omega \ll V$) and antiadiabatic ($\omega \gg V$) limits. For the antiadiabatic case, we obtain

$$P(t) \approx (1/2)(1 + \cos(2 V_{\text{eff}} t))$$  \hspace{1cm} (15)

where the effective hopping term $V_{\text{eff}}$ decreases with $g/\omega$ as $V_{\text{eff}} = V \exp(-\alpha (g/\omega)^2)$ (with $\alpha = 0.434$ for $\omega \geq 10$). This is the well-known “polaronic narrowing” of the band, a limit
well understood which can be obtained by first performing a Lang-Firsov transformation on the Holstein Hamiltonian \( \mathcal{H} \), followed by the \( \omega/V \gg 1 \) limit. For the adiabatic regime, the behavior of the electron is similar to the previous case only in the limits \( g/\omega < 1 \) or \( g/\omega \gg 1 \). In the intermediate region, \( P(t) \) displays a complex behavior due to the simultaneous presence of many different frequency scales. \[1\]

Even though \( < P >_\infty = 1/2 \) would seem to preclude any selftrapping phenomenon like the one observed in DNLS, it is clear from (13), that the effective hopping can decrease to the point of “trapping” the electron for long times on the initial site. For a given observational time scale \( T \), we examine \( < P >_T \). Results are shown in Figs. 3a and 3b. We note that the behavior of \( < P >_T \) is analogous to its DNLS counterpart, with a “critical” value of \( g/\omega \) above which the electron tends to confine itself at the initial site, forming a “small polaron”, a localized electron with low mobility.

The transition region between free electron and a small polaron behavior, can be determined by the following criterion [13]: examination of the correlation \( \chi_{1,1} = < \phi_0 | C_1^\dagger C_1(a_1^\dagger + a_1^\dagger) | \phi_0 \rangle \) where \( | \phi_0 \rangle \) is the ground state. As \( g/\omega \) is increased, \( \chi_{1,1} \) decreases initially from zero, reaches a minimum value and rises again back to zero. The maximum value of its slope \( d\chi_{1,1}/d(g/\omega) \) determines the critical point \( (g/\omega)_c \) and the maximum (minimum) value of its second derivative \( d^2\chi_{1,1}/d(g/\omega)^2 \) mark the lower (upper) boundary of the transition region. This is shown in Fig. 3c. Other criteria, such as variations of the ground state energy or the examination of \( \chi_{1,0} = < \phi_0 | C_1^\dagger C_1(a_1^\dagger + a_1^\dagger) | \phi_0 \rangle \) give the same qualitative results. The features displayed in Fig. 3c seem to remain valid also for longer chains, as shown by Capone et al. [13] for \( N = 4 \) sites using exact diagonalization and by Romero et al. [14] for \( N = 32 \) sites, who relies on a variational scheme to obtain \( | \phi_0 \rangle \) plus an energetic criteria. The latter group propose for the “critical” line the relation \( (g/\omega)_c = 1 + (V/\omega)^{1/2} \). The agreement for the dimer case is good, as can be seen in Fig. 3c. According to Capone et al. [13], a criterion for the formation of a small polaron is the simultaneous occurrence of

\[
\left( \frac{g}{\omega} \right) > 1 \quad \text{and} \quad \left( \frac{g^2}{\omega^2} \right) > \frac{1}{2}
\]

that is, there must be a net phononic displacement \( g/\omega \) correlated with an energetic gain \( g^2/\omega \) from polaron formation greater than the kinetic energy (\( \sim 2V \)) lost due to electron localization.

### 4.2 General quantum dimer

The general quantum dimer is described by two parameters: the asymmetry \( \Delta = \delta + (g_2^2 - g_1^2)/\omega \) and the effective coupling \( G = (g_2^2 + g_3^2)^{1/2} \). We focus on the values for \( < P >_\infty \) in terms of \( \Delta \) and \( G \), and for the two phonon initial conditions: undisplaced and relaxed. Results are displayed in Fig. 4 in the form of probability density phase diagrams.

