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Localization of electronic states in finite ladder models: Participation ratio and localization length as measures of the wave-function extension

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In this work we discuss and compare different definitions for localization of electronic states in low-dimensional systems. We choose a heuristic model for DNA-like molecules as a system prototype in order to establish ranges of equivalence for the localization length obtained from both the conductance and participation ratios. The results suggest also criteria to infer the extension of wave function in mesoscopic systems within the diffusive transport regime as complementary information to the localization length.

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I. INTRODUCTION

The recent interest on the electronic applications of DNA molecules has already motivated an astonishing amount of literature,[1] although a consensus on the transport properties of such a complex system has not yet been established. From the theoretical point of view, such complexity has to be handled having in mind different length scales:[2] first-principles calculations for short base-pair (bp) sequences and heuristic models for sequences in the long macromolecular range. The disclosure of the electronic structure of short DNA chains in very strict conditions begins to be addressed by combining powerful experimental tools such as scanning tunneling microscopy and first-principles calculations for few tens of base pairs.[3] On the other hand, one of the frameworks to investigate the electronic properties on a mesoscopic scale is the use of tight-binding ladder models with effective sites for the nucleotides.[4] Experimental results may be simulated by such simple models, such as the semiconductorlike current-voltage characteristics[5] or interband optical transitions.[6] Having in mind the possible nanoelectronic applications of DNA, transport properties have been the focus of a wide span of investigative approaches, since such a fascinating system with intrinsic (due to the huge number of degrees of freedom of macromolecules) and extrinsic (due to the environmental aspects, presence of substrate, solvent, counterions, and electric contacts) complexities may show competing transport mechanisms. Although a comprehensive scenario is out of the scope of this paper, an overview may be gained in recent reviews:[7] static and dynamic fluctuations as well as environmental effects have to be considered. Here again two strategies in addressing the charge-transfer problem can be identified. We may find either examples of heuristic models for polarons in DNA (Refs. 8 and 9) or first-principles combined to empirical approaches as a methodology for investigating the charge transfer.[10]

One of the several aspects involved in the analysis of the DNA electronic and transport properties is the localization of states, here from the point of view of heuristic ladder models: possible localization-delocalization transitions have been alluded as a consequence of long-range correlations along the DNA-like chains.[11] One previous work[12] has called the attention to the localization length enhancement due to the base pairing, when comparing a single strand to the corresponding double-strand structure. This localization length enhancement, leading to a wave-function extension comparable to the sample length, has been called effective delocalization. The expression "effective delocalization" has been sparsely used in different contexts, from charge spreading out through a polymer chain[13] to transport in finite quantum Hall systems.[14] In this last situation, an effective delocalization is defined in the exact sense we are dealing with in the present paper: *in a finite sample the states with a localization length larger than the sample size L are effectively delocalized.*[14]

The use of effective delocalization in the present paper is motivated by the fact that the quest for DNA-based electronics has to be set for mesoscopic and not macroscopic dimensions. Therefore, the system size becomes an important length scale in the problem, and if we recall the transport regimes in mesoscopic systems, the localization length, l_c, and the system size, L, are directly compared within two regimes:[15] the localized regime with $L \gg l_c$ and the diffusive regime where $L \ll l_c$. Finite DNA-like double strands seem to offer an intermediate stage to these two regimes, which is related, as will be seen, to wave-function extensions that are much longer than l_c, suggesting a less restrictive definition of effective delocalization, namely, in the range $L \approx l_c$. Another previous result suggests a bona fide delocalization for the same ladder model, but for unrealistic interchain coupling, therefore, of prospective academic interest as a toy model.[12] The properties of this model, with stronger interchain than intrachain coupling, are indeed in the diffusive regime, showing a significant conductance even at very long length scales, although not truly delocalized in the thermodynamic limit, as pointed out by other groups.[16–18] Nevertheless, a comprehensive scenario for the possible resonant effects and intrachain correlations, even for effective DNA models, is not fulfilled,[19] and analyzing transport properties of effective DNA models is also relevant from a biological point of view, such as studying DNA repairing deficiencies.[20] However, one should keep in mind the aforementioned complexity of the problem and how transmission resonance effects may be conciliated with other charge-transfer mechanisms, for instance, polaron hopping, or even combined polaron-bipolaron-tunneling pathways,[21] continuous an open question.

