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Electronic transport through dsDNA based junction: a Fibonacci model

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Abstract

A numerical study is presented to investigate the electronic transport properties through a synthetic DNA molecule based on a quasiperiodic arrangement of its constituent nucleotides. Using a generalized Green's function technique, the electronic conduction through the poly(GACT)-poly(CTGA) DNA molecule in a metal/DNA/metal model structure has been studied. Making use of a renormalization scheme we transform the Hamiltonian of double-stranded DNA (dsDNA) molecule to an effective Hamiltonian corresponding to a one-dimensional chain in which the effective on-site energies are arranged as a quasiperiodic lattice according to Fibonacci sequence. The room temperature current-voltage characteristic of dsDNA has been investigated in this Fibonacci model and compared with those corresponding to poly(GACT)-poly(CTGA) DNA molecule. Our results indicate the main effect of the quasiperiodic arrangement of the nucleotides as the Fibonacci sequence on the electronic spectrum structure of the dsDNA is that the energy band gaps of the molecule have a tendency for suppression. The room temperature I-V characteristic of the DNA Fibonacci model shows a linear and ohmic-like behavior.

Keywords: DNA, current-voltage characteristic, Fibonacci model, Green's function

1. Introduction

During the last years, many studies have been devoted to the understanding of the conduction properties of electrode-molecule-electrode junctions (magnetic and non-magnetic) in the framework of molecular electronics [1-7]. The single molecules with semiconducting behavior play an important role in the development of these systems for electronic applications. The discovery that deoxyribosenucleic acid (DNA) can conduct an electrical current has made it an interesting candidate for the role that in itself does not occur in nature. The potential application of DNA as an important part in molecular electronics is one of the several cases. DNA could be useful in molecular electronics for design of the electric circuits, which could help to overcome the limitations that silicon-based electronics is facing in the recent years. Several studies on the structural, electronic and conduction properties of the biological DNA molecules have been published [8-13]. Most studies have been focused on the understanding of how the details of the DNA electronic structures, the nature of electronic states, impurities and geometrical factors affect the conduction properties of DNA molecule in the nanoelectronic devices [14-18].

In biological dsDNA, several thousands to millions of base pairs are aperiodically distributed, exhibiting self-similar characteristics due to the presence of long-range correlations in certain regions [19]. The role of long-range correlations in the electronic delocalization has been anticipated [20]. On the other hand, the synthetic DNA molecules as the single and double-stranded sequences, the nature of the long-range correlations and also the self-similar behavior of the electronic wave functions in their structures have been the subject of intense debate [11, 19, 21, 22]. In general, the self-similar characteristic of the wave functions is attributed to the quasiperiodicty feature of the structures. The effects of quasiperiodicity on the electronic spectra can be studied in most simple quasiperiodic structures, Fibonacci chains which are able to support the extended electronic states as well. Accordingly, Fibonacci systems may be regarded as useful model prototype, able to mimic relevant features related to long-range correlation effects in biological DNA molecules. In this work, we consider a dsDNA molecule as poly(GACT)-poly(CTGA) sequences containing four different nucleotides, thymine (T), cytosine (C), adenine (A) and guanine (G) which may be arranged as a one-dimensional site Fibonacci lattice. By means of a renormalization process, we transform the Hamiltonian of

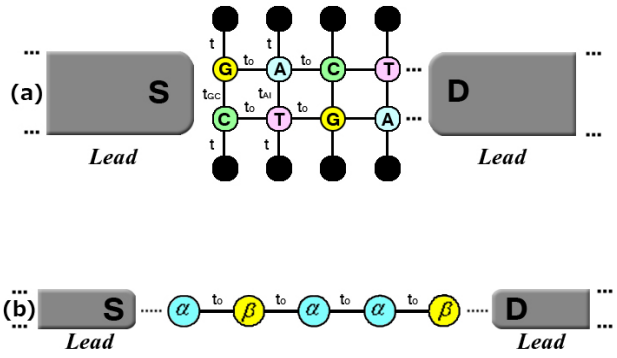

Figure 1. (color online) The plots show a schematic representation of the metal/DNA/metal structure. (a) The ladder model contains M cells connected to each other. Parameters are defined as mentioned in the text. (b) The suggested DNA Fibonacci model in which the effective on-site energies $\tilde{\varepsilon}_\alpha$ and $\tilde{\varepsilon}_\beta$ are arranged according to the Fibonacci sequence.

