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Characteristics of alternating current hopping conductivity in DNA sequences*

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This paper presents a model to describe alternating current (AC) conductivity of DNA sequences, in which DNA is considered as a one-dimensional (1D) disordered system, and electrons transport via hopping between localized states. It finds that AC conductivity in DNA sequences increases as the frequency of the external electric field rises, and it takes the form of $\sigma_{\rm ac}(\omega) \sim \omega^2 \ln^2(1/\omega)$. Also AC conductivity of DNA sequences increases with the increase of temperature, this phenomenon presents characteristics of weak temperature-dependence. Meanwhile, the AC conductivity in an off-diagonally correlated case is much larger than that in the uncorrelated case of the Anderson limit in low temperatures, which indicates that the off-diagonal correlations in DNA sequences have a great effect on the AC conductivity, while at high temperature the off-diagonal correlations no longer play a vital role in electric transport. In addition, the proportion of nucleotide pairs p also plays an important role in AC electron transport of DNA sequences. For $p < 0.5$, the conductivity of DNA sequence decreases with the increase of p, while for $p \geq 0.5$, the conductivity increases with the increase of p.

Keywords: DNA sequences, AC hopping conductivity, off-diagonal correlations
PACC: 8715, 7220

1. Introduction

Electronic transport in DNA, first proposed by Eley in 1962,[1] is attracting a considerable amount of attention for its biological functions such as the repair mechanism after radiation damage and biosynthesis.[2,3] Recent experiments have demonstrated that there is a long-range charge migration in DNA, indicating that the DNA sequence is a candidate for one-dimensional (1D) molecular electronics.[4,5] It is thus not surprising that DNA molecules have become the subject of intense study concerning their potency to carry electric charges,[6−9] and to provide a scaffold for a metal assembling of highly conductive nano-wires.[10,11] Although the use of DNA molecules in nano-electronic circuits is a very promising task due to their self-assembly and molecular recognition abilities, their conductivity properties of direct experimental measurements are still under intense debate due to the complexity of the environment and the molecule itself.[12−19]

Concerning theory, both *ab initio* calculation[20−22] and model-based Hamiltonians[23−25] are extensively adopted to interpret the diversity of the experimental results and to ascertain the underlying charge transport mechanisms. Some consensus on the mechanisms of charge migration in DNA seems to emerge. The charge transport may result from either coherent tunneling or hopping. Recently, the energy levels and electronic states of DNA were studied,[26,27] the observed temperature dependence of the conductivity in DNA was studied by considering DNA as a 1D disordered system, and electrons transport via hopping between localized states.[28] The effects of interfacial couplings on transport properties in DNA were studied by Meng *et al.*[29] It is also found that the introduction of diagonal disorder and the role of on-site energy correlations in DNA sequences markedly changes their physical characteristics and can play a crucial role in their electron transport.[30−32] In addition, the influence of off-diagonal correlations which is proved to affect the transport properties effectively

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was investigated,[33,34] and the current-voltage (I–V) characteristics showed typical semi-conducting features with a gap ranging from 0.8 eV to 2.1 eV.[35] However, these studies concentrated on the case of an externally applied direct current (DC) field, and paid little attention to the case of an AC field which is proved to affect the electronic localization properties effectively.[36,37] In this paper, by considering that electrons transport via hopping in DNA sequences which can be viewed as a 1D disordered system, we consider the influence of an applied AC field on electron transport in DNA sequences, and present the characteristics of frequency dependence of the conductivity in DNA. We also study the effect of concentration of one type of nucleotide pairs on the AC conductivity, and the temperature dependence of the AC conductivity. Moreover, we also investigate the role of correlated off-diagonal disorder and their impact on the AC hopping conductivity in DNA sequences.

This paper is organized as follows. In the next section, we present our theoretical model based on an electronic tight-binding Hamiltonian suitable to describe DNA sequences. Then we introduce the AC conductivity formula in the externally applied AC field. In Section 3 we investigate frequency dependence of the conductivity in DNA, we also investigate the effect of proportion of nucleotide pairs and temperature on the electric transport of DNA sequences, and the role of correlated off-diagonal disorder and their impact on the AC hopping conductivity in DNA sequences. Finally, the conclusions of this work are presented in Section 4.

