Superexchange interaction enhancement of the quantum transport in a DNA-type molecule*

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We use the transfer matrix method and the Green function technique to theoretically study the quantum tunnelling through a DNA-type molecule. Ferromagnetic electrodes are used to produce the spin-polarized transmission probability and therefore the spin current. The distance-dependent crossover comes from the topological variation from the one-dimensional to the two-dimensional model transform as we switch on the interstrand coupling; a new base pair will present $N-1$ extrachannels for the charge and spin as N being the total base pairs. This will restrain the decay of the transmission and improve the stability of the quantum transport. The spin and charge transfer through the DNA-type molecule is consistent with the quantum tunneling barrier.

Keywords: quantum transport, DNA, superexchange

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1. Introduction

In the last decades, the fact that electrons and holes transporting through a DNA molecule depends on long distance has attracted considerable attention in the fields of physics, biology and chemistry.[1−4] Studying the charge and spin migration in DNA will help people have a better understanding of DNA oxidation and self-repair, and explore the functioning of molecular electronic devices.[5−10] In particular, in the last decade, progress in the nanofabrication of quantum devices has enabled us to study the electron transport through the mesoscopic system in a controllable way,[11−15] since a wide range of energy can be achieved by continuously changing the applied external potential in contrast with the real atoms. Then the DNA molecule can be regarded as a double-stranded artificial quantum-dot (QD) array.[16] Experiments and theoretical calculation on mesoscopic transport on a single artificial or DNA duplex chain contacted between metallic electrodes have also been the kernel subject of controversial debates.

When the size of the system is reduced to the electron coherence length, the transmission probability will play an essential role in the quantum transport and we can gain the conductance of the devices by the Landauer–Büticker formula $G = (2e^2/h)T$, where e is the electron charge, h is the Planck constant and T is the transmission probability of the devices.[17,18] Spintronics has become a new branch of condensed matter physics and materials science where not only the charge but also the spin degree of freedom of electrons play an essential role.[19] It is of fundamental importance to study the mechanism to generate and control the spin-polarised currents in spintronic devices, which may promise practical applications in quantum computing and information.[20,21]

Recently, calculations and experiments both have shown that the transmission in a quantum-dot array is strongly dependent on the length of the model and exponential decay;[22,23] but when we come to the double-stranded model, experiments have shown that the transfer rate is of weak dependence on the length.[24] How to explain this profound difference of the transmission between these two models and what is the underlying mechanism of the carrier transfer in double-stranded DNA molecule are not clear as yet.[25,26] People have developed several methods to deal with the transfer in DNA, but an accepted fact is that the sequence of the base pairs (made up from nucleotides: guanine G; adenine A; cytosine C and thymine T)[27−30] in the DNA will account for the transmission between the strong and weak dependence on the length of the molecule. In 2001, Giese *et al.*[26] measured the transmission through DNA based on the

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sequence $(G:C)(T:A)_N(G:C)_3$ for different N. They have shown that $N=3$ is a critical value in such a system: when $N<3$ the transmission is strongly dependent on the length and when $N>3$ the transfer rate will be weakly dependent on the distance. They even pointed out that the transfer rate will be almost not dependent on distance when $N>3$. It is shown that although the long-range efficiency will be reduced by the length of the single lane, but the π-stacked array of DNA base pairs could provide an extended path to convey the long-range charge transport. Berlin *et al.* suggested that the variable-range hopping model (combination of coherent superexchange and a hopping mechanism) can allow for such a transition,[22] i.e., the superexchange in the DNA bridge may play an important role in the charge and spin transport.

In the former process, the discussion about DNA is focused on the charge transfer through it, while the spin-polarised transport in a DNA-type molecule has not yet been investigated. In this paper, ferromagnetic electrodes are used to produce the spin-polarised transmission probability. In terms of the tight-binding model and the Green function technique, we will demonstrate that the spin transport distance-dependent crossover is a quantum tunneling barrier in nature. This paper is organised as follows. In Section 2, the Hamiltonian of a tight binding two-strand DNA model is given. In Section 3, the calculation, based on transfer matrix and Green function technique, and the numerical results are presented. The final section gives a conclusion.

