



Optical absorption and delocalization in a quaternary tight-binding chain with correlated disorder



L.D. da Silva^a, A. Ranciaro Neto^{a,b}, M.O. Sales^a, M.L. Lyra^a, F.A.B.F. de Moura^{a,*}

^a Instituto de Física, Universidade Federal de Alagoas, Maceió AL 57072-970, Brazil

^b Faculdade de Economia, Administração e Contabilidade, Universidade Federal de Alagoas, Maceió AL 57072-970, Brazil

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ABSTRACT

We study electronic transport in a one-dimensional model with four kinds of atoms. The quaternary energy distribution is chosen in such a way that it contains long-range correlations. Localization properties and optical absorption are obtained by using numerical methods. Regarding the localization properties of the wave function, we show numerically that, for weak correlation on disorder, the participation number is finite, and the eigenstates are localized. For strong correlation, our results suggest that the model displays fully delocalized eigenstates. Weakly localized states are present in the regime of intermediate correlations. Our analysis of the optical absorption showed an interesting pattern of peaks and a strong dependence on the correlation degree. The structure of peaks is discussed in the light of the underlying density of states.

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1. Introduction

According to Anderson localization theory, it is well known that extended eigenstates are absent in low-dimensional disordered systems for any level of uncorrelated disorder [1]. Exceptions to this statement are related to those cases with intrinsic correlation in the disorder distribution [2–21]. In general, it has been demonstrated that correlated disorder promotes the appearance of a metal–insulator transition, even in one-dimensional models. Moreover, the effect of correlated disorder on optical spectroscopy properties is also a key problem in the context of condensed matter physics [22–27]. It is well known that optical spectroscopy usually fails in detecting localization–delocalization transitions. However, a double-peak absorption spectrum profile has been numerically reported in a 1d lattice with long-range correlated diagonal disorder [24]. This phenomenon was also observed in a 1d lattice with long-range off-diagonal correlated disorder [25]. In [26], the optical properties in 1d models with heavy-tailed Levy disorder distribution were studied. These authors found a broadening of the optical line and a non-universal scaling of the distribution of exciton localization lengths. The optical absorption bandwidth and the non universality of the localization length within models with Levy disorder distribution were re-visited in [27].

Works on correlated disorder have considered systems in which the site energies are uniformly distributed in a finite range $[-W/2, W/2]$. Some years ago, models were considered in which the on-site energy could assume only discrete values (for example, binary and ternary models) [28–34]. The Anderson model with long-range correlated ternary disorder sequence was studied in [28]. These authors demonstrated that if the ternary sequence is generated in a fully random way, then the system is an insulator. Nevertheless, by creating a ternary diagonal disorder with long-range correlations, a localized–delocalized phase transition was observed [28]. The effect of long-range correlations in the sequence of

* Corresponding author.

E-mail address: fidelis@fis.ufal.br (F.A.B.F. de Moura).

capacitances of classical transmission lines (TL) was studied by Lazo and Diez [30,31]. To generate the ternary correlated distribution of capacitances, the Fourier filtering method was used [30] and also the Ornstein–Uhlenbeck (OU) process [31]. In both cases, a transition from non-conducting to conducting state of the TL induced by strong correlations was observed. More recently, a one-dimensional classical ternary harmonic chain with the mass distribution constructed from an OU process was studied [32]. The localization aspects of all vibrational eigenstates were obtained using the transfer matrix formalism. These authors concluded that only the zero frequency mode can propagate through the chain, thus contradicting previous works [28–31]. It has been latter clarified that the transition observed in Refs. [28–31] was actually a transition between disordered and ordered chains because the global disorder effectively vanished as the internal correlations were increased in their model systems. Forthcoming works unveiled the role played by local and global disorder in several models with discrete diagonal disorder [32–34], showing that a localization–delocalization transition can indeed occur when the global disorder is kept finite while the local disorder vanishes. In this case, delocalized states emerge when the two point correlation function develops a slow power-law decay. In the present work, we will focus in a model system with a finite global disorder and explore the signature of the localization–delocalization transition in the optical absorption spectrum.

