International Journal of Modern Physics C
Vol. 26, No. 12 (2015) 1550133 (9 pages)

DOI: 10.1142/S0129183115501338

Stability of uniform electronic wavepackets in chains and fullerenes

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Received 28 January 2015
Accepted 13 March 2015
Published 4 May 2015

In this paper, we investigate the influence of electron-lattice interaction on the stability of uniform electronic wavepackets on chains as well as on several types of fullerenes. We will use an effective nonlinear Schrödinger equation to mimic the electron–phonon coupling in these topologies. By numerically solving the nonlinear dynamic equation for an initially uniform electronic wavepacket, we show that the critical nonlinear coupling above which it becomes unstable continuously decreases with the chain size. On the other hand, the critical nonlinear strength saturates on a finite value in large fullerene buckyballs. We also provide analytical arguments to support these findings based on a modulational instability analysis.

Keywords: Nonlinearity; localization; self-focusing; self-trapped.

PACS Nos.: 62.30.+d; 62.20.dq; 72.15.Rn.

1. Introduction

The direct connection between the time-dependent behavior of electronic wavepackets and the electrical properties of materials has impelled the development of new studies devoted to the one-electron dynamics within the context of condensed matter physics.[1–3] Since the works by Anderson and co-workers, it is known that the presence of disorder is a key factor governing the extension of the wave function.[3] It was demonstrated that all states in a disordered system with dimension below two are localized in a small fraction of the lattice, even for a small disorder degree. We emphasize that, although the Anderson localization has been developed in the electronic context, such prediction is still valid for every field described by a wave equation. For instance, Anderson localization of electromagnetic fields,[4] water waves[5] and Bose–Einstein Condensates (BEC)[6] has been reported in the literature. One interesting issue concerning the BEC issue is that its dynamic is well described

by the Gross–Pitaevskii equation[7] and the nonlinearity present in this equation reveals exciting new physical properties.[8–10] In general, even within the electronic context, nonlinearity also can be present. It was shown that the interaction between electrons and optical phonons is well described by a nonlinear Schrödinger equation.[10,11] One of the most interesting phenomenon promoted by the nonlinearity is the self-trapping (ST) which occurs when the nonlinearity strength exceeds a critical value of order of the band width.[11–17] In this regime, an initially localized wavepacket does not spread over the system, remaining localized around its initial position. Usually, the electron–phonon interaction included in the nonlinear Schrödinger equation commonly found in the literature describes the coupling between the lattice vibration and the diagonal electronic matrix elements of the electron Hamiltonian, namely, diagonal linearity. In fact, it was shown that the lattice vibration also can couple with the off-diagonal electronic matrix elements.[18] In Ref. 19 the effect of off-diagonal nonlinearity on the electronic time-evolution was investigated. The authors analyzed the second momentum of the electronic probability and the participation number for different nonlinearity strengths. In general, it was demonstrated that the off-diagonal nonlinearity also provides the trapping of the wavepacket in a finite fraction of the system.

The competition between topology and non-linearity has attracted a great interest in the last years. In Ref. 20, a detailed study of the ST transition in square and honeycomb lattices were reported showing that the ST threshold continuously grow as a function of the initial wavepacket width. By using a tight-binding Hamiltonian approach, the dynamics of one-electron wavepackets in a twisted ladder geometry with adiabatic electron–phonon interaction was investigated.[21] The considered model mimics the electronic wavepacket dynamics in DNA-like segments. In the presence of electron–phonon interaction, the Anderson localization existent in DNA segments is suppressed and a delocalized sub-diffusive regime takes place. A partially self-trapped behavior develops at strong nonlinearities.[21] Self-trapping at large nonlinearities was also reported to take place in carbon nanotubes with strong electron–phonon coupling.[22] More recently, it has been evidenced that the relaxation process of the nonlinearity has a profound impact in the wavepacket dynamics and in the formation of self-trapped stationary states in C_{60} buckyballs.[23]

In this paper, we advance in the study of the wavepacket electron dynamics in the presence of an effective nonlinearity by numerically investigating the time-evolution of an initially uniform electronic wavepacket on chains as well as on several fullerene topologies. In our model, we include the effect of electron-lattice interaction through an effective nonlinear Schrödinger equation. We will solve numerically the dynamic nonlinear equation for an uniform wavepacket and investigate its stability. Our numerical calculations reveal the existence of a strong dependence of the critical nonlinear coupling above which the uniform state is unstable in linear chain, contrasting with its weak size dependence on fullerene buckyballs. We will also provide some analytical arguments supporting these findings based on the modulational instability analysis of the uniform state.