dsDNA molecule to an effective Hamiltonian corresponding to a one-dimensional chain in which the effective on-site energies are arranged according to a Fibonacci sequence. Here, in the framework of this site Fibonacci model, we investigate the electronic conduction and current-voltage characteristic of the DNA Fibonacci model and the results are compared with those corresponding to poly(GACT)-poly(CTGA) DNA molecule. The model and description of the computational methods are introduced in Section 2. The results and discussion are presented in Section 3, followed by conclusion in section 4.

2. Model and the computational scheme

The nature of charge transfer and the electronic transport through the DNA molecule depends on the understanding of its structural parameters. The biological DNA consists of a double-helix with an aromatic π-stack core, where four types of nucleobase participate in Watson-Crick base pairing, G:C and A:T [23]. A detailed knowledge of the dsDNA structure suggests that the π-π interaction between the stacked base pairs in the dsDNA could support extended charge transport [24,25]. The electronic coupling through a duplex stack of nucleobases is expected to involve in both intrastrand and interstrand pathways [24]. Here, we consider a ladder model to study the dsDNA electronic properties, which includes the backbone structure of the dsDNA, explicitly. As shown in figure 1(a), the dsDNA molecule in the ladder model has two central conduction channels in which G and C (A and T) sites represent a base-pair, which are interconnected and further linked to the upper and lower sites, representing the backbone. The backbone sites, however, are not interconnected along the DNA sequence [10]. In the presented model, the DNA structure as a 2-dimentional layer, consisting of M cells and further four sites in each cell, is set between two semi-infinite metallic leads (electron reservoirs) as the electrode described by an effective self-energy function and then the conductance properties of the system is numerically calculated, based on the well-known procedures particularly suitable for treating the electron transmission through the mesoscopic structures, Landauer formalism and a generalized Green's function method. As illustrated in figure 1(a), each cell connects to the left (right) cell through two horizontal hopping integrals, t_0. Also, in a typical cell, the base-pair sites connect to each other through a hopping integral t_{GC} (t_{AT}) and connect to the upper and lower backbone sites via vertical hopping integrals, t. In this study, GC (TA) base pair in the first (end) cell is connected to the left (right) semi-infinite metallic leads as electron reservoirs. The full system is modelled by a generalized non-interacting Hamiltonian, namely,

$$H = H_{\mathrm{DNA}} + H_L + H_R + H_C \tag{1}$$

Where H_{DNA}, $H_{L(R)}$ and H_C represent the DNA Hamiltonian, the semi-infinite metallic leads, and their interaction with DNA molecule, respectively. The metallic leads are modelled within the tight-binding approximation with only one electron per atom. Accordingly, $H_{L(R)}$ describes reasonably the band structure of an isolated metallic wire, especially near the Fermi level, which is considered zero, thus,

$$H_\gamma = \sum_{i_\gamma} \varepsilon_{i_\gamma} c_{i_\gamma}^{+} c_{i_\gamma} - \sum_{i_\gamma} t_{i_\gamma, i_\gamma+1} \left(c_{i_\gamma}^{+} c_{i_\gamma+1} + c_{i_\gamma+1}^{+} c_{i_\gamma} \right). \tag{2}$$

Here $c_{i_\gamma}^{\dagger}$ ($c_{i\gamma}$) denotes the creation (annihilation) operator of an electron at site i in the metallic electrode $\gamma(=L \text{ or } R)$. ε_{i_γ} and $t_{i_\gamma, i_\gamma+1} = t_{i_\gamma}$ are the on-site energy and the nearest neighbour hopping integral, respectively. Also, the dsDNA ladder model Hamiltonian can be expressed in the form,

$$H_{\mathrm{DNA}} = \sum_n \hat{\varepsilon}_n d_n^{+} d_n - \sum_n \hat{t}_{i,i+1} \left(d_n^{+} d_{n+1} + d_{n+1}^{+} d_n \right). \tag{3}$$