On the other hand, a central discussion is related to the consistency among distinct measures of localization, such as

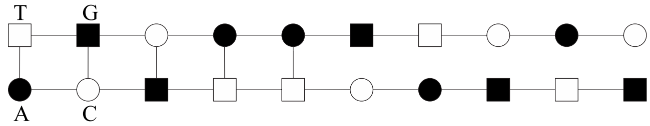

FIG. 1. The model DNA system with the four effective nucleotides: A, T, C, and G. At the left side in a double chain with base pairing and at the right side separated in single strands. Throughout this work we will consider the limiting case of a completely random sequencing along the chains.

the Thouless number,[22] spectral statistics,[23] inverse participation ratio (PR) (Ref. 24) (and the related participation ratio), Lyapunov exponents, localization length,[25] or the geometric average of the density of states.[26,27] Here we focus on the comparison between two definitions of localization: the PR (Ref. 28) together with the derived participation number (PN) and l_c obtained from the conductance[25] in the situation depicted above, namely, for $L \approx l_c$, i.e., an effective delocalization regime. This comparison leads to similar localization lengths for both methods, whenever $l_c \ll L$, while for $L \approx l_c$ the PN and l_c from conductance strongly fluctuate and deviate from each other. Actually such fluctuations and deviations may start already for system lengths an order of magnitude longer than l_c obtained in the thermodynamic limit. These results suggest that for finite systems, the extension of the wave function may be as important as the localization length[17] in order to characterize the electronic properties. It should be also mentioned that the question posed here—effects of localization lengths comparable to the system lengths on transport properties in a mesoscopic length scale—is closely related to a growing branch in measuring localization, namely, probing localization in open systems.[29,30] Therefore, comparing closed and open systems localization measures, such as PR and l_c, starts also to become relevant.

In what follows we briefly describe our model Hamiltonian for a double chain simulating a double-strand DNA, as well as the approach necessary for obtaining the conductance. The localization definitions, PR and l_c, used throughout the paper are also introduced. Careful comparisons between both methods are undertaken afterwards and used in the qualitative characterization of a wave-function extension, L_{wf}.

II. NUMERICAL MODEL

The starting point for analyzing the localization of electronic states in a heuristic DNA-like system is to consider an effective double-strand model, consisting of two parallel chains N_L sites long that can be connected at both ends to semi-infinite ideal nondisordered leads. The base paring is introduced by means of four different effective sites, representing the nucleotides, A, T, C, and G with specific binding properties: only A-T and C-G pairs are possible (see Fig. 1). This base paring correlation is the only one to be taken into account in the present work since the nucleotide sequencing is completely random. The DNA-like double strands are, therefore, described by a two-channel tight-binding model with nearest-neighbor interactions only. In this approximation, the suitable Hamiltonian can be written as

$$H = \sum_{i=1}^{N_L} \sum_{j=1}^{2} \left(\varepsilon_{i,j} |i,j\rangle\langle i,j| + V|i,j\rangle\langle i+1,j| + V|i,j\rangle\langle i-1,j| \right.$$
$$\left. + V'|i,j\rangle\langle i,j+1| + V'|i,j\rangle\langle i,j-1| \right). \quad (1)$$

According to values suggested in the literature, we use the following site energies: $\varepsilon_A = 8.24$ eV, $\varepsilon_T = 9.14$ eV, $\varepsilon_C = 8.87$ eV, and $\varepsilon_C = 7.75$ eV.[31] The intrachain hopping parameters, between two adjacent nucleotides and for interchain coupling, are $V = 1.0$ eV and $V' = 0.5$ eV,[12] respectively. This set of parameters has been used in several ladder models for DNA (Refs. 16–19 and 32–34) and represents a qualitatively sound parametrization obeying the expected condition of $V' < V$. Other parameters set (within the same model) may simulate more properly the semiconductorlike electronic structure,[5,6,35] but we considered the present set as adequate for the focus of the present paper: the localization of the states in ladder models and the possible comparison to previous works on the subject.[4] One should recall, however, that the parameters set used here is an empirical choice and that the problem of reducing the full DNA Hamiltonian to a single (effective) orbital tight-binding model is still debatable.[36]