2. Tight binding Hamiltonian of the DNA

The system consisting of a DNA duplex chain with N Watson-Crick base pairs connected to four ferromagnetic electrodes with one for each end as shown in Fig. 1. The first strand has the base sequence $G(T)_NG$ and the second has sequence as $C(A)_NC$, the electrodes contact the left ends of each DNA strand as the injector and the others contact the right ends as the drain. The magnetic moment M of the left electrodes are pointing to the z-direction, the electric current is flowing in the y-direction, while the moments of the right electrodes make angles θ_1 and θ_2 with the z-axis in the x–z plane on the first and second strands, respectively. In terms of the tight binding model, the Hamiltonian of this duplex could be written as

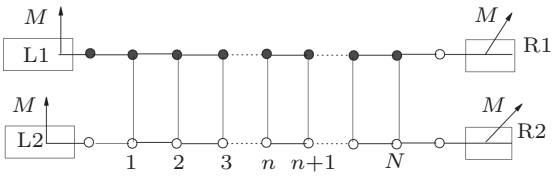


Fig. 1. DNA duplex chain with N Watson–Crick base pairs connected to four ferromagnetic electrodes.

$$H = H_{\rm L} + H_{\rm R} + H_0 + H_{\rm I},$$

where $H_{\rm L}$ and $H_{\rm R}$ denote the left and the right electrodes, respectively, to which a DC bias potential is applied,

$$\begin{aligned} H_{\rm L} &= \sum_{k,\sigma}(\varepsilon_{k,{\rm L}} + \sigma M)C^{+}_{k{\rm L}\sigma}C_{k{\rm L}\sigma}, \\ H_{\rm R} &= \sum_{k,\sigma,i}(\varepsilon_{k,{\rm R}} + \sigma M\cos\theta_i)C^{+}_{k{\rm R}\sigma}C_{k{\rm R}\sigma} \\ &\quad + \sum_{k,\sigma,i} M\sin\theta_i C^{+}_{k{\rm R}\sigma}C_{k{\rm R}\bar{\sigma}}; \end{aligned}$$

H_0 describes the duplex chain of DNA,

$$\begin{aligned} H_0 &= \sum_{n=1}^{N}[\varepsilon_n a^{+}_{n,\sigma}a_{n,\sigma} - t_{n,n+1} \\ &\quad \times (a^{+}_{n,\sigma}a_{n+1,\sigma} + a^{+}_{n+1,\sigma}a_{n,\sigma})] \\ &\quad + \sum_{n=1}^{N}[u_n d^{+}_{n,\sigma}d_{n,\sigma} - h_{n,n+1} \\ &\quad \times (d^{+}_{n,\sigma}d_{n+1,\sigma} + d^{+}_{n+1,\sigma}d_{n,\sigma})] \\ &\quad - \sum_{n=1}^{N}\lambda_n(a^{+}_{n,\sigma}d_{n,\sigma} + d^{+}_{n,\sigma}a_{n,\sigma}). \end{aligned}$$

$H_{\rm I}$ models the coupling between electrodes and the DNA chain region with hopping integral constant. To simplify the analysis and calculation, we assume the hopping constant to be independent of spin index, then we have

$$\begin{aligned} H_{\rm I} &= \sum_{k} t_{0,1}(C^{+}_{k{\rm L}\sigma}a_1 + C^{+}_{k{\rm L}\sigma}d_1 + {\rm h.c.}) \\ &\quad + t_{N,N+1}(C^{+}_{k{\rm R}\sigma}a_N + C^{+}_{k{\rm R}\sigma}d_N + {\rm h.c.}). \end{aligned}$$

In these expressions, $C^{+}_{k{\rm L}\sigma}$ and $C^{+}_{k{\rm R}\sigma}$ are the creation operators for the electron with spin index σ inside the L- and R-electrodes, $\varepsilon_{k,{\rm L}} = \varepsilon^0_k + qv_{\rm L}$ ($\varepsilon_{k,{\rm R}} = \varepsilon^0_{\rm R} + qv_{\rm R}$) is the on-site energy. For DNA region, $a^{+}_{n,\sigma}$ ($d^{+}_{n,\sigma}$) is the creation operator in the first (second) strand on site n ($1 \leqslant n \leqslant N$), the corresponding on-site energy is denoted by $\varepsilon_n(u_n)$, which is equal to the highest occupied molecular orbit (HOMO) energy of the base on this site in the DNA chain and the centre of conduction band in the electrodes.The coupling parameter of the first (second) strand $t_{n,n+1}$ ($h_{n,n+1}$)