Here $\hat{d}_n = \begin{pmatrix} d_n^{S_1} \\ d_n^{S_2} \end{pmatrix}$ where $d_n^{S_1}$ and $d_n^{S_2}$ are the charge destruction operators at site n in the two central conduction channels, named S_1 and S_2. $\hat{\varepsilon}_n = \begin{pmatrix} \varepsilon_n^{S_1} & t_{GC(AT)} \\ t_{GC(AT)} & \varepsilon_n^{S_2} \end{pmatrix}$, where ε_n^m with $n \in \{G, C, A, T\}$ is the on-site energy of a typical nucleobase in the strand $m \in \{S_1, S_2\}$. Moreover, $\hat{t}_{n,n+1} = \begin{pmatrix} t_{n,n+1}^{S_1} & 0 \\ 0 & t_{n,n+1}^{S_2} \end{pmatrix}$ in which $t_{n,n+1}^{m} = t_0$ is the nearest-neighbour transfer integral between nucleobases located at sites nth and $(n+1)$th in strand m. We note that

while the intrastrand hopping terms $t^{m}_{n,n+1}$, describing the $\pi-\pi$ orbital coupling, are included into $\hat{t}_{n,n+1}$ matrix, and the hopping integral $t_{GC(AT)}$ mediated interstrand coupling is incorporated into the $\hat{\varepsilon}_n$ matrix instead. Finally, H_C the DNA-metallic electrode coupling Hamiltonian is described.

Here $V_{L(R)}$ denotes the tunnelling coupling between the single electron states of the GC (AT) base pair in the cell of number 1(M) from the left (right) and the electron states in the left (right) metallic electrode.

Following the discovery quasicrystalline structures and the pioneer work of Schrödinger, who introduced the notion of a one-dimentional aperiodic crystal in order to describe the basic structure of genetic material [26], several models suggested to investigate the nature of charge transport in biological DNA molecule with millions of base pairs are aperiodically distributed. To describe the higher chemical complexity in DNA structure, determined by the base pair sequencing, we transform, by means of a renormalization process, the Hamiltonian of dsDNA molecule in ladder model to an effective Hamiltonian corresponding to a one-dimensional chain in which the effective on-site energies are arranged according to a Fibonacci sequence. The contributions of the hopping integrals t_{GC} and t_{AT} between the base-pairs and also the lateral hopping integrals t between each base and the backbone sites are decimated to obtain the one-dimensional array, as sketched in figure 1(b). Accordingly, we could arrange the GC and AT complementary pairs according to the Fibonacci sequence. Choosing $\alpha \equiv$ GC and $\beta \equiv$ AT and applying the inflation rule $\alpha \to \alpha\beta$ and $\beta \to \alpha$, one may construct the series of unit cells $\alpha, \alpha\beta, \alpha\beta\alpha, \alpha\beta\alpha\alpha\beta, \alpha\beta\alpha\alpha\beta\alpha\beta\alpha, \ldots$ for sequences of length 1, 2, 3, 5, 8, …, respectively. In this model the effective on-site energies $\tilde{\varepsilon}_\alpha$ and $\tilde{\varepsilon}_\beta$ are given as [19],

$$\tilde{\varepsilon}_{\alpha(\beta)}(E) = t_{\alpha(\beta)} + \frac{2t^2}{E - \varepsilon_{\text{backbone}}} \quad , \tag{5}$$

where, $t_\alpha = t_{GC}$ and $t_\beta = t_{AT}$ are the hopping integrals interconnecting the base-pair sites to each other. Then the renormalized one-dimensional monoatomic lattice Hamiltonian in the Fibonacci model is written as,

$$H_r = \sum_{j=1}^{N} \tilde{\varepsilon}_j(E)\, b_j^{+} b_j - \sum_{j=1}^{N-1} t_{j,j+1}\left(b_j^{+} b_{j+1} + b_{j+1}^{+} b_j\right) , \tag{6}$$

where $b_j^{\dagger}(b_j)$ denotes the creation (annihilation) operator of an electron at site j on the monoatomic chain. $t_{j,j+1} = t_0$ is the hopping between the renormalized "atoms" corresponding to complementary GC (AT) base pairs in the original dsDNA molecule whose on-site energies $\tilde{\varepsilon}_j(E)$ depend on the charge carrier energy, the hopping integral $t_{GC}\left(t_{AT}\right)$ between G and C (A and T) bases, and also on the hopping t between each base and the upper and lower backbone sites. Because of ignored inelastic scatterings, to calculate the current through the model structure, we use Landauer-Büttiker formalism under an applied bais of V_a. It is written in the form [27, 28],