2. Method of calculations

A DNA chain is a sequence of four possible nucleotides which define the structure of the amino acids to form proteins. Thus the DNA nucleotide sequences can be considered as a symbolic sequence of a four-letter alphabet, namely, guanine (G), adenine (A), cytosine (C) and thymine (T). Considering the pairing of complementary bases (A with T and G with C) in the inside of the chain, where each base is covalently bounded to the backbone and bind to its complementary via hydrogen binding, we can view a DNA chain with random base pair sequences as a 1D binary disordered system composed of base pairs A–T and C–G in a typically random order.[31]

In order to study the electronic property of a DNA sequence, we map it onto a binary alphabet, in which the unit of nucleotides A and T gives $\varepsilon_{\rm A} = 0.1$ eV and that of nucleotides C and G $\varepsilon_{\rm B} = 0$ (reference point),[33,34] according to the *ab initio* calculation of Endre *et al.*[38] Using the simplest form of the tight-binding disordered model representing a single electron band, we find that the Hamiltonian is[31]

$$H = \sum_{i=1}^{N} \varepsilon_i \left|i\right\rangle \left\langle i\right| + \sum_{i=1}^{N} \sum_{j \neq i}^{N} t_{ij} \left|i\right\rangle \left\langle j\right|, \qquad (1)$$

where N is the number of base pairs. The length of a DNA sequence is $L = Na$ ($a = 0.34$ nm). The quantity ε_i is the energy assigned to each random base pair site i which takes two values $\varepsilon_{\rm A}$ and $\varepsilon_{\rm B}$ with probabilities p and $1 - p$. Obviously, $|\varepsilon_{\rm A} - \varepsilon_{\rm B}|$ quantifies the spread of the site energy distribution, namely, the degree of disorder W. The quantities t_{ij} are the off-diagonal elements which represent electronic overlap (hopping term) between the wave-functions of electrons centered at two neighboring sites i, $j(|i - j| = 1)$, and according to the *ab initio* calculation of Endre *et al*,[38] the values of t_{ij} are assumed to be

$$t_{ij} = \begin{cases} t_{\rm AA} = -0.0695 \text{ eV}, & \varepsilon_i = \varepsilon_j = \varepsilon_{\rm A} \\ t_{\rm BB} = -0.1403 \text{ eV}, & \varepsilon_i = \varepsilon_j = \varepsilon_{\rm B} \\ t_{\rm AB} = \dfrac{t_{\rm AA} + t_{\rm BB}}{2} = -0.1049 \text{ eV}, & \varepsilon_i \neq \varepsilon_j \end{cases}, \quad |j - i| = 1. \qquad (2)$$

Obviously, the off-diagonal correlation is built in through the values of t_{ij}. This model is simply generalized from the Anderson model, which is the limit for $t_{\rm AA} = t_{\rm BB} = t_{\rm AB} = -0.1403$ eV.[33] Then the Schrödinger equation for the Hamiltonian (Eq.(1)) becomes

$$E\phi_i = t_{i-1,i}\phi_{i-1} + \varepsilon_i\phi_i + t_{i,i+1}\phi_{i+1}, \quad (3)$$

where E is the energy corresponding to the electron wave-function. The $|\phi_i|^2$ is the probability of finding an electron at site i in a DNA random base pair sequence, and $|\varphi\rangle = \sum_i \phi_i |i\rangle$ is an eigenstate with energy E. Based on Dean and Martine's negative eigenvalue theorem[39] and Wu and Zheng's infinite order perturbation theorem,[40] the eigenstates of the DNA chain can be solved. As expected, the electron wave-functions of the DNA sequence present the property of localization, each localized wave-function has its own central localized position, and the localization length $\lambda(E_i)$ can also be computed. As we know, an electron can hop between pairs of localized states and give rise to a finite energy dissipation, which is responsible for the AC conductivity of DNA sequences.