In order to obtain the localization length from the conductance we will focus on the transmission probabilities, which are directly related to the total Green's function of the system calculated by means of the recursive approach for a lattice of sites treated in a tight-binding approximation.[37,38] A detailed description of this procedure for the transmission probabilities in terms of the Green's function is already given in textbooks.[39]

At zero-temperature limit, the linear conductance is related to the transmission probability by the Landauer formula[40]

$$G = \frac{2e^2}{h} \mathrm{Tr}(\hat{t}\hat{t}^\dagger), \quad (2)$$

where $\hat{t}$ is the 2×2 transmission matrix of the system. Our results are given in terms of the dimensionless conductance $G_0 = 2e^2/h$, being therefore equivalent to simply the trace of the transmission matrix $\mathrm{Tr}(\hat{t}\hat{t}^\dagger)$. The localization length for the DNA-like double-strand model is computed from the exponential decrease in the conductance and, based on the result by Johnston and Kunz,[25] defined by the relation

$$l_c^{-1}(E) = -\lim_{N_L \to \infty} \frac{1}{2N_L} \langle \ln \mathrm{Tr}(\hat{t}\hat{t}^\dagger) \rangle, \quad (3)$$

where $\langle \cdots \rangle$ means an average over several hundred different double-chain configurations. This is done to avoid spurious effects due to a particular configuration.

Another way established in the literature to determine the degree of localization of a state, directly from diagonalizing the Hamiltonian, is the PR,[22,24] defined, in a tight-binding approximation, by

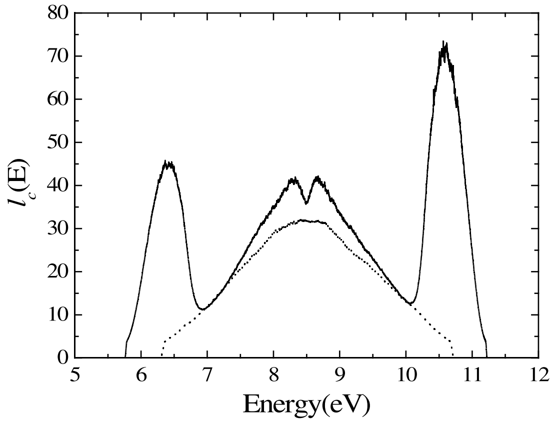

FIG. 2. Localization length as a function of energy for single (dotted line) and double-strand (continuous line) DNA.

$$\mathrm{PR} = \frac{1}{2N_L \sum_{i=1}^{N_L} \sum_{j=1}^{2} |a_i|^4}, \tag{4}$$

where $a_{i,j}$ is the wave-function amplitude in the (i,j) site. In the limit $N_L \to \infty$, the PR$\to 0$ for localized states. Considering finite systems, i.e., rigid boundary conditions, truly delocalized states PR reaches the maximum value of 2/3 for one-dimensional ordered case.[28] A quantity related to the PR is PN=PR$\times 2N_L$,[41] considering a double strand as depicted in Fig. 1. While PR is a measure of a fraction, PN would be a measure of the actual number of sites having appreciable wave-function amplitudes at a given energy.