We would like to stress that the study of models with discrete correlated disorder has received considerable interest from the experimental point of view. The possibility of generating real systems with a discrete correlated disorder can be a possible tool to compare theoretical and experimental procedures [35], also allowing for the design of new materials with adjustable properties.

Here, we will study the electronic transport in systems with correlated quaternary disorder distribution. In particular, we will examine localization properties and optical absorption using numerical methods. The term “quaternary disorder” represents a disorder distribution containing only four values ($\epsilon_1, \epsilon_2, \epsilon_3, \epsilon_4$). We will be particularly interested in identifying the distinct regimes of eigenstates localization and relate them with the typical regimes local disorder that emerge when the correlation degree is increased. Further, we will explore the optical absorption spectrum and relate its structure of peaks with the underlying characteristics of the energy band.

2. Model and formalism

We consider a standard one-electron Anderson Hamiltonian written as

$$H = \sum_n \epsilon_n |n\rangle \langle n| + \sum_n \tau_n (|n\rangle \langle n+1| + c.c) \quad (1)$$

where (ϵ_n) represents the quaternary correlated disorder distribution. We construct this sequence by mapping a continuous correlated series $\{V_n\}$ in a discrete group of four values ($\epsilon_1, \epsilon_2, \epsilon_3, \epsilon_4$). The continuous series $\{V_n\}$ is obtained through the trace of a fractional Brownian motion defined as [4–7]:

$$V_n = \sum_{k=1}^{N/2} \frac{1}{k^{\gamma/2}} \cos\left(\frac{2\pi nk}{N} + \phi_k\right). \quad (2)$$

Here, ϕ_k represent random phases distributed within the interval $[0, 2\pi]$.

The above series has no typical length scale, which is a characteristic of several stochastically generated natural series [36]. These sequences have a power-law spectral density of the form $S(k) \propto 1/k^\gamma$ derived from the Fourier transform of the two-point correlation function. k is related to the wavelength λ of the undulations on the random landscape. The widespread occurrence in nature of sequences with power-law noise is related to the general tendency of large driven dynamical systems to evolve for a self-organized critical state [37]. For $\gamma = 0$, the sequence is completely uncorrelated. The exponent γ is directly related to the Hurst exponent H of the rescaled range analysis ($\gamma = 2H + 1$) which describes the self-similar character of the series and the persistent character of its increments. In the case of $\gamma = 2$ the sequence resembles the trace of the usual Brownian motion. Long-range correlated sequences with persistent (anti-persistent) increments are obtained in the regime $\gamma > 2$ ($0 < \gamma < 2$).

According to the approach used in [4], we also perform a normalization process such that: $\langle V_n \rangle = 0$ and $\sqrt{\langle V_n^2 \rangle - \langle V_n \rangle^2} = 1$. Once the correlated sequence $\{V_n\}$ was built, we proceed to the mapping in order to generate the quaternary correlated sequence $\{\epsilon_n\}$. The mapping is defined by the equation below:

$$\epsilon_n = \begin{cases} \epsilon_1 & \text{if } V_n < -b \\ \epsilon_2 & \text{if } -b < V_n < 0 \\ \epsilon_3 & \text{if } 0 < V_n < b \\ \epsilon_4 & \text{if } V_n > b. \end{cases} \quad (3)$$

The mapping parameter b introduced into this equation controls the type of mapping we use. In general, b controls the probability of each of the four values ($\epsilon_1, \epsilon_2, \epsilon_3, \epsilon_4$) appearing on the quaternary distribution. We choose $b \leq 2$, once 2 is approximately the largest value of the V_n sequence after the normalization process. Moreover, we set the following values for the on-site energies: $\epsilon_1 = -2, \epsilon_2 = -1, \epsilon_3 = 1$ and $\epsilon_4 = 2$.