In the presence of an external electric field $F(t) = F_0\cos(\omega t)$, an electron hops between states $|i\rangle$ and $|j\rangle$ with energy E_i and E_j respectively (E_i and E_j are the eigenvalues of the states localized on sites i and j respectively, which are measured from the Fermi level), due to resonant absorption or emission of photons. By absorbing a photon with energy $\hbar\omega$, electron hops from state $|i\rangle$ to state $|j\rangle$ and the exchanged energy is given by

$$q_{ij} = \Gamma_{ij}\hbar\omega f(E_i)[1 - f(E_j)]\delta[\hbar\omega - (E_j - E_i)], \quad (4)$$

where

$$f(E_i) = \left[\exp\left(\frac{E_i}{k_{\rm B}T}\right) + 1\right]^{-1}$$

is the occupation factor of the i-th site, T is the temperature and $k_{\rm B}$ is the Boltzmann constant. The Γ_{ij} is the hopping rate, which can be given by

$$\Gamma_{ij} = \begin{cases} v_0 \exp\left(-2\alpha x_{ij} - \dfrac{E_j - E_i}{k_{\rm B}T}\right), & E_j > E_i, \\ v_0 \exp(-2\alpha x_{ij}), & E_j < E_i, \end{cases} \quad (5)$$

where v_0 is the attempt-to-jump frequency which should be a typical phonon frequency, α^{-1} is the average localization length, $x_{ij} = |x_j - x_i| = |j - i|\, a$ is the electron hopping distance.

Corresponding to a reversed analog of the absorption process, an electron hops from state $|j\rangle$ to state $|i\rangle$ by emission of a photon with energy $\hbar\omega$, the exchanged energy can be given by

$$q_{ji} = \Gamma_{ji}\hbar\omega f(E_j)[1 - f(E_i)]\delta[\hbar\omega - (E_j - E_i)]. \quad (6)$$

Obviously,

$$\Gamma_{ji} = \Gamma_{ij}\exp\left(-\frac{\hbar\omega}{k_{\rm B}T}\right),$$

and we can obtain the exchanged energy between the nucleotide pairs per unit time as

$$q_i = \frac{2\hbar\omega}{Z}\Gamma_{ij}\exp\left(\frac{E_i}{k_{\rm B}T}\right)\sinh\left(\frac{\hbar\omega}{k_{\rm B}T}\right) \times \delta[\hbar\omega - (E_j - E_i)], \quad (7)$$

where

$$Z = 1 + \exp\left(\frac{E_i}{k_{\rm B}T}\right) + \exp\left(\frac{E_i}{k_{\rm B}T}\right) + \exp\left(\frac{E_i + E_j}{k_{\rm B}T}\right),$$

and the total energy dissipation in a DNA sequence per unit time can be described as $q = \sum_i q_i$. In terms of electrodynamics, the total work done over a time interval by electric field is equal to the total energy dissipation in a DNA sequence, that is,

$$q = \frac{1}{2}\sigma_{\rm ac}(\omega)F_0^2,$$

then the AC conductivity of a DNA sequence can be gotten as

$$\sigma_{\rm ac}(\omega) = \frac{4\hbar\omega}{F_0^2}\sum_i \frac{\Gamma_{ij}}{Z}\exp\left(\frac{E_i}{k_{\rm B}T}\right)\sinh\left(\frac{\hbar\omega}{k_{\rm B}T}\right) \times \delta[\hbar\omega - (E_j - E_i)]. \quad (8)$$

3. Numerical results and discussions

Based on the AC conductance formula, we calculate the AC conductivity of DNA sequences with $N = 2000$ base pairs, and F_0 fixed at $F_0 = 1 \times 10^3$ V/cm.

