Localization Phenomena in a DNA Double Helix Structure: A Twisted Ladder Model

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In this work we propose a model for DNA double helix within the tight-binding framework that incorporates the helicity of the molecules. We have studied localization properties of three DNA sequences, the periodic poly(dG)-poly(dC) and poly(dA)-poly(dT) sequences and the random ATGC sequence, all of which are coupled to backbone with random site energies representing the environmental fluctuations. We observe that due to helicity of DNA, electron transport is greatly enhanced and there exists almost a disorder-strength independent critical value of the hopping integral, that accounts for helicity of DNA, for which the electronic states become maximally extended. We have also investigated the effect of backbone energetics on the transmission and $I - V$ characteristics of DNA.

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I. INTRODUCTION

In recent years interest on DNA mediated charge migration has enhanced remarkably because its potential for the development of new generation DNA-based nano-electronic devices and computers. Also in biology, a precise understanding of the mechanism of electron transport along DNA could play an important role for the description of the processes like damage sensing, protein binding, gene regulation, cell division, etc. However, the question that does DNA conduct electron is quite intriguing to the physicists as well as biologists, and, even today the electronic and transport properties of DNA are not well understood. Inspite of the current intense debate, the electron transport properties of DNA were being addressed soon after the discovery of the double helix structure of DNA by Watson and Crick [1]. Eley and Spivey [2] first suggested that DNA could behave like an electric conductor. More recently with the advent of measurements on single DNA molecule, Kelley et al. [3] showed that DNA behaves like a conducting molecular wire. But a number of conflicting experimental results [4–11] and a variety of theoretical models [12–22] appeared in the literature. Experiments on periodic double stranded poly(dG)-poly(dC) DNA sequence reported both conducting [4] as well as semiconducting [5] behavior. Measurements on aperiodic λ-phage DNA sequence suggested that it could be metallic [4], superconducting [23] at low temperature or insulating [6]. Unambiguous and reproducible experimental results are still a great technological challenge due to the complexity of environment, thermal vibrations, contact resistance and sequence variability of the DNA molecules. On the other hand, lack of clear understanding of charge transfer mechanism in DNA leads to various phenomenological models in which charge transport is mediated by polarons [24], solitons [13], electrons or holes [25–27].

The electronic properties and the conducting behavior of DNA thus remain highly controversial and Ref. [28] provides an excellent review on this issue.

In order to explain the diverse experimental results, one should consider three different contributions that are present in an experiment with DNA, namely, the sequence of the pairs of nucleotides (i.e., base-pairs) in DNA, the presence of backbones and the influence of environment. The environment in turn pieces out into three main parts, the substrate on which experiments are performed, the surrounding temperature [29] and the humidity. As the sugar-phosphate backbones are negatively charged and hanging outside the double-helix structure of DNA, they can easily interact with the substrate and the effective on-site energies of the backbone get modulated to a great extent [7–9, 23, 30, 31]. The experiments with DNA are performed mostly in two different conditions: i) in natural aqueous buffer solutions, and ii) in dry conditions. In DNA’s natural aqueous buffer solution, the backbone phosphates usually attract counter-ions and polar water molecules to neutralize the phosphates, and in the process modify their ionization potential [32]. Even for the dry case in vacuum, there are few counter-ions or water molecules that may still reside on the phosphates of the backbones even after the drying process and hence can change the effective on-site energy of the backbone sites. Though the main conducting pathway is believed to be the $\pi - \pi$ interaction of the stacked base-pairs in double stranded DNA and the backbones do not take part directly in the electronic transport process of DNA, one can significantly control the transport behavior of DNA just by tuning the environment which effectively modifies the backbone on-site energies introducing disorder. We have shown that this kind of control can ignite a semiconducting to metal transition in the conducting behavior of DNA.

In this paper we study the electronic conduction properties of DNA double helix within the tight-binding framework where environmental fluctuations are modelled in terms of disordered on-site potentials of the backbone sites. We propose a model that explicitly takes into account the helicity of the DNA molecule. We have ob-

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served that helicity significantly enhances electronic conduction, and, the interplay of helicity and backbone disorder has nontrivial effects on the transmission characteristics of the DNA molecules.

This paper is organized as follows. In Sec. II we introduce the model Hamiltonian and briefly describe our theoretical formulation. We analyze our numerical results in Sec. III and finally conclude in Sec. IV.