In our calculations, hopping energy is $\tau_n = 1$. By exact diagonalization we compute all eigenstates $|\Phi(E)\rangle = \sum_n f_n(E) |n\rangle$, where $f_n(E)$ represents the Wannier amplitude on site n . From these eigenfunctions, we estimate participation average

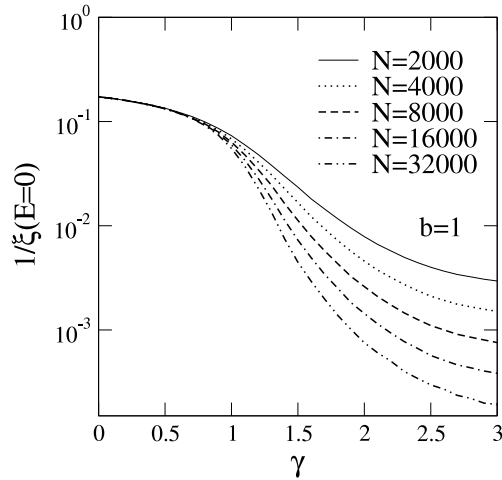


Fig. 1. Inverse of the participation number $\xi(E = 0)^{-1}$ versus the correlation parameter γ considering $N = 2000, N = 40\,000, N = 8000, N = 16\,000, N = 32\,000$ and $b = 1$.

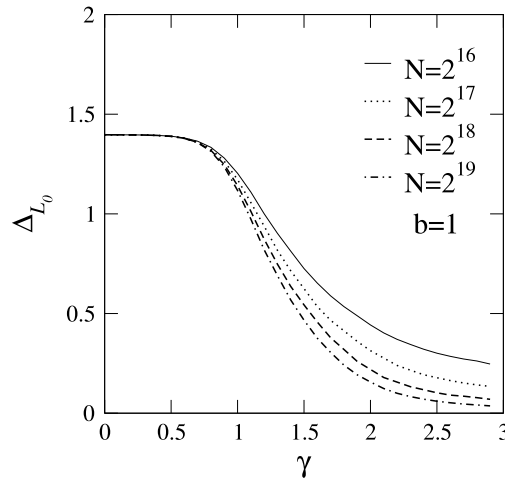


Fig. 2. Local standard deviation Δ_{L_0} versus the degree of correlation γ for $b = 1$ and $N = 2^{16}$ until $N = 2^{19}$.

number $\xi(E)$ [5,6,1,38] and we analyze the extended/localized nature of the eigenfunctions. The participation is defined as:

$$\xi(E) = \frac{\sum_{n=1}^N (f_n(E))^2}{\sum_{n=1}^N (f_n(E))^4}. \tag{4}$$

Participation is a measure of the number of sites in which contains the wave function. We study the inverse of the participation number $\xi(E = 0)^{-1}$ at the center of the band ($E = 0$). Furthermore, we investigate the absorption spectrum defined as [38,39,25,33]:

$$A(E) = \frac{1}{N} \sum_{\beta} \delta(E - E_{\beta}) F_{\beta}, \tag{5}$$

where F_{β} is the oscillator strength associated with the eigenvalue β , namely $F_{\beta} = [\sum_n f_n(E_{\beta})]^2$. For the off-diagonal terms, positive ($\tau_n > 0$) and the diagonal disorder following an uncorrelated distribution, the eigenstates with the largest oscillator strength are those of the top of the band.

3. Results and discussion

We start by presenting some results obtained performing the exact diagonalization of the quantum Hamiltonian. In Fig. 1, we report the inverse of the participation number, calculated in the vicinity of the band center ($E \rightarrow 0$) ($\xi(E = 0)^{-1}$) versus

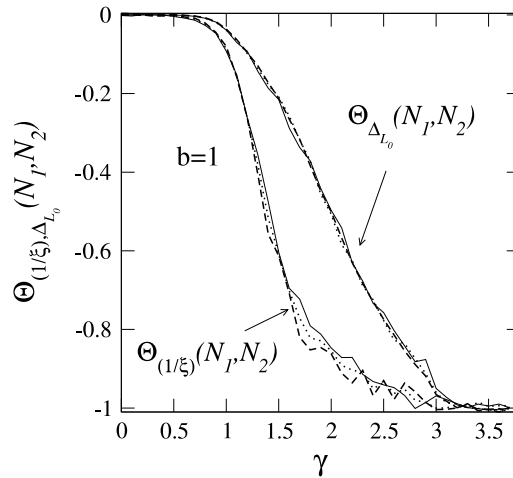


Fig. 3. $\Theta(N_1, N_2)$ function computed for the participation number and the local disorder.