The frequency-dependent AC conductivity $\sigma_{\rm ac}(\omega)$ in DNA sequences with off-diagonal correlations which can be seen as a 1D chain is plotted in Fig.1, in which $\log_{10}\sigma_{\rm ac}(\omega)$ is plotted vs. $\log_{10}\omega$. From Fig.1, we can find that the AC conductivity $\sigma_{\rm ac}(\omega)$ in DNA sequences with off-diagonal correlations increases with the increase of the frequency of the external electric field. In our opinion, this phenomenon is due to the fact that, in a 1D disordered chain, the states are localized, the conductivity of the chain stems from electron hopping between the localized states by resonantly absorbing or emitting photons. That is, when the frequency of the external electric field rises, the energy of the photon is increased. Therefore, the AC conductivity of DNA sequences increases as the frequency of the external electric field rises.

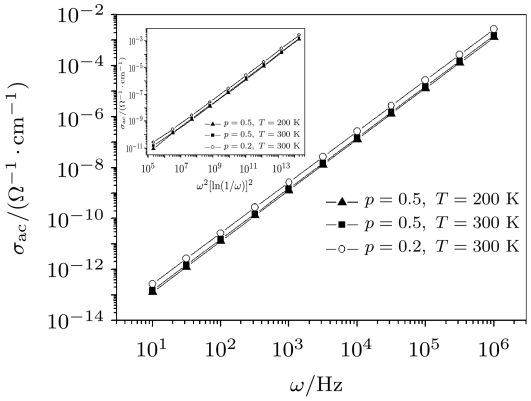


Fig.1. Frequency-dependence of AC conductivity in DNA sequences.

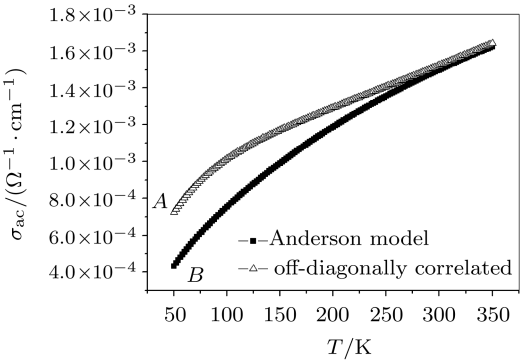


Fig.2. The temperature dependence of the AC conductivity in DNA sequences.

According to the result of Rosenow,[41] which suggests that the AC conductivity in the 1D chain takes the form of $\sigma_{\rm ac}(\omega) \sim \omega^2 \ln^2(1/\omega)$, in the inset of the figure, we plot the AC conductivity as a function of $\omega^2 \ln^2(1/\omega)$ for the same parameter. It clearly shows that the AC conductivity $\sigma_{\rm ac}(\omega)$ increases with $\omega^2 \ln^2(1/\omega)$ in DNA sequences, which fits well with the result of Rosenow. Therefore, we think that the AC conductivity of the DNA sequence also takes the form $\sigma_{\rm ac}(\omega) \sim \omega^2 \ln^2(1/\omega)$.

Meanwhile, it can be seen in Fig.1 that the AC conductivity of DNA sequences shows a weak dependence on the temperature and concentration of the nucleotide pairs. To investigate the effect of temperature on the AC conductivity, the conductivity at a fixed frequency ($\omega = 10^6$ Hz) was shown between 50 K and 350 K in Fig.2. And in order to investigate the role of off-diagonal correlation and their impact on the AC conductivity in DNA sequences, we also study the case of the Anderson model with the same parameter for the purpose of comparison. It can be seen that just as in the DC case,[42] the AC conductivity of DNA sequences (both curve A and curve B) also increases with the increase of temperature, revealing that with the increase of temperature, which corresponds to an increase in delocalization, the thermal movement of base-pairs will be stimulated gradually, which will be beneficial to the hopping of electrons. That is, the AC conductivity also has an activated behavior with the increase of temperature. However, comparing with the DC case, we can find that the absolute value of the DC conductivity increases several orders of magnitude, while the AC conductivity shows only a weak increase when the temperature goes from 50 K to 350 K, revealing that the thermal activated behavior has more effect on the DC conductivity than that on the AC conductivity.