III. RESULTS AND DISCUSSION: LOCALIZATION LENGTH AND WAVE-FUNCTION EXTENSION

The localization length as a function of energy, calculated within the scheme delineated by Eq. (3), is depicted in Fig. 2 for two illustrative situations. The continuous line shows the double ladder case emulating a double-strand DNA, while the dotted line corresponds to the results for a single-strand DNA-like chain. The interesting point here is the significant enhancement of the localization length in the energy window near the edges of the single-strand density of states. The effect of connecting both chains and respecting the base pairing is to enhance the localization length up to an order of magnitude. Such enhancement has been reported in a previous work by means of a PR calculation[12] and similar findings have been published afterwards.[17] The present calculations are for finite systems 1250 bps long. Localization lengths of the same order are obtained for fishbone and double strands with backbone models treated also within a tight-binding framework of effective sites.[4] The highest value of $l_c \approx 70$ bps would validate that the limit $N_L \to \infty$ has been effectively reached since $l_c / N_L \ll 1$. It should be mentioned that double strands without base pairing (not shown here) also present higher l_c than a single strand, but still significantly lower than in the cases including base pairing.

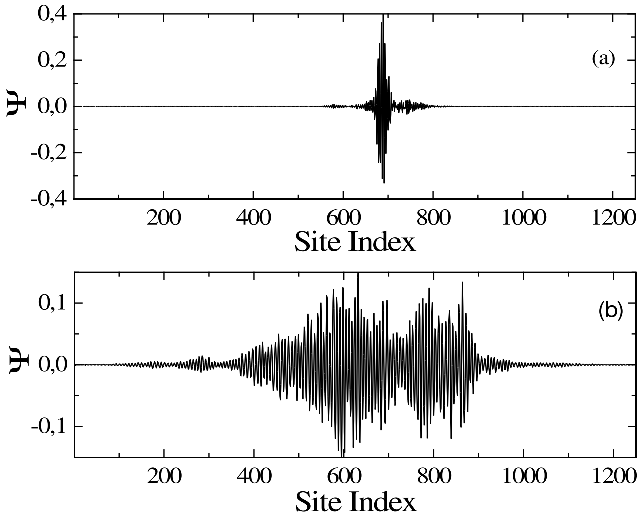

FIG. 3. Wave function for states of the double-strand DNA-like chain with different localization length. (a) A state with energy $E \approx 10$ eV and $l_c \approx 15$ bps and (b) a state with energy $E \approx 10.5$ eV and $l_c \approx 70$ bps.

A localization length $l_c \approx 70$ bps in a 1250-bp-long chain would also suggest at a first glance that the corresponding wave function would not extend significantly throughout the ladder, but one should remind that in general l_c is not a measure of L_{wf}.[42] For very large systems this rather subtle conceptual difference has no further consequences. However, for smaller systems the difference between the l_c and L_{wf} could be important, and indeed they may differ by an order of magnitude as discussed below.

In Fig. 3 two wave functions of the double strand 1250 bps long are depicted. In Fig. 3(a) a state with $l_c \approx 15$ bps is clearly confined to a small portion of the system, while in Fig. 3(b), a state with $l_c \approx 70$ bps spreads out over almost the entire double chain. Also interesting is the fact that these states show strong modulations, consistent with the presence of resonances along the system.[19] In other words, an exponential envelope characterized by a localization length may not grasp all the important aspects of the problem.

The degree of the wave-function localization depends on the length of a finite system, whenever it is in the diffusive regime up to the crossover to a localized regime. The two quantities, PN and l_c, are analyzed as functions of chain length at the energy $E=10.6$ eV [Figs. 4(a) and 4(b), respectively]. Both quantities initially increase for short systems, as expected for a diffusive regime or what we call as effectively delocalized. More important are the strong fluctuations shown by l_c, up to a chain length of $N_L \approx 1000$ bps. For longer chains, the localization length remains constant, $l_c \approx 70$ bps. Hence, $N_L \approx 1000$ is the lower limit for a well-defined l_c from Eq. (3) applied to the present system. Approximately at the same length, on one hand, the PN begins to saturate, Fig. 4(a), as generally expected for $L > l_c$.[43] On the other hand, fluctuations in PN become more important, in spite of the numerical average over many disorder configurations. Indeed, fluctuations in PR have also been discussed as a possible order parameter for localization-delocalization transitions.[44] In summary, these are the aspects we focus on: the lengths at which (i) the saturation of the PN and (ii) the quenching of the fluctuations of l_c occur.[45]