2. Model and Numerical Calculation

Within the context of electronic dynamics under the influence of lattice vibrations, the effective nonlinear Schrödinger equation appears as an interesting tool that allows a close investigation of the nonlinear electron wavepacket dynamics on distinct topologies. By considering an adiabatic approximation, the nonlinear term can be considered as the on-site potential depending on the local electron density. Using a tight-binding approach and considering localized orbital basis, the dynamics of the electronic wavepacket can be described following the discrete nonlinear Schrödinger equation written as

$$i\dot{\psi}_n(t) = \varepsilon_n\psi_n(t) + \sum_m V_{nm}\psi_m(t) + \chi|\psi_n(t)|^2\psi_n(t), \tag{1}$$

where we used $\hbar = 1$. $\psi_n(t)$ is the wavevector coefficient at the local orbital at site $n : |\Psi(t)\rangle = \sum_n \psi_n(t)|n\rangle$. The sum is taken over the first nearest neighbors. V_{nm} represents the energy hopping and ε_n is the on-site energy at site n. The dynamic equations will be solved by using an eighth-order Runge–Kutta algorithm and the wave function norm ($N(t) = \sum_n |\psi_n|^2$) is accompanied with an accuracy of $|1 - N(t)| < 10^{-8}$ to ensure the numerical accuracy.

To describe the spatial extent of the electron wavepacket, we calculate its participation function defined as

$$P(t) = \left[\sum_{n=1}^{N} |\psi_n(t)|^4\right]^{-1}, \tag{2}$$

$P(t)$ provides a measure of the fraction of sites over which the wavepacket is spread at time t. $P(t)$ becomes equal to N for a wavepacket evenly distributed over the entire system, while $P = 1$ for a state located in a single site.

3. Results

In this paper, we consider the wavepacket initially uniformly distributed over the entire lattice. Therefore, the initial wavepacket coefficients are $\phi_n = 1/N$ where N is the number of sites. Our focus consists in investigating the stability of the uniform wavepacket profile and its relation with the degree of nonlinearity. We initially study the wavepacket dynamics in a discrete chain with periodic boundary conditions. In Fig. 1(a), we plot the normalized participation number $P(t)/N$ versus time t for $N = 50$ and $\chi = 0.3$ and 0.6. We observe that, for $\chi = 0.3$, the participation number is constant for all times considered, thus indicating that the uniform wavepacket remains stable. For $\chi = 0.6$ the participation number exhibits an oscillating profile, which signals the instability of the wavepacket width. Such instability is related to the mechanism of polaron formation in the underlying electron–phonon system. Therefore, our calculations indicate that there is a critical value of the nonlinearity (χ_c) below which the uniform wavepacket remains stable and spread over the entire chain. For $\chi > \chi_c$, a dynamic instability leads to an oscillating behavior of the

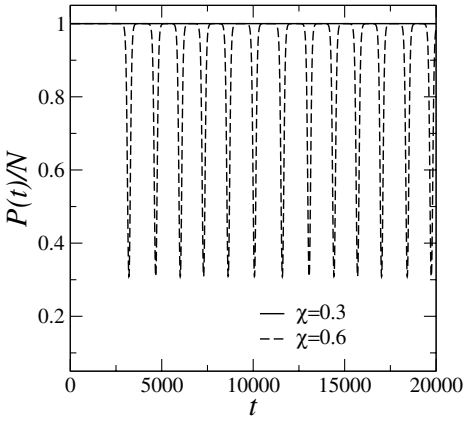

Fig. 1. The normalized participation number $P(t)/N$ versus time t for linear chains with $N = 50$ and $\chi = 0.3$ and 0.6. In our calculations, we considered the wavepacket initially distributed over the entire chain. Therefore, the initial wavepacket has $\phi_n = 1/N$ where N is the number of sites. We observe that for $\chi = 0.3$ the participation number is constant for all times considered, thus indicating that the wavepacket is stable. For $\chi = 0.6$ the participation number exhibits an oscillating profile, signaling the instability of the uniform wavepacket.