γ for a quaternary chain with $b = 1.0$ and distinct chain sizes $N = 2000, N = 4000, N = 8000, N = 16\,000$ and $N = 32\,000$. We observe that when the correlation parameter $\gamma < 1$, $\xi(E = 0)^{-1}$ remains independent of N . This result suggests that all eigenstates for $\gamma < 1$ are strongly localized. As we increase the value of γ , $\xi(E = 0)^{-1}$ starts to decay when the system size is increased. We also observed that the decay of $\xi(E = 0)^{-1}$ with γ becomes more pronounced for $\gamma > 2$.

To investigate in detail some intrinsic characteristics of the quaternary disorder distribution, we address the specificity of the local disorder distribution and its relation with N and γ . In the following, we present an analysis of the amount of intrinsic disorder in such quaternary distribution. As a quantifier of the degree of disorder, we compute the local standard deviation Δ_{L_0} of the on-site energies of a segment with L_0 atoms. The local standard deviation Δ_{L_0} is defined by

$$\Delta_{L_0} = \left(\sum_{k=1}^M \Delta_{k,L_0} \right) / M, \tag{6}$$

where Δ_{k,L_0} is given by

$$\Delta_{k,L_0} = \sqrt{ \sum_{n=(k-1)L_0+1}^{n=kL_0} (\epsilon_n^2) / L_0 - \left(\sum_{n=(k-1)L_0+1}^{n=kL_0} \epsilon_n / L_0 \right)^2 }, \tag{7}$$

and $M = N/L_0$. Δ_{L_0} is a measure of the intensity of local disorder in a segment with L_0 atoms. In Fig. 2, the local standard deviation Δ_{L_0} is shown versus the degree of correlation γ for $b = 0.5, 1.0, 1.5$, $L_0 = 5000$ and system sizes ranging from $N = 2^{16}$ up to 2^{19} . We can clearly observe that, for small values of γ (roughly smaller than unit), the local disorder remains constant. For $\gamma > 1$, it also exhibits a decreasing pattern as the system size is increased. In fact, it is a consequence of the Fractional Brownian motion (Eq. (2)) that was used to generate the quaternary disorder distribution [4]. Therefore, the increase of the localization length seems to be directly related to the decrease of the degree of intrinsic disorder. To further explore such relationship, we compute the dependence of Δ_{L_0} with the chain size and compare with the results for the size scaling of the participation number (Fig. 1). We apply a standard finite-size scaling analysis given by the following function:

$$\Theta(N_1, N_2) = \frac{\log(Z(N_2)/Z(N_1))}{\log(N_2/N_1)}. \tag{8}$$

The $\Theta(N_1, N_2)$ function represents an interesting way to obtain the size dependence of a generic quantity $Z(N)$. Formally, calculations of the $\Theta(N_1, N_2)$ function for several pairs (N_1, N_2) provide the power-law exponent of the quantity Z with the system size (i.e. $Z(N) \propto N^{\Theta(N_1, N_2)}$). We apply this analysis to both cases, i.e., by considering $Z \rightarrow \Delta_{L_0}$ and $Z \rightarrow \xi(E = 0)^{-1}$. The main results are reported in Fig. 3. We used the data of Fig. 1 to compute $\Theta_{1/\xi(E=0)}(N_1, N_2)$ and the results of Fig. 2 to obtain $\Theta_{\Delta_{L_0}}(N_1, N_2)$. We notice that the decay of Δ_{L_0} and $1/\xi(E = 0)$ with the system size follows quite distinct trends, especially in the regime of intermediate correlation parameter γ . For $\gamma < 1$, a size independent degree of disorder implies in fully localized states. In the opposite regime of large correlation parameter $\gamma > 3$, both quantities decay as $1/N$. In this regime, disorder is effectively absent and, consequently, the states are fully delocalized. However, for $1 < \gamma < 3$ these two quantities depict distinct dependences on the system size. There are two intermediate regimes. In the first intermediate with $1 < \gamma < 2$, the disorder strength decreases with an exponent smaller than $1/2$. Such slowly decaying disorder strength leads to weakly localized states which have localization lengths depicting a sub-linear growth with the system size. In the