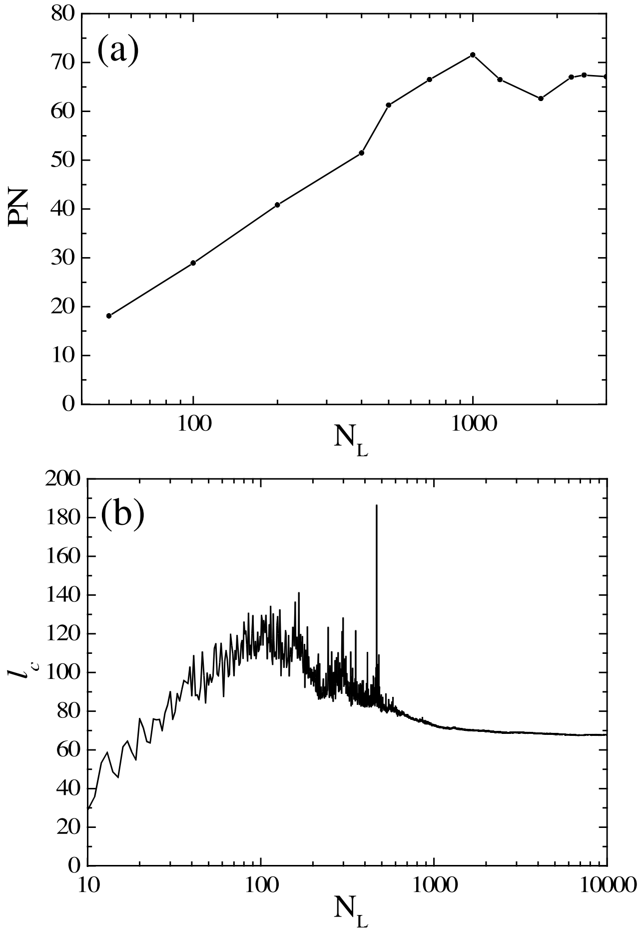

FIG. 4. The participation number, (a), and localization length, (b), at $E=10.6$ eV for the double-strand DNA-like model as a function of chain length in units of base pairs.

A measure of L_{wf} seems, therefore, to be possible by comparing the quenching of the fluctuations in l_c, together with the clear saturation of the PN around 70 bps. Indeed, simple inspection of the wave-function extension in Fig. 3(b) delivers an estimative comparable to the common threshold at $N_L \approx 1000$ for the result trends in Fig. 4.

The qualitative measurement procedure for L_{wf} suggested by the common threshold for both quantities (quenching of the fluctuations of the localization length and saturation of the PN as function of chain length) may be further refined by a direct comparison of both l_c and PN quantities. In Fig. 5 the PN and l_c are compared for the entire energy spectrum already depicted in Fig. 2. Figure 5(a) is for a 500-bp-long double chain, while Fig. 5(b) is for a 2000-bp-long system.

This direct comparison delivers a precise evaluation of L_{wf} in the transition from localized to diffusive regimes, where the localization length is comparable to the system size. It is interesting to notice that l_c represents an almost exact envelope for PN for systems longer than L_{wf}. Naively one should expect that PN could be closer to a measure of L_{wf} than to l_c. It should be mentioned that the PN shows a stronger fluctuation than the values of l_c since an appropriate average over several hundred configurations in the case of PN are numerically too costly. In the limit of long enough systems, PN and l_c show these similarities in values in spite of being calculated in completely different ways. While the PN is obtained from the diagonalization of a finite system, l_c is obtained from the transmission probability in an open system, the scattering region embedded by semi-infinite leads. The deviations between both quantities occur only for chains that are shorter than L_{wf}. Indeed, in Fig. 5(a) we see that for smaller localization length (around $E \approx 7.0$ eV and $E \approx 10.0$ eV) the deviations are also smaller since in this limit $N_L \gg l_c$ is better satisfied. Therefore we can envisage a further characterization of the wave-function localization, beyond simply defining l_c, by means of comparing two definitions of the degree of localization: PN and l_c obtained from the transmission probability. Such characterization should be valid in both localized and diffusive regimes. It is obviously meaningless in a truly delocalized system for which l_c diverges and the PN has an upper bound of the finite chain length taken for the calculation.