participation function of the electronic wavepacket, which is associated with periodic breathings. However, the participation function oscillates around a relatively large value, indicating that the wavepacket remains extended, at least slightly above the transition. We consider also two chains with distinct sizes and exactly the same value of the nonlinear coupling (see Fig. 2(a)). For $N = 40$, $\chi = 0.45$ is weak enough to keep the wavepacket stability and the participation number remains constant. However, for $N = 50$, the same nonlinear strength $\chi = 0.45$ in strong enough to trigger the wavepacket instability and periodic breathings emerge. As a result, the critical nonlinearity χ_c decreases as the chain size increases. The size dependence of the critical nonlinear strength was numerically determined, with the resulting curve reported in Fig. 2(b). We found numerically that the critical nonlinearity decreases proportional to $1/N$. In Fig. 2(b), the circles represent the values of χ_c obtained numerically for different sizes of linear chains. The dotted curve represents a best fitting to a $1/N$ power-law.

We now extend our analysis of the wavepacket stability for the case of lattice topologies on which the sites are distributed over a closed surface. In particular, we consider the lattice topology of fullerene buckyballs. We initially consider the wavepacket uniformly distributed over all fullerene sites and we follow numerically its time-evolution. We pay particular attention to the characteristics of the transition from stable to unstable uniform wavepackets and its dependence on the buckyball size. In Fig. 3, we show a schematic representation of two typical fullerenes: namely $C60$ and $C180$. However, our study was extended to the whole buckyball family ranging from $N = 20$ up to $N = 720$. The initial uniform wavepacket still has coefficients $\phi_n = 1/N$ where N is the number of sites in a given

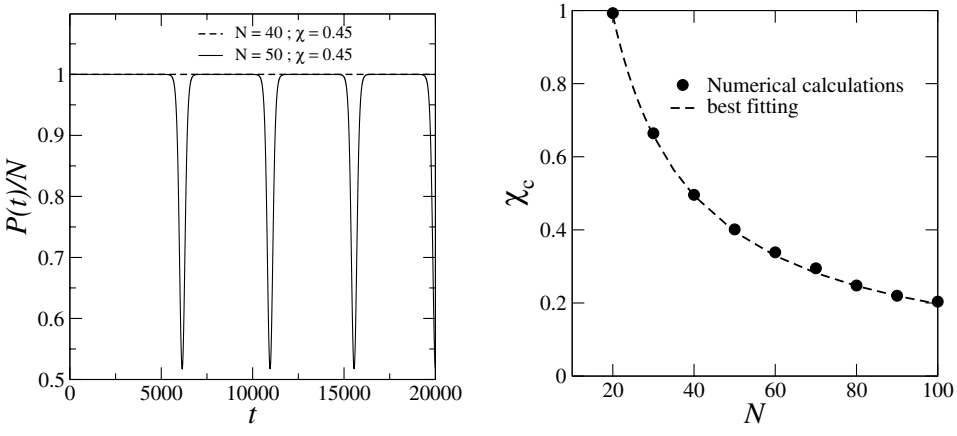

Fig. 2. (a) Normalized participation number $P(t)/N$ versus time t for linear chains with distinct sizes and degree of nonlinearity ($\chi = 0.45$). We observe that for $N = 40$, such nonlinear coupling is sufficient to keep the wavepacket stability. However, for $N = 50$, the uniform state becomes unstable and the participation number develops oscillations, signaling the wavepacket breathing. (b) We numerically demonstrated that the value χ_c that separates the stable phase from the unstable phase decreases as $1/N$. Error bars are smaller than the symbol sizes.

fullerene. In Fig. 4, we plot the scaled participation function $P(t)/N$ versus time t for $\chi = 7$ and 7.5. We have used the $C60$ topology for these calculations. We can observe that there are two distinct regimes depending on the degree of nonlinearity. One of these regimes is characterized by the stability of the wavepacket occupying the entire buckyball. Consequently, the scaled participation function remains constant for all times. In the other regime, the wavepacket focuses around a small fraction of the buckyball sites, a phenomenon similar to the well-known wavepacket self-focusing. For the case of $C60$, we can see through the evolution of the normalized participation function P/N that the transition occurs for a typical value of the nonlinearity