$$I(V) = \frac{e}{h}\int_{-\infty}^{+\infty} \left[f_L(E) - f_R(E)\right] T(E)\, dE \quad , \tag{7}$$

where $f_{L(R)} = f(E - \mu_{L(R)})$ gives the Fermi distribution function of the electrons within the metallic electrodes having chemical potentials $\mu_{L(R)} = E_F \pm eV_a/2$. E_F is the equilibrium Fermi energy. The driving force, here, is the electric potential bias. Furthermore, $T(E) = \mathrm{Tr}\left\{\hat{\Gamma}_L G \hat{\Gamma}_R G^{+}\right\}$ is the energy-dependent transmission function. The matrix $\hat{\Gamma}_{L(R)} = i\left(\hat{\Sigma}_{L(R)} - \hat{\Sigma}^{+}_{L(R)}\right)$ corresponds to the chemisorption's coupling at the electrode-molecule interface, where $\hat{\Sigma}_{L(R)}$ represents the self-energy matrix introduced to incorporate the effects of the semi-infinite metallic electrodes coupled to the molecule. In addition, the Green's function of the molecule coupled to the electrodes (as the source and drain) in the presence of the bias voltage is given as,

$$\hat{G}(E, V_a) = \left[E\hat{1} - \hat{H}_{\text{DNA}} - \hat{\Sigma}_L\left(E - \frac{eV_a}{2}\right) - \hat{\Sigma}_R\left(E + \frac{eV_a}{2}\right)\right]^{-1} . \tag{8}$$

Thus, the core of the problem lies in the computing of the energy-dependent self-energy matrices $\hat{\Sigma}_L$ and $\hat{\Sigma}_R$. In the case of coupling through a GC (AT) base pair, only one element of each self-energy matrix is non-zero. Based on Dyson equation, the matrix elements of the self-energy $\Sigma_{L(R)}(E)$ may be carried out as [29],

$$\Sigma_{L(R)}(E) = \frac{V^2_{L(R)}}{E - \varepsilon_i - \Gamma_{L(R)}} \quad , \tag{9}$$

where Γ^{L}_{11} and Γ^{R}_{MM} are the only non-zero elements of matrix $\hat{\Gamma}_{L(R)}$ and given as the following expression [30],

$$\Gamma^{L(R)}_{11(MM)}(E) = \left(\frac{E - \varepsilon_i}{2}\right) - i\left[t_i^2 - \left(\frac{E - \varepsilon_i}{2}\right)^2\right]^{1/2} \tag{10}$$

Here $\varepsilon_i = 0$ and $t_i = 1\text{eV}$ are the onsite energy and the nearest-neighbor hopping integral of the metallic electrodes, respectively.

3. Results and discussion

Based on the formalism presented in section 2, some significant properties of the electronic conduction of the

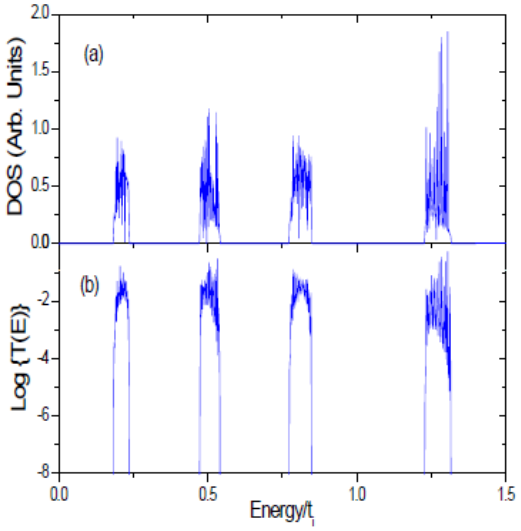

Figure 2. (color online) Panels (a) and (b) show the electronic density of states (DOS) and the logarithm of the transmission coefficient versus the dimensionless parameter $Energy / t_i$ for the dsDNA in the metal/DNA/metal junction.