It is obvious that the AC conductivity in a correlated case (curve A) is much larger than that in an uncorrelated case of the Anderson limit (curve B) at low temperatures, while at high temperatures, curve A merges with curve B. The result indicates that off-diagonal correlations have a great effect on the AC conductivity, especially at low temperatures. As we know, correlation leads to delocalization of the electrical wave-function, and delocalization of the electrical wave-function drastically enhances the hopping rate of electrons. Therefore, the AC conductivity in a correlated case (curve A) is much larger than that in an uncorrelated case of the Anderson limit (curve B) at low temperatures. But as temperature increases, the effective disorder also increases to some extent, and the scattering increases, which leads to the decrease of the mean hopping rate of the electrons. The "good" transport due to off-diagonal correlations is cut down. As a result, the off-diagonal correlations which lead to "good" transport at low temperatures do not play a vital role in AC conductivity of DNA sequences at high temperatures. That is, the off-diagonal correlations have more effect on the conductivity at low temperatures than that at high temperatures.

Figure 3 shows the dependence of the AC conductance on the concentration p of the A–T base pair. It can be seen that the AC conductivity of DNA sequences (both curve A and curve B) firstly decreases and then increases with the increase of p. It implies that the concentration of one type of nucleotide pairs plays an important role in electronic transport in DNA. Obviously, probability p represents the disorder in a DNA sequence to some extent. For example, when $p = 0$ or $p = 1.0$, the DNA chain is composed of only one kind of base pairs, so it can be regarded as a perfect periodic sequences with no disorder, and

$p = 0.5$ represents the maximum disorder case. For $p < 0.5$, the strength of disorder increases with the

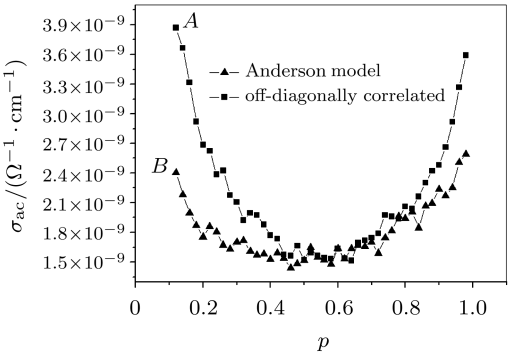


Fig.3. The dependence of the AC conductance on the concentration of A–T base pair p.

increase of the percentage of nucleotide pairs A–T, which leads to an increase of localization of wave-functions and results in the decrease of the jump rate. However, for $p \geq 0.5$, the strength of disorder decreases with the increase of the percentage of nucleotide pairs A–T, and results in the delocalization of wave-functions and the jump rate increases. Consequently, the conductivity of DNA sequences decreases with the increase of $p < 0.5$, while for $p \geq 0.5$, the conductivity increases with the increase of p.

This result also coincides with the result of Dunlap and Phillips, which suggests that in a binary disordered system,[43,44] when the concentration of one type of site is small, the probability for two nearby sites to have the same on-site energy is small. In this case, extended states tend to exist, which leads to the drastic increase of conductivity of the system. As a result, in DNA sequences which can be viewed as a 1D binary disordered system composed of base pairs A–T and C–G in a typically random order, the AC conductivity presents the characteristic that the conductivity is high when the concentration of one type of base pairs is small.

4. Conclusions

From the above discussion, we can draw conclusions as follows:

(i) AC conductivity $\sigma_{ac}(\omega)$ of DNA sequences increases as the frequency of the external electric field rises, and takes the form of $\sigma_{ac}(\omega) \sim \omega^2 \ln^2(1/\omega)$.

(ii) AC conductivity of DNA sequences increases with the increase of temperature.

(iii) AC conductivity in an off-diagonally correlated case is much larger than that in an uncorrelated case of the Anderson limit at low temperature, while at high temperature the off-diagonal correlations no longer plays a vital role in electronic transport.

(iv) The proportion of nucleotide pair concentration p plays an important role in AC electronic transport of DNA sequences. For $p < 0.5$, the conductivity of DNA chains decreases with the increase of p, while for $p \geq 0.5$, the conductivity increases with the increase of p.

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