is equal to the intrastrand coupling parameter t_d between neighbouring sites n and $n+1$ of the DNA chain, one-fourth of the conduction bandwidth in the ferromagnetic electrodes t_m for $n \leqslant 0$ and $n \geqslant N+1$, and the coupling strength $t_{0,1}$and $t_{N,N+1}$ between the DNA chain and the electrodes at the sites of $n = 0$ and $n = N$. The interstrand coupling between sites in the same Watson–Crick base pair, i.e., the strength of the superexchange between the two strands, is described by λ_n. To minimize the contact effect, we assume a strong coupling between the ferromagnetic electrodes and the ends of the DNA region, such as $t_{0,1} = t_{N,N+1} = t_{dm} \geqslant 1.5$ eV. The main quantities which we have to calculate in our formulation are the transmission probabilities T_{ij} of electrons from electrode j to electrode i, and the reflection probabilities R_{jj} in the electrode j which is considered as an input terminal. For the four-terminal system we have $j = 1, 2$ for the left electrodes and $i = 3, 4$ for the right electrodes with $T_3(\sigma)$ denoting the transmission probability from the left electrodes to the right electrodes of the first strand, and $T_4(\sigma)$ for the other right electrode connected to the second strand. We begin with the stationary Schrödinger equation $H|\Psi\rangle = E|\Psi\rangle$, which possesses a general solution of the form

$$|\Psi\rangle = \phi_{n,\sigma} + \varphi_{n,\sigma} = C_{1,n}a^{+}_{n,\sigma}|0\rangle + C_{2,n}d^{+}_{n,\sigma}|0\rangle\,, \quad (1)$$

where $\phi_{n,\sigma} = C_{1,n}a^{+}_{n,\sigma}|0\rangle$ is the wave function of the first strand and $\varphi_{n,\sigma} = C_{2,n}d^{+}_{n,\sigma}|0\rangle$ is the wave function of the second strand. Inserting Eq. (1) into the Schrödinger equation, we obtain

$$\begin{aligned}
&t_{n-1,n}\phi_{n-1,\sigma} + (\varepsilon_n - E)\phi_{n,\sigma} \\
&+\lambda_n\varphi_{n,\sigma} + t_{n,n+1}\phi_{n+1,\sigma} = 0, \\
&h_{n-1,n}\varphi_{n-1,\sigma} + (u_n - E)\varphi_{n,\sigma} \\
&+\lambda_n\phi_{n,\sigma} + h_{n,n+1}\varphi_{n+1,\sigma} = 0; \qquad (2)
\end{aligned}$$

especially at the right end of the DNA chain, the ferromagnetic electrodes with the moment at the angles θ_1 and θ_2 to the z-axis will result in spin flip

$$\begin{aligned}
&t_{N,N+1}\phi_{N,\sigma} + (\varepsilon_{k,\mathrm{R}} + \sigma M\cos\theta_1 - E)\phi_{N+1,\sigma} \\
&+M\sin\theta_1\phi_{N+1,\overline{\sigma}} + t_{N+1,N+2}\phi_{N+2,\sigma} = 0, \\
&h_{N,N+1}\varphi_{N,\sigma} + (\varepsilon_{k,\mathrm{R}} + \sigma M\cos\theta_2 - E)\varphi_{N+1,\sigma} \\
&+M\sin\theta_2\varphi_{N+1,\overline{\sigma}} + h_{N+1,N+2}\varphi_{N+2,\sigma} = 0. \qquad (3)
\end{aligned}$$

Note that in the input electrodes 1 and 2, the wave functions are superposition of the incoming plane wave of unit amplitude and a reflected wave with amplitudes r_{11} and r_{22} , while in the output electrodes 3 and 4, the outgoing plane waves possess transmission amplitudes t_3 and t_4, respectively.

$$\begin{aligned}
\Psi_{\mathrm{L}} &= \sum_{i=1}^{2}(\,\mathrm{e}^{\mathrm{i}\,K^{\mathrm{L}}_{i,\sigma}na} + r_{ii}\,\mathrm{e}^{-\mathrm{i}\,K^{\mathrm{L}}_{i,\sigma}na}), n < 0; \\
\Psi_{\mathrm{R}} &= [t_3(\sigma)\,\mathrm{e}^{\mathrm{i}\,K^{\mathrm{R}}_{i,\sigma}na} + t_4(\sigma)\,\mathrm{e}^{\mathrm{i}\,K^{\mathrm{R}}_{i,\sigma}na}], n > N.
\end{aligned}$$