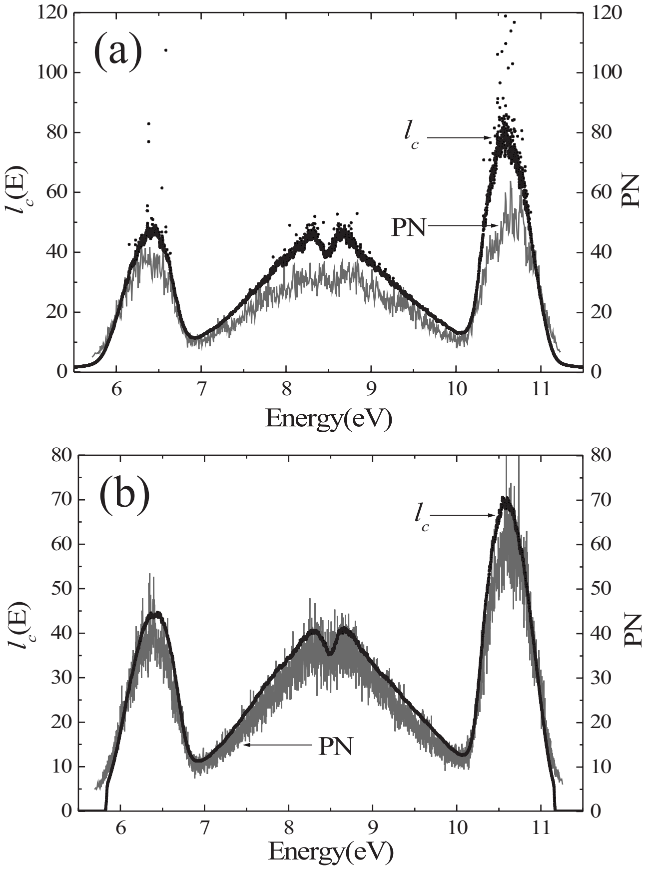

FIG. 5. A comparison between localization length and PN through the entire energy spectrum for DNA-like double chains of different lengths: (a) 500 bps and (b) 2000 bps.

A complementary insight to the problem may be obtained by directly inspecting the conductance as a function of the length, instead of a derived quantity as l_c. In Fig. 6 we depict the conductance (averaged over several disorder configurations for each length) at the energy associated to the highest PN. In Fig. 6(a), for the DNA-like double chain, this energy is $E=10.6$ eV, as already discussed. In Fig. 6(b) we illustrate the behavior for a "fake" DNA, namely, a double chain with similar on-site energies [$\varepsilon_A=10.5$ eV, $\varepsilon_T=9.5$ eV, $\varepsilon_C=10.3$ eV, and $\varepsilon_G=9.7$ eV (Ref. 12)] but with a stronger

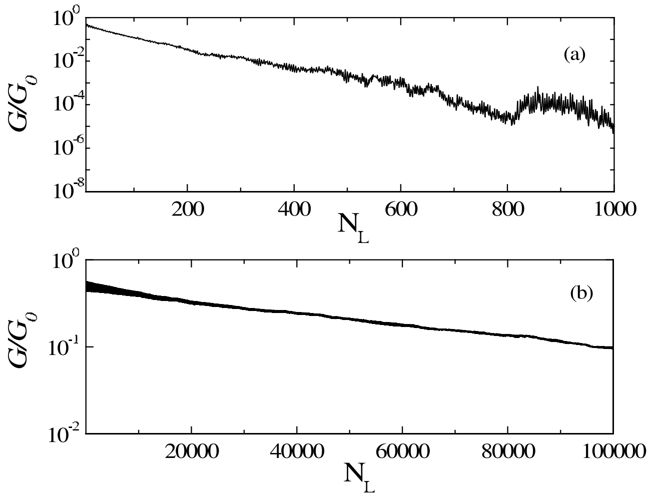

FIG. 6. Conductance at a selected energy as a function of double-chain length in units of base pairs. (a) The DNA-like system and (b) for interchain stronger than intrachain hopping, $V'/V=3/2$.

interchain hopping: $V'/V=3/2$. In this case, the highest PN occurs around $E=14.7$ eV. For both situations only one transmission channel is connected to the leads used here, and therefore, the upper bound for the conductance is $G=G_0$.