II. MODEL AND THEORETICAL FORMULATION

DNA, carrier of genetic code of all living organism, is a long and complex biological macro-molecule, a π-stacked array of base-pairs made from four nucleotides guanine (G), adenine (A), cytosine (C) and thymine (T) coupled via hydrogen bond and forms a double-helix structure associated with sugar-phosphate backbone attached to each base-pair [21]. In most of the theoretical models it is assumed that electronic transport [17, 33, 35] is through the long-axis of the DNA molecule. In the present study, the helicity of the DNA molecule is incorporated in a tight-binding (TB) dangling backbone ladder model [21, 36] by adding hopping integrals due to the proximity of atoms in the upper strand with the corresponding atoms of the lower strand in the next pitch (see Fig. 1). The Hamiltonian for the twisted ladder model can be expressed as

\[
H_{DNA} = H_{ladder} + H_{helicity} + H_{backbone},
\]

where,

\[
H_{ladder} = \sum_{i=1}^{N} \sum_{j=I,II} \left( \epsilon_{ij} c_{ij}^{\dagger} c_{ij} + t_{ij} c_{ij}^{\dagger} c_{i+1,j} + \text{H.c.} \right) + \sum_{i=1}^{N} v \left( \epsilon_{II}^{I} c_{II\uparrow} + \text{H.c.} \right),
\]

\[
H_{helicity} = \sum_{i=1}^{N} v' \left( \epsilon_{II}^{I} c_{II\uparrow} + \text{H.c.} \right),
\]

\[
H_{backbone} = \sum_{i=1}^{N} \sum_{j=I,II} \left( \epsilon_{q(j)}^{I} c_{iq(j)}^{\dagger} c_{iq(j)} + t_{q(j)}^{I} c_{iq(j)}^{\dagger} c_{iq(j)} + \text{H.c.} \right),
\]

where \( c_{ij}^{\dagger} \) and \( c_{ij} \) are the creation and annihilation operators for electrons in the \( i \)th Wannier state, \( t_{ij} \) is hopping integral between neighboring nucleotides along each strand of the ladder, \( \epsilon_{ij} \) is on-site potential energy of the nucleotides, \( t_{q(j)}^{I} \) is hopping amplitude between a nucleotide and the corresponding backbone site, \( \epsilon_{q(j)}^{I} \) is on-site potential energy of the backbone sites with \( q(j) = \uparrow, \downarrow \) for \( j=I \) and \( II \) respectively denoting upper and lower backbone sites, \( v \) is interstrand hopping integral between two neighboring sites of DNA within a given pitch, \( v' \) is interstrand hopping integral between neighboring atomic sites in the adjacent pitches which actually accounts for the helical structure of DNA. Here \( n \) is the number of sites in each strand within a given pitch. For simplicity we set \( \epsilon_{q(j)}^{I} = \epsilon_{b} \), \( t_{ij} = t_{i} \) and \( t_{q(j)}^{I} = t_{b} \).

It is possible to obtain an analytical expression for the dispersion relation of an infinite homogeneous DNA chain (setting \( \epsilon_{ij} = \epsilon \) and \( t_{i} = t \)) modeled by twisted ladder in the absence backbones. Using Bloch’s theorem, the dispersion relations for the highest occupied molecular orbital (HOMO)(\( E_{\text{-}} \)) and the lowest unoccupied molecular orbital (LUMO)(\( E_{\text{+}} \)) can be expressed as

\[
E_{\pm} = \epsilon + 2t \cos(k) \pm \sqrt{v^{2} + v'^{2} + 2vv' \cos(nk)},
\]

where \( v_{n} \) and \( v'_{n} \) are the interstrand hopping (\( v \)) between neighboring nucleotides within a given pitch and dashed lines represent the interstrand hopping (\( v' \)) between neighboring nucleobases of adjacent pitches. Backbones are not shown in the figure.

In order to study the transport behavior of DNA, we use semi-infinite 1D chains as leads connected cross-wise to the left (L) and right (R) ends of the DNA double helix and the Hamiltonian of the entire system is given by

\[
H = H_{DNA} + H_{L} + H_{R} + H_{\text{tun}}.
\]

The explicit form of \( H_{L}, H_{R} \) and \( H_{\text{tun}} \) are

\[
H_{L} = \sum_{i=-\infty}^{0} \left( \epsilon_{i} c_{i}^{\dagger} c_{i} + t_{i} c_{i+1}^{\dagger} c_{i} + \text{H.c.} \right).
\]
by the two-terminal Landauer formula

\[ I(V) = \frac{2e}{h} \int_{E_F-V/2}^{E_F+V/2} T(E) dE, \]

where \( V \) is the applied bias voltage, \( E_F \) is the Fermi energy, and \( T(E) \) is the transmission probability at energy \( E \). We have assumed that there is no charge accumulation within the system, i.e., the voltage drop occurs only at the boundaries of the conductor.