Using these functions, we can rewrite Eqs. (2) and (3) as the matrix equations

$$\begin{aligned}
&(\phi_{n+1,\sigma}, \varphi_{n+1,\sigma}, \phi_{n,\sigma}, \varphi_{n,\sigma}, \\
&\phi_{n+1,\overline{\sigma}}, \varphi_{n+1,\overline{\sigma}}, \phi_{n,\overline{\sigma}}, \varphi_{n,\overline{\sigma}})^{\mathrm{T}} \\
&= M(n,E)(\phi_{n,\sigma}, \varphi_{n,\sigma}, \phi_{n-1,\sigma}, \\
&\varphi_{n-1,\sigma}, \phi_{n,\overline{\sigma}}, \varphi_{n,\overline{\sigma}}, \phi_{n-1,\overline{\sigma}}, \varphi_{n-1,\overline{\sigma}})^{\mathrm{T}}, \qquad (4)
\end{aligned}$$

where $\boldsymbol{M}(n,E)$ is called the transfer matrix,[18,31,32] which links the coefficient vector $(\phi_{n,\sigma},\ \varphi_{n,\sigma},\ \phi_{n-1,\sigma},\ \varphi_{n-1,\sigma},\ \phi_{n,\overline{\sigma}},\ \varphi_{n,\overline{\sigma}},\ \phi_{n-1,\overline{\sigma}},\ \varphi_{n-1,\overline{\sigma}})^{\mathrm{T}}$ to the vector $(\phi_{n+1,\sigma},\ \varphi_{n+1,\sigma},\ \phi_{n,\sigma},\ \varphi_{n,\sigma}, \phi_{n+1,\overline{\sigma}},\ \varphi_{n+1,\overline{\sigma}},\ \phi_{n,\overline{\sigma}},\ \varphi_{n,\overline{\sigma}})^{\mathrm{T}}$, defined by

$$\boldsymbol{M}(n,E) = \begin{bmatrix} \boldsymbol{M}_{11} & \boldsymbol{M}_{12} \\ \boldsymbol{M}_{21} & \boldsymbol{M}_{22} \end{bmatrix},$$

with

$$\boldsymbol{M}_{11} = \begin{bmatrix} \dfrac{E-\varepsilon_n}{t_{n,n+1}} & -\dfrac{\lambda_n}{t_{n,n+1}} & -\dfrac{t_{n-1,n}}{t_{n,n+1}} & 0 \\ -\dfrac{\lambda_n}{h_{n,n+1}} & \dfrac{E-u_n}{h_{n,n+1}} & 0 & -\dfrac{h_{n-1,n}}{h_{n,n+1}} \\ 1 & 0 & 0 & 0 \\ 0 & 1 & 0 & 0 \end{bmatrix}$$

describing the electron with spin index σ transport in the DNA lane, and $\boldsymbol{M}_{22}$ describing the electron with spin index $-\sigma$ in the similar form. In DNA region, $\boldsymbol{M}_{12}$ and $\boldsymbol{M}_{21}$ are zero matrix, but when it comes to the ferromagnetic leads, they will have the form

$$M_{12} = \begin{bmatrix} \dfrac{M\sin\theta_1}{t_{N+1,N+2}} & 0 & 0\ 0 \\ 0 & \dfrac{M\sin\theta_2}{h_{N+1,N+2}} & 0\ 0 \\ 0 & 0 & 0\ 0 \\ 0 & 0 & 0\ 0 \end{bmatrix} = M_{21},$$

which will lead to the spin-flip in the right ferromagnetic leads. Then we can write the inside wave function at the left ends and the transmission function at the right ends in one matrix equation

$$\Psi_{\mathrm{R}} = M^{N}(n,E)\Psi_{\mathrm{L}}, \qquad (5)$$

where N is the number of the transfer matrix, i.e., the base pairs of the DNA, or the length of DNA.

Using Eq. (4), we can obtain the transmission $T(E,\sigma)$ as

$$\begin{aligned} T(E,\sigma) &= T_3(E,\sigma) + T_4(E,\sigma) \\ &= |t_3|^2(\sigma) + |t_4|^2(\sigma). \end{aligned} \tag{6}$$

To obtain the transmission of the charge and spin from the donor at the left-end site to the acceptor at the right-end site, the chemical potential at each end is necessary, according to the Giese's results, the chemical potential at the left ends is approximately equal to the on-site energy of the DNA line, and the chemical potential at the right ends is smaller than the former.