For the DNA-like double chain in Fig. 6(a) a fast decrease in the conductance with the system length can be observed, while the fluctuation amplitude becomes comparable to the conductance for lengths beyond a few hundred base pairs. Besides rapid fluctuations associated to resonant effects that are revealed at low conductances,[19] important deviations from a decrease described by a simple and smooth exponential envelope start to show up. It should be noticed, however, that the conductance still shows appreciable values up to several hundred base-pair-long chains.

On the other hand, the fake DNA in Fig. 6(b) is somehow intriguing. The decrease in the conductance is very slow with increasing length and remains smooth through the entire range depicted here. It is noticeable that no significant fluctuations in the conductance appear in the whole range of the figure, meaning that the wave function continuously extends up to the maximum length depicted. Our simulations reach double chains as long as 10^5 bps, which may be considered already within a macroscopic length scale. Having in mind a measure of the base pairs spacing of 3.4 Å, 10^5 bps represent 34 μm. In fact, these results reveal not only such large L_{wf} but also a $l_c \approx 10^5$ bps, a value at which a tendency to saturation only starts to show up. Such systems will show a smooth PR with values close to the maximum (0.667 in one dimension)[28] for very long chains, as pointed out previously.[12,16]

IV. CONCLUSIONS

The question of how electronic states are localized in double chains with a base-pairing correlation, emulating effective DNA-like systems, has motivated a wide range of investigations, by far not completely covered in the references mentioned in the above introduction. The main question posed here, concerning an effective delocalization in systems eligible for molecular electronic wiring, focuses on a rather long length scale, suitable for tight-binding effective models. Nevertheless, resonance transmission could not have the highest impact on the transport properties as a whole, albeit the interesting findings based on this view is concerning point mutation effects in DNA.[20] Hole polaron formation and hopping play an important role and lead to smaller length scales, sometimes of the order of solely ten base pairs,[8] nevertheless comparable to l_c for the ladder models used here, both being smaller than the proposed wave-function extension here.

In such an involved context, together with the importance of the transport properties on a mesoscopic scale, emerges the necessity of measuring the wave-function extension, L_{wf}, besides solely the localization length, l_c. In the present work two well-known definitions of wave-function localization are compared: the participation number and the localization length from the conductance. The main findings are: (i) both quantities deliver almost coincident values for $L \gg l_c$, and (ii) the system size for which deviation between both quantities start to be appreciable is proposed to be a measure of L_{wf}. This measure coincides with the system length at which, on one hand, the fluctuations in l_c are suppressed, and on the other hand, the participation number begins to saturate. These findings are in the context of a dual scenario, namely, addressing on one hand probing localization in open systems[29,30] compared to closed ones and, on the other hand, establishing comparisons among different localization definitions.[46] It is worth mentioning that $L_{\mathrm{wf}} \gg l_c$ for some of the electronic states discussed here and, therefore, called as effectively delocalized, while some toy models, like the one illustrated in Fig. 6(b), may lead the measure of L_{wf} to a macroscopic length scale. These findings point to the necessity of a comparative study of the various definitions of localization originally thought for defining delocalization-localization transitions in the thermodynamic limit. As soon as transport properties in a mesoscopic scale are concerned, these usual definitions might be misleading and a careful insight into the extension of the wave function is necessary.

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[45] A further aspect is the comparison of a localization measure previously used for closed systems, PN, with the conductance obtained from open systems. Besides that, the interesting behavior of l_c at lower lengths should be addressed in future works but is not within the scope of the present one.
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