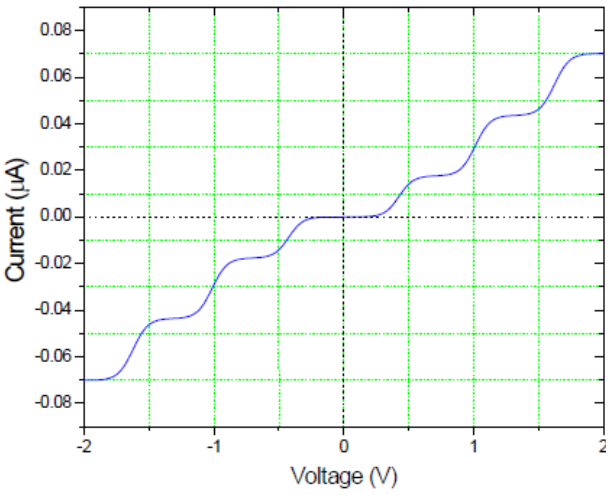

Figure 3. (color online) The current-voltage characteristic of the dsDNA molecule at room temperature (T=300 K) for the metal/DNA/metal junction. The curve shows clearly a nonlinear dependence. The model parameters used as mentioned into the text.

metal/DNA/metal model junction were studied. To proceed, we study the electronic density of states (DOS) and the electron transmission coefficient, $T(E)$ through the DNA molecule in the foregoing junction. Panels (a) and (b) in figure 2, illustrate the DOS and the logarithm of $T(E)$ for the junction, respectively. Here, the rescaled parameters are used for the onsite energies of poly(GACT)-poly(CTGA) structure. The parameters adapted for the dsDNA Hamiltonian in this work are rescaled by the backbone on-site energy $\varepsilon_{\text{backbone}} = 12.27\,\text{eV}$ [31]. Thus, $\varepsilon_x = \varepsilon_x^0 / \varepsilon_{\text{backbone}}$ with x = G, C, A and T is eventuated. Based on this relation $\varepsilon_G = 0.633$, $\varepsilon_C = 0.723$, $\varepsilon_A = 0.672$ and $\varepsilon_T = 0.744$ are obtained. Moreover, the hopping integrals in dsDNA structure are set as $t_{GC} = 0.90\,\text{eV}$, $t_{AT} = 0.34\,\text{eV}$, $t = 1.5\,\text{eV}$ and $t_0 = 0.15\,\text{eV}$. In addition, the tunnelling coupling strength between molecule and the left (right) metallic electrode and the number of cells for the dsDNA are set $V_{L(R)} = 0.3\,\text{eV}$ and M=245, respectively. Figure 2 displays the four allowed bands for poly(GACT)-poly(CTGA) structure, which are separated, from right to left, by the relatively broad gaps $\Delta_{GA} \cong 0.459\,\text{eV}$, $\Delta_{AC} \cong 0.305\,\text{eV}$ and $\Delta_{CT} \cong 0.3\,\text{eV}$. As expected, the states in the guanine band have more contribution to the charge conduction. This may be attributed to the richness of π-orbitals on the base of guanine [28]. In addition, as mentioned before, the onsite potential of guanine is lower than that of cytosine, adenine and thymine. Figure 3 shows the room temperature I-V characteristic of the junction in a step-wise and smoothed curve. This plot shows clearly a nonlinear dependence. Our results represent a good qualitative agreement with the electronic structure of the system (figure 2) along the zero current part of the I-V curve. In other words, there is a zero-current gap $\cong 315\,\text{V}$ in the I-V characterization of the junction coincidence with the band gap with the average magnitude of $\cong 321\,\text{eV}$ between the nucleobase bands. The zero-current gap reflects that many of the electronic states close to the gaps in the electronic structure have a very low transmission probability. In addition, the low-voltage part of the I-V curve arises from the semiconducting behaviour of the poly(GACT)-poly(CTGA) DNA molecule. Figures 4 and 5 show the corresponding results for the renormalized structure of dsDNA molecule in Fibonacci model. According to figure 1(b), a one-dimensional site Fibonacci lattice with $\widetilde{\varepsilon}_\alpha$ and $\widetilde{\varepsilon}_\beta$ as the effective onsite energies, are arranged based on Fibonacci sequence with generation 15 and 987 sites connected to two semi-infinite metallic leads. Panels (a) and (b) in figure 4, illustrate the DOS and the logarithm of $T(E)$. As expected, the spectra are fully self-similar, i.e., the peak clusters and the gaps are arranged in a very similar way. In fact, the self-similarity in DOS and thereby in the transmittance spectrum is the reflection of the existence of the self-similarity in the corresponding energy spectrum. Figure 4(b) in a remarkable coincidence with figure 4(a), shows that the electronic states associated to the corresponding energy eigenvalues are non-localized and from view point of the electronic conduction, these eigenstates are extended. However, in order to obtain a complete picture of the role of the extended states on the electronic conduction, it is necessary to calculate the I-V characteristic of the system. Figure 5 illustrates the current-voltage