3. Calculation and results

In terms of Eqs. (5) and (6), we calculate the transmission probability through the DNA duplex as we have shown in Fig. 1, the HOMO energies for bases in the DNA G, C, T, A are $E_G = 7.75$ eV, $E_C = 8.87$ eV, $E_T = 9.14$ eV, $E_A = 8.24$ eV.[5,31,33] In the actual calculation, we assume that all the intrastrand coupling parameters have a fixed value as $t_{n,n+1} = h_{n,n+1} = t_{\rm d}$ $(1 \leqslant n \leqslant N-1)$, and the interstrand coupling parameter is set to be $\lambda_n = \lambda_{\rm d}$ $(1 \leqslant n \leqslant N)$ between the neighbouring bases in the DNA molecule.

First, we consider a simple limit: the interstrand coupling will be cancelled, i.e., $\lambda_{\rm d} = 0$. Then our model will contain two independent quantum dot lines. We calculate the transmission $T_3\ (E,\sigma)$ for the first brand as shown in Fig. 2(a) and $T_4\ (E,\sigma)$ for the second brand as shown in Fig. 2(b) under different intrastrand coupling parameters. The results have shown that when the interstrand coupling is switched off, the transmission probability of the spin will drop sharply with the number of the DNA base pairs N. In our calculation, the vertical coordinate is in logarithm of the transmission probability, so we can say that the transmission damping exponentially with the length of the line, spin and charge transport in such a formation has a very low efficiency. It also indicates that the intrastrand coupling parameter $t_{\rm d}$ could enhance the transport efficiency. In our Hamiltonian, $t_{\rm d}$ is the hopping integral between the nearest neighbouring site in the line, the increase of it means a stronger interaction between the neighbouring sites, and this will help the charge and the spin transport from n to $n+1$ site.

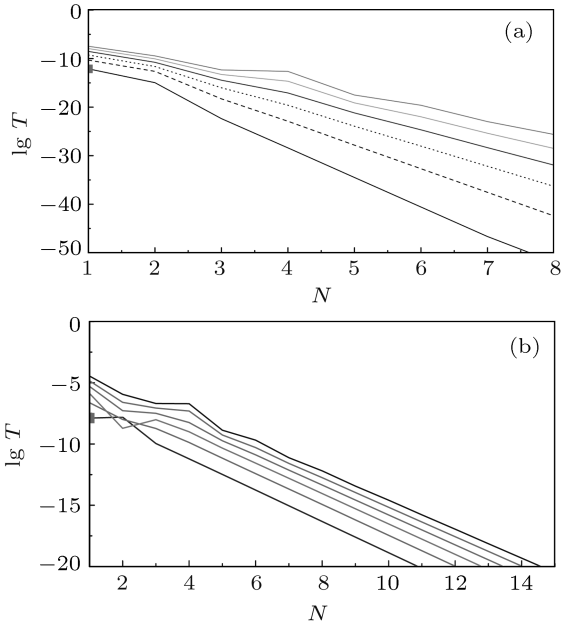


Fig. 2. Transmission for (a) the first brand and (b) the second brand for $\lambda_{\rm d} = 0$.

Then, we fix the intrastrand coupling parameter as a constant $t_{\rm d} = 0.5$ eV. Figure 3 shows the result of the transmission versus N for different interstrand coupling, with $\lambda_{\rm d} = 0$, 0.01, 0.05, 0.1 eV from lower to upper curves. For a weak interstrand interaction, $\lambda_{\rm d}$ is not stronger than $t_{\rm d}$. The calculation illustrates that the interstrand coupling can also increase the transporting efficiency. The lowest curve is for the case of $\lambda_{\rm d} = 0$, the transmission probability decreases exponentially as shown in Fig. 2, other curves are obtained under the condition of nonzero interstrand coupling. It is significant to point out that when the number of the DNA base pairs N is no more than three, there are no clear difference among these curves, but when $N > 3$, there exists obvious variation, the curves versus N become more flat and we can define the crossover point $N_{\rm c}$. More calculations indicate that the value of $N_{\rm c}$ depends on the strength of the interstrand coupling parameter $\lambda_{\rm d}$ partly, smaller $\lambda_{\rm d}$ corresponds to bigger $N_{\rm c}$, and smaller $N_{\rm c}$ for stronger coupling. The distance-dependent crossover arises from the topological variation of the model: when we switch off the interstrand coupling, the model in Fig. 1 will be simplified to two independent QD lines. This is a typical one-dimensional model, charge and spin transporting in terms of this model means tunneling through N barriers. This will lead to the exponential damping of transmission probability. When we switch on the interstrand coupling, the model turns to a two-dimensional ladder network and the interstrand cou-