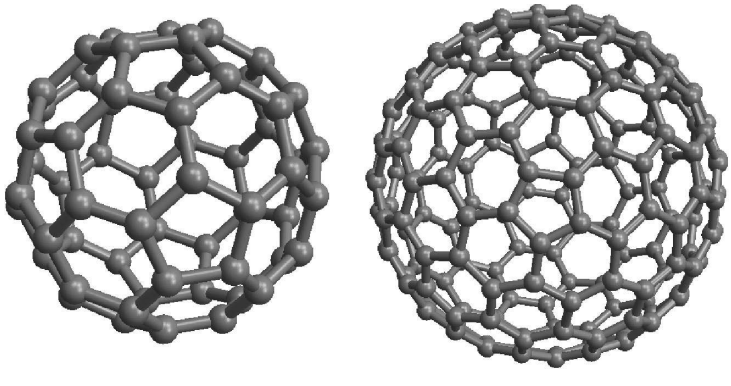

Fig. 3. Schematic representation of two fullerene buckyballs, namely $C60$ and $C180$.

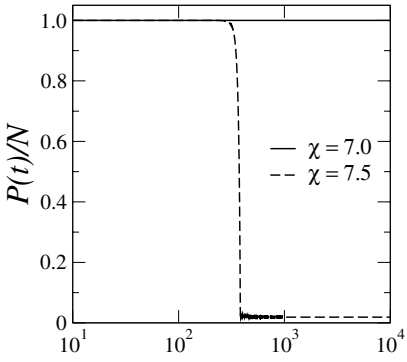

Fig. 4. Scaled participation function $P(t)/N$ versus time t for $\chi = 7$ and 7.5. We have considered here the wavepacket uniformly distributed over a $C60$ geometry. We can observe that for $\chi = 7$ the wavepacket remains stable occupying the entire buckyball. For $\chi = 7.5$, we observe that the wavepacket localizes over a small fraction of sites, signaling a self-focusing phenomena.

around $\chi_c = 7.32$. Further, no oscillations are observed above the transition. Instead of developing breathings, as it occurs in a linear chain, there is a direct transition from a stable uniform extend state to a strongly localized state. We numerically determined the critical nonlinear strength governing this transition for the entire buckyball family. We found that the critical value χ_c necessary to promote the transition slightly decreases as the number of sites of the fullerene is increased, as reported in Fig. 5. However, it saturates at a finite value for very large buckyballs, in contrast with the reported continuous decrease of the critical nonlinearity with the size of linear chains. It is important to stress that the above features characterizes the transition from stable to unstable initially uniform wavepackets. The influence of the nonlinearity on the stability of initially localized or extended nonuniform states is strongly dependent on the specific initial wavepacket distribution.[20]

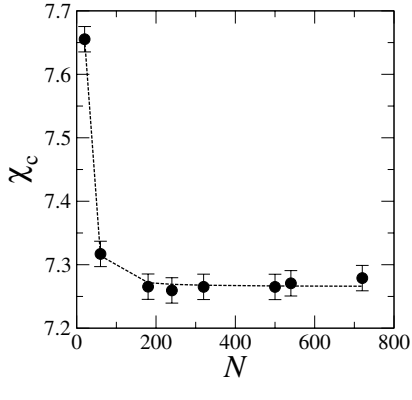

Fig. 5. The critical nonlinear strength separating the stable uniform state phase and the self-focusing phase as a function of the number of sites in fullerene buckyballs. The critical value χ_c necessary to promote the transition depicts a weak size dependence for fullerene buckyballs.