A comparative study of \( < P >_\infty \) in terms of \( \Delta \) requires to consider two scales. One of them is the hopping \( V \). For small \( G/\omega \) values, variations in \( < P >_\infty \) are determined by this scale. The other relevant scale is the frequency \( \omega \). When the phonons are strongly coupled to the electron (i.e., when \( G/\omega \) is large enough), the effective electronic hopping increases when the asymmetry value is a multiple of the frequency: \( \Delta \approx n\omega \) with \( n = 0, 1, 2, \ldots \) for relaxed initial conditions and \( n = 0, \pm 1, \pm 2, \ldots \) for undisplaced initial conditions. On these
“resonance” channels, $< P >_{\infty} = 1/2$. As $\omega$ is decreased, these channels become narrower and begin to distort considerably the low asymmetry region ($|\Delta| \leq 5$) giving rise to a probability pattern of great complexity. One of its most salient features is the presence of basins or low-probability “pockets” where $< P >_{\infty}$ is significantly smaller than $1/2$. These pockets appear for $\Delta > 0$ and can be clearly appreciated in Fig. 5, which shows a blow-up of the density phase diagram for the adiabatic case with relaxed initial conditions. Inside one of the “pockets”, the phonons conspire to push the electron towards the other site, the one with lower site energy. On the contrary, for the adiabatic case with undisplaced phonon initial conditions, no such pockets have been found and, in fact, $< P >_{\infty}$ is always greater or equal to $1/2$.

Figure 5 also shows a blow-up of the probability density phase diagram for the DNLS case. We observe, near the DNLS critical line separating localized from non-localized states, a low-probability pocket, whose origin is uncertain at this stage.

The onset of the secondary resonances ($n \neq 0$) tends to occur for ever increasing values of $G$ as $\omega$ decreases and, for fixed $\omega$, as $n$ increases. The beginning of fully consolidated channels can be used to define the position of a critical coupling value $G_c = G_c(\omega, \Delta/\omega)$. Above this value, the average probability scales as $< P >_{\infty} = f(\Delta/\omega)$ where $f$ is a smooth periodic function ranging from $1/2$ to 1. We believe this is the small polaron region for the general case. A finite-time average $< P >_{\infty}$ of order unity in this region would confirm this conjecture. This would imply a DNLS-type behavior for $< P >_{\infty}$. It remains to be determined whether the $G_c$ line approaches the DNLS selftrapping line (Fig. 1). If true, DNLS could be consider to appropriately describe the selftrapping dynamics of the quantum case, at least qualitatively, when finite-time average is considered. We are currently endeavoring to explain and model the resonance channels observed. They have also been seen for the case of a single vibrational impurity in a finite linear chain[15].

One of the authors (C.A.B) acknowledges partial support from a FONDECYT doctoral grant (project 2980033).
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Captions List

**Fig. 1 :** DNLS dimer: probability density phase diagram of $< P >_T$ in asymmetry–nonlinearity space ($T = 300 V^{-1}$). The transition line begins at $\chi = 3.09$, $\Delta = -0.383$.

**Fig. 2 :** Semiclassical dimer: Time-averaged probability at the initially occupied site $< P >_T$ (vertical) versus nonlinearity parameter $\alpha^2/k$ (horizontal) for different oscillator masses. Left column: Relaxed oscillator initial conditions. Right column: Undisplaced oscillator initial conditions ($T = 200 V^{-1}$, $k = 1$).

**Fig. 3 :** Symmetric quantum dimer: The transition region from free electron to small polaron behavior is depicted by the shadowed area in (c). In (a) and (b) we show representative $< P >_T$ plots in the adiabatic and antiadiabatic region, respectively ($T = 500 V^{-1}$, $V \equiv 1$). The thick (thin) line corresponds to initially relaxed (undisplaced) oscillators.

**Fig. 4 :** General quantum dimer: Probability density phase diagrams for $< P >_\infty$ in asymmetry–“nonlinearity” space. Upper row: Adiabatic case ($\omega = 0.5$). Lower row: Antiadiabatic case ($\omega = 10$). The left (right) column refers to relaxed (undisplaced) initial oscillators condition. ($V \equiv 1$).

**Fig. 5 :** Blow-up of the probability density phase diagrams for the semiclassical nonlinear dimer (upper) and the general adiabatic quantum dimer (lower, $\omega = 0.5$).
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