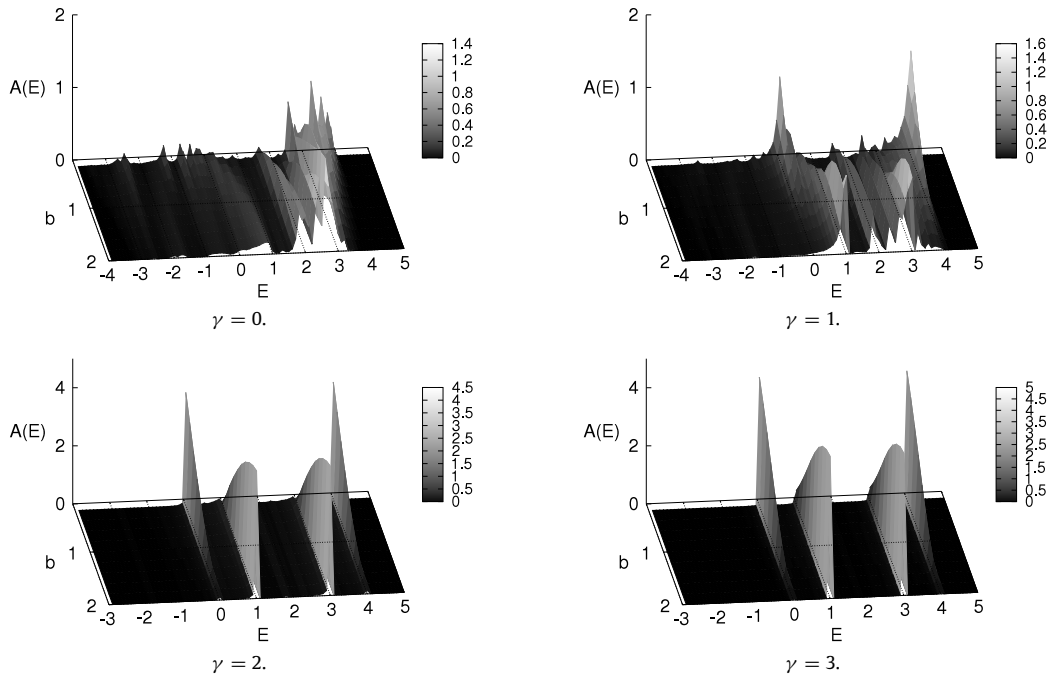


Fig. 4. Absorption spectrum versus energy versus disorder parameter b , for $N = 2000$ and b varying from 0.2 until 3.0. Case (a) refers to uncorrelated distribution ($\gamma = 0$). We observed a wide absorption peak around ($E \approx 2$). Figure (b) show the results for ($\gamma = 1$). Absorption spectrum depicts a wide energy range with small absorption. However, some absorption peaks arises within this region. These peaks are not well defined. Figure (c) for $\gamma = 2$ and (d) for $\gamma = 3$: The absorption spectrum exhibits an interesting structure of peaks strongly related to the superposition of sub-bands (see text).

second regime with $2 < \gamma < 3$ the disorder strength decays with a finite-size scaling exponent larger than $1/2$. In this case of quickly decaying disorder, the states already become fully delocalized. Deviations from the zero exponent at $\gamma = 1$ and unitary exponent at $\gamma = 2$ (ξ) and $\gamma = 3$ (Δ) are due to logarithmic corrections to scaling which are present at the border of distinct scaling regimes.