### III. RESULTS AND DISCUSSIONS

We first study the localization properties of the system. The localization length \( L \) of the system is calculated from the Lyapunov exponent \[ \gamma = 1/l = -\lim_{\lambda \to \infty} \frac{1}{\lambda} < \ln(T(E)) >, \]

where \( \lambda \) is the length of the system in terms of basepairs, and \(< >\) denotes average over different disorder configurations. In actual experimental situations there are various environmental fluctuations. We have simulated these environmental fluctuations in the model by considering the backbone site energy \( \epsilon_b \) to be randomly distributed within the range \([\epsilon_b-w/2, \epsilon_b+w/2]\), where \( \epsilon_b \) is the average backbone site energy and \( w \) represents the disorder strength. For the purpose of numerical investigation the on-site energies of the nucleotides are taken as the ionization potentials, and the following numerical values are used throughout this work: \( \epsilon_G = -0.56eV \), \( \epsilon_A = -0.07eV \), \( \epsilon_C = 0.56eV \), \( \epsilon_T = 0.83eV \). The intranucleotides between like nucleotides are taken as \( t = 0.35eV \) while those between unlike nucleotides are taken as \( t = 0.17eV \). We take intranucleotides hopping parameter to be \( v = 0.3eV \). As all the nucleobases are connected with sugar-phosphate backbone by identical C-N bonds, the corresponding hopping parameter between the nucleobase and backbone is taken to be equal for all the cases and we take \( t_b = 0.7eV \) \[19\]. The parameters used here are the same as those used in \[11\] which were extracted from the \textit{ab initio} calculations \[40–42\].

In Fig. 2 we have plotted inverse localization length \( \gamma \) of the periodic poly(dG)-poly(dC) and poly(dA)-poly(dT) sequences and the random ATGC sequence with respect to \( v' \) (where \( v' \) accounts for the helicity of DNA) for various values of the disorder strength \( w \). It has been observed that all the curves have a general shape for the periodic as well as random DNA sequences. The variation of \( \gamma \) with \( v' \) is not monotonic, there exists a flat minimum of \( \gamma \) for each sequence and the position of this minimum for a specific sequence is almost insensitive to the disorder strength \( w \). As we increase \( v' \) starting from zero, \( \gamma \) starts decreasing, which is quite natural because by allowing \( v' \) we are opening new channels for...
electron conduction, so the system becomes much more conductive and $\gamma$ decreases. Now there is a flat minimum which signifies that in this region the system is most conducting as both the channels, one along the main DNA ladder and one arising due to helicity, are contributing almost equally. Then $\gamma$ starts increasing implying that $v'$ channel starts to dominate over the other channels due to $t$ and $v$, and as we further increase $v'$ the system becomes more and more like a 1D disordered one (as the role of $t$ and $v$ becomes negligible). As in the 1D systems Anderson localization sets in if we introduce small role of $\Gamma$ coming more and more like a 1D disordered one (as the value of $\Gamma$ gets reduced). Now there is a flat minimum $\gamma_t$ irrespective of the disorder strength. Disorder can change the magnitude of $\gamma$ but it has nothing to do with the nature of this competition, so the minima occurs almost at the same value of $v'$ irrespective of the disorder strength.

In Fig. 3 we have plotted $\gamma$ versus $v'$ for a fixed disorder strength ($w=5$) for three DNA sequences for the two cases $\bar{\epsilon}_b = 0$ eV and $\bar{\epsilon}_b = 3$ eV showing significant drop in $\gamma$ for higher $v'$.

In Fig. 4 we have shown the behavior of $\gamma$ with disorder strength $w$ for two cases with $v' = 0$ and $v' = 0.3$. It is clearly visible that for the higher value of $v'$, there is a significant drop in $\gamma$ which is obvious because by introducing $v'$ we are adding other channels for conduction and the localization length of the system increases. Initially if the localization length for $v' = 0$ was a few base-pairs in presence of backbone disorder, it increases three to four times if we consider the effect of helicity of

![Graphs showing Lyapunov exponent $\gamma$ versus a parameter $v'$ for three DNA sequences at a fixed disorder strength ($w=5$) for two cases $\bar{\epsilon}_b = 0$ eV and $\bar{\epsilon}_b = 3$ eV. Figure shows that there is a shift in positions of the minima for $\gamma$ versus $v'$ curves depending on the value of the average backbone site-energy ($\bar{\epsilon}_b$).](image1)