The above trends associated with the distinct size dependence in linear chains and buckyballs of the nonlinear coupling above where a uniform wavepacket becomes unstable can be understood under the light of a modulational instability analysis. As the initial state is uniform, the discrete nature of the lattice is not relevant to describe the general aspects related to its stability. Within this context, one may perform the modulational instability analysis in the continuous version of the nonlinear Schrödinger equation, which may be put in the form

$$i\frac{d\psi}{dt} = \nabla^2\psi + \chi|\psi|^2\psi \tag{3}$$

from which a linear diagonal term was dropped as it does not influence the wavefunction dynamics. The above continuous nonlinear Schrödinger equation has as CW solution $\psi(t) = \psi_0 e^{-i\chi|\psi_0|^2 t}$. In order to investigate its stability, one adds a small perturbation to its amplitude as

$$\psi(\mathbf{r},t) = [\psi_0 + \varepsilon(\mathbf{r},t)]e^{-i\chi|\psi_0|^2 t}. \tag{4}$$

The time evolution of the perturbation $\varepsilon(\mathbf{r},t)$ obeys

$$i\frac{d\varepsilon}{dt} = \nabla^2\varepsilon + \chi|\psi_0|^2(\varepsilon + \varepsilon^*), \tag{5}$$

where ε^* stands for the complex conjugate of ε and nonlinear terms are disregarded. The above equation has harmonic solutions in the form

$$\varepsilon(\mathbf{r},t) = Ae^{i(\mathbf{k}\cdot\mathbf{r}+\Omega t)} + Be^{-i(\mathbf{k}\cdot\mathbf{r}+\Omega t)}, \tag{6}$$

subjected to the dispersion relation

$$\Omega = \sqrt{k^2(k^2 - 2\chi|\psi_0|^2)}. \tag{7}$$

According to the above dispersion relation, harmonic perturbations with large wavevectors remain stable (real Ω), while those with $k < k_{MI} = \sqrt{2\chi|\psi_0|^2}$ grow exponentially (imaginary Ω). As a consequence, the CW solution will be unstable whenever $k_{MI} = \sqrt{2\chi/N} > 2\pi/L$, where L is the typical linear dimension of the system and $2\pi/L$ is the minimum allowed wavevector for an harmonic perturbation. This last expression ultimately determines a characteristic nonlinear strength $\chi_{MI} = 2\pi^2 N/L^2$ above which the CW solution is unstable. In linear chains $N \propto L$, leading to $\chi_{MI} \propto 1/N$. In large chain sizes, the critical modulational instability threshold is quite small. Although the uniform wavepacket becomes unstable above χ_{MI}, the nonlinear coupling is not strong enough to promote the wavepacket localization and breathing pattern develops. According to the dispersion relation (Eq. 7) the period of the breathing oscillations scales as $\tau \propto (\chi - \chi_{MI})^{-1/2}$ above the modulational instability threshold. On the other hand, $N \propto L^2$ in a buckyball geometry. In this case, one results with a size-independent characteristic nonlinear strength for the modulational instability of the CW solution. This critical value is large enough to drive

the wavepacket toward its strong localization. These analytical results are in full agreement with the numerical data.

4. Summary and Conclusions

In summary, we study the instability of an electronic wavepacket evolving in discrete chains and fullerene buckyballs. In the case of the discrete linear chain, we found that the value of χ_c separating the phase of stable from unstable wavepackets profile decays proportional to $1/N$, with the wavepacket developing breathings right above the instability threshold. In fullerenes, there is also a χ_c below which the electronic wavepacket remains uniformly distributed. For values of nonlinearity above χ_c, the electronic wavepacket focuses around a small fraction of the buckyball sites. While in linear chains this critical nonlinear coupling decreases monotonically with the chain size, in fullerenes χ_c displays a very weak size dependence. We demonstrated that the distinct reported wavepacket dynamics in linear chains and buckyballs can be understood within the framework of the modulational instability phenomenon. The present study can be extended to the analysis of the stability of uniform wavepackets in other nanoscopic structures (nanotubes, nanostripes, etc) of current scientific and technological interest, as well as to the stability of soliton-like solutions. Within this context, the relaxation time of the nonlinearity shall be a relevant ingredient to be included in the stability analysis.[23] These are topics that deserve to be investigated in the near future in order to provide a more complete scenario regarding the electronic wavepacket dynamics in nanostructures.

Acknowledgments

This work was partially supported by CNPq, CAPES (PVE A121), and FINEP (Federal Brazilian Agencies), CNPq-Rede Nanobioestruturas, as well as FAPEAL (Alagoas State Agency).

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