In order to analyze the optical absorption properties of the present model system, we compute numerically Eq. (5). We diagonalize the model Hamiltonian for $N = 2000$, $\gamma = 0, 1, 2$ and 3. In the following analysis, we plot the optical absorption $A(E)$ as a function of energy E and the parameter b . We observe initially the case $\gamma = 0$ (see Fig. 4(a)). In the absence of correlation, the absorption is practically null in a large part of the energy band. However, around the top of the energy band ($E \approx 2$), there is a broad absorption peak. This phenomenon is expected in the literature for systems with uncorrelated disorder and positive hopping. The states at the top of the band exhibit larger oscillator strength and, thus, effectively contribute to the optical absorption. For $\gamma = 1$ (see Fig. 4(b)), we observe that absorption is small in a wide energy range. However, again around the band edge ($E \approx 2$), there is an increase of the absorption region. We note that in the case of $\gamma = 1$, the width of the energy region with more pronounced absorption is considerably wider than in the case of $\gamma = 0$. In some works [39,25], this expansion of the absorption bandwidth was also observed for intermediate correlations. In general, a weak degree of correlation promotes the decreasing of the localization length of those eigenstates around the band edge, thus promoting the expansion of the absorption band. In Fig. 4(c) we show some results of the optical absorption for $\gamma = 2$. We observe that the absorption has a structure of peaks within the region of $E \geq 0$. For $\gamma = 3$ (see Fig. 4(d)), the optical absorption results are similar to those obtained for $\gamma = 2$. However, the structure of peaks is more pronounced.

The reported structure of peaks deserves a more detailed analysis in order to predict the position of each peak. Let us begin by discussing what happens to the quaternary disorder when the degree of correlation is increased ($\gamma \gg 0$). Based on the analysis presented in Fig. 2, the intrinsic local disorder within this quaternary sequence decreases as the correlation degree grows. In this way, the quaternary sequence evolves into a more ordered structure containing finite segments with energies $(\epsilon_1, \epsilon_2, \epsilon_3, \epsilon_4)$. Therefore, we can approximately consider that, for large γ , such quaternary distribution can be pointed out as a concatenation of four one-dimensional systems, each of energies $\epsilon_1, \epsilon_2, \epsilon_3$, and ϵ_4 , respectively. Thus, for large γ , the energy band of our system consists of an overlapping of four one-dimensional bands: $[\epsilon_1 - 2, \epsilon_1 + 2]$, $[\epsilon_2 - 2, \epsilon_2 + 2]$, $[\epsilon_3 - 2, \epsilon_3 + 2]$, $[\epsilon_4 - 2, \epsilon_4 + 2]$. Therefore, assuming that this system has positive hopping, the states with non-zero absorption should be around the top of each sub-band, i.e. $\epsilon_1 + 2, \epsilon_2 + 2, \epsilon_3 + 2, \epsilon_4 + 2$. Using the values of $(\epsilon_1, \epsilon_2, \epsilon_3, \epsilon_4)$, this mapping $(2, -1, 1, 2)$ reproduces accurately the position of the absorption peaks $(0, 1, 3, 4)$.

4. Summary and conclusions

We have studied electronic transport in quaternary systems with correlated disorder. Our goal was to understand the localization properties of the electronic wave function and optical absorption properties. In general, we considered a one-dimensional model with four kinds of atoms. The on-site diagonal energy at each of these atoms was given by $(\epsilon_1, \epsilon_2, \epsilon_3, \epsilon_4)$. The distribution of these energies was chosen in order to exhibit long-range correlations Eq. (2) (i.e. fractional Brownian motion with power spectrum $S(k) \propto k^{-\gamma}$). By using exact diagonalization, we studied the localization properties and the optical absorption spectrum of all eigenstates. Regarding the localization properties of the wave function, we have shown numerically that, for $\gamma \leq 1$, the participation number does not depend on the system size (fully localized states). For $1 < \gamma < 2$ the states are weakly localized depicting a sub-linear dependence on the system size [34]. For $\gamma > 2$, our results points to fully delocalized states. We unveiled that the local disorder have a wider regime of sub-linear size dependence. Our analysis of the optical absorption showed an interesting pattern that was highly dependent on the degree of correlation. In the absence of correlation $\gamma = 0$, the optical absorption in mostly of the band is zero. However, around the top of the band ($E \approx 2$), there is a region of non-zero absorption. In general, the same behavior was obtained for different energy values with weak correlations $\gamma \leq 2$. For $\gamma > 2$, our results indicate the presence of several peaks. Using a heuristic formalism, we explained qualitatively the existence of each peak as resulting from the superposition of the energy bands of disorder free segments.

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