![Graphs showing Lyapunov exponent $\gamma$ versus a parameter $w$ for three DNA sequences with disordered backbone sites. Figure 4. (Color online). Lyapunov exponent $\gamma$ vs $w$ for the three DNA sequences for the two cases $v'= 0$ eV and $v'= 0.3$ eV showing significant drop in $\gamma$ for higher $v'$.](image2)
the DNA molecules. There is another thing to be noted is that there exists a maximum in each of these curves, beyond which $\gamma$ starts decreasing even if we increase $w$. This feature was already reported by Guo et al. for the fishbone and dangling backbone ladder models [11] and the same effect is also present in our model, but this anomalous effect of backbone disorder is much smaller in our model which turn also signify that helical nature of DNA can minimize the effect of disorder i.e., environmental fluctuations.

![Graph](image)

**FIG. 5:** (Color online). Transmission coefficient as a function of energy for the poly(dA)-poly(dT) and poly(dG)-poly(dC) sequences for the cases $\epsilon_b = 0$ eV and $\epsilon_b = 3$ eV without backbone disorder (i.e., $w=0$) with $\nu' = 0.3$ eV.

We have also investigated the $I-V$ response of the two periodic sequences. The temperature is set to 0 K. To minimize the contact effects we choose tunnelling parameter $\tau$ to be optimum i.e., $\tau = \sqrt{\epsilon_b \times t}$ between ds-DNA and the electrodes, where $t$ is the hopping parameter for the electrodes [10]. In Fig. 6 we have shown variation of transmission probability $T$ with respect to energy $E$ for these two sequences, and it clearly shows that by tuning the backbone site energy one can control the energy gap of the system. In Fig. 6 the $I-V$ characteristics are shown without backbone disorder and it has been observed that for $\epsilon_b = 3$, the cut-off voltage becomes nearly equal to zero for both the periodic poly(dG)-poly(dC) and poly(dA)-poly(dT) sequences. For poly(dG)-poly(dC) case the response is almost linear which indicates that the system would undergo a transition from semiconducting to metallic phase depending on on-site energies of the backbone, not only disorder can induce that kind of transition [11]. $I-V$ characteristics for $\epsilon_b = 0$ case are in good agreement with experimental results [5]. We have checked the $I-V$ response for different system sizes, and observed that there is no significant change in the characteristics. In Fig. 7 we show the $I-V$ responses for the two periodic sequences for $\epsilon_b = 3$ case in presence of the backbone disorder. It clearly shows that with increasing disorder the current in the system decreases.

![Graph](image)

**FIG. 6:** (Color online). $I-V$ characteristics for the poly(dA)-poly(dT) and poly(dG)-poly(dC) sequences for the cases $\epsilon_b = 0$ eV and $\epsilon_b = 3$ eV without backbone disorder (i.e., $w=0$) with $\nu' = 0.3$ eV.

![Graph](image)

**FIG. 7:** (Color online). $I-V$ characteristics for the poly(dA)-poly(dT) and poly(dG)-poly(dC) sequences for the $\epsilon_b = 3$ eV case in presence of backbone disorder ($w$) with $\nu' = 0.3$ eV.
IV. CONCLUDING REMARKS

Within the tight-binding framework, the fishbone and the dangling backbone ladder models are generally used to study the transport properties of DNA like systems, but none of these models have incorporated the effect of helicity of DNA molecules. Helicity is a very fundamental aspect of the DNA structure and gives the possibility of new conduction channels in DNA. In this paper we propose a model that accounts for the helical structure of the DNA molecules and show that there is a significant change in the localization properties and $I - V$ response of the systems. One of the main results is that there exists a almost disorder independent critical hopping ($v_c'$) for which localization length becomes maximum for each of the sequences that we have considered. At this critical hopping strength system is least affected by the external disturbances, i.e., environmental effects are least at this point. If one can utilize this information properly then it might be possible to minimize the environmental effects in the actual experiments. We have also shown that though backbones do not play any role directly in electron conduction but they can significantly contribute by narrowing the energy gap and reducing the cut-off voltage. It might also be possible to find DNA in a complete metallic phase even in presence of environmental fluctuations as evident from the almost linear $I - V$ response of poly(dG)-poly(dC) sequence for $\bar{\epsilon}_c = 3$. We look forward that there might be experimental investigations as well as ab initio calculations in near future to find the exact value of the interstrand hopping integral ($v_c'$) between nucleobases of adjacent pitches and its actual role in transport.
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