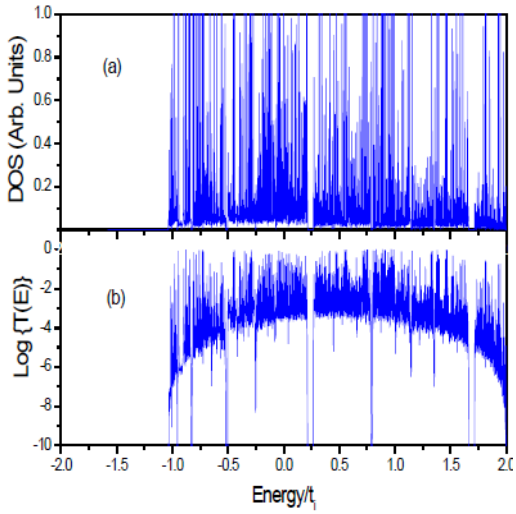

Figure 4. (color online) Panels (a) and (b) show the electronic density of states (DOS) and the logarithm of the transmission coefficient versus the dimensionless parameter $Energy / t_i$ for the DNA Fibonacci model. The model parameters are used as mentioned in the text.

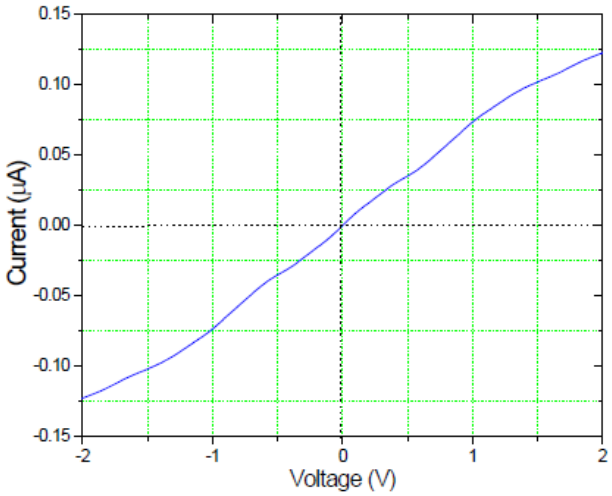

Figure 5. (color online) The current-voltage characteristic of the DNA molecule in the Fibonacci model at room temperature (T=300 K). As it is clear, the curve shows an ohmic-like behavior.

characteristic of one-dimensional Fibonacci lattice according to figure 1(b). Our results indicate that the DNA molecule which primarily acts as a semiconductor in the ladder model, moves toward becoming an ohmic-like conductor in the site Fibonacci model. In fact, in the suggested Fibonacci lattice for DNA molecule, the band gaps of the semiconductor structure of dsDNA are basically modified and have a tendency for suppression in such a way that its I-V characteristic shows an ohmic-like behavior.

4. Conclusion

In conclusion, we have investigated the effect of the mapping of double-stranded DNA molecule (dsDNA) in a ladder model to a one-dimensional site Fibonacci lattice on its electronic conduction properties. Our results indicate that in this process the semiconducting behaviour of dsDNA molecule is basically modified and the band gaps of dsDNA structure have a tendency for suppression when the dsDNA structure is mapped to a one-dimensional site Fibonacci lattice and the room temperature I-V characterization of the molecule shows an ohmic-like behavior.

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