pling interaction between the bases T and A will provide an extra superexchange channel for the charge and spin. For example, if we consider the first chain of the DNA, there is only one channel when there is no-interstrand coupling, and when there exists one base pair in the duplex, the number of the channel is still one. Then, when two base pairs exist, the charge and spin will have two channels from the left electrode to the right electrode as we have shown in Figs. 4(a) and 4(b). When there exist N base pairs, a new base pair will present $N-1$ extrachannels for the spin and the charge, then the total channels will be the sum from 1 to $N-1$ plus 1 (Fig. 4(c)).

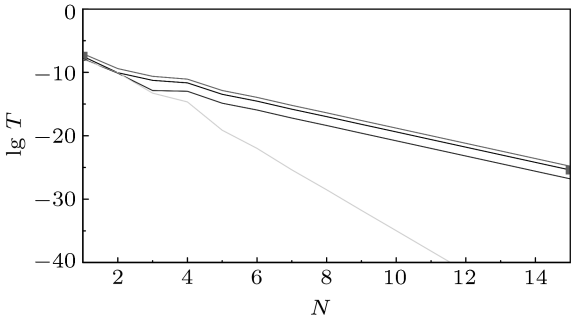


Fig. 3. Transmission versus N for different interstrand couplings.

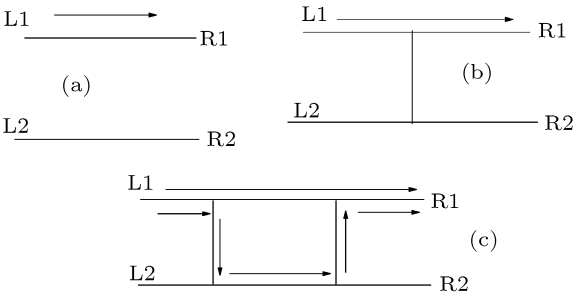


Fig. 4. Schematic for carrier transport through the DNA molecule: (a) two separate lain, (b) one pair in DNA, (c) two pairs in DNA.

Figure 5 is the result of the net transmission probability defined by $\Delta T = T(E,\sigma) - T(E,\bar{\sigma})$ versus the number of the base pairs N. The lower dashed line is for the first chain of the duplex and the upper solid line is for the second one. When ΔT is positive, it means the electrons with spin-up property is predominant in the output electrons. Calculation also indicates that we can obtain different spin currents at these two right electrodes for different on-site energies and angles θ_1 and θ_2. In some suitable regions, we can have pure spin-current. When we obtain the standard spectrum, by comparing the test spectrum with the standard one, we can make a verdict of the change of the corresponding parameters such as the on-site energy and the hopping integral. In Figs. 6(a), 6(b), 6(c), and 6(d), we fix the number of the base pairs as $N = 30$, 1, 2, 7,[34] and give the transmission probability T as a function of the electronic energy E. The efficiency of the spin transport will decrease sharply, this is obviously different from the previous theory as we have proposed in Section 1, where $N = 3$ is the crossover point in the DNA duplex, when $N > 3$, the transmission will be weakly distance-dependent on the length of the molecule, and our results demonstrate that the transmission probability exponentially decays as a function of the number N. It strongly depends on the distance even for $N > 3$ and the crossover point is not a constant, it depends on the intrastrand and interstrand coupling, which is consistent with the quantum tunneling barrier.

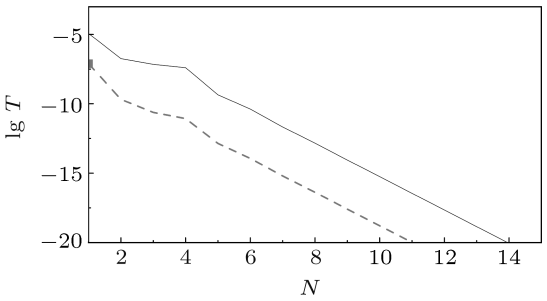


Fig. 5. Results for the net transmission probability, the dashed line is for the first brand and the solid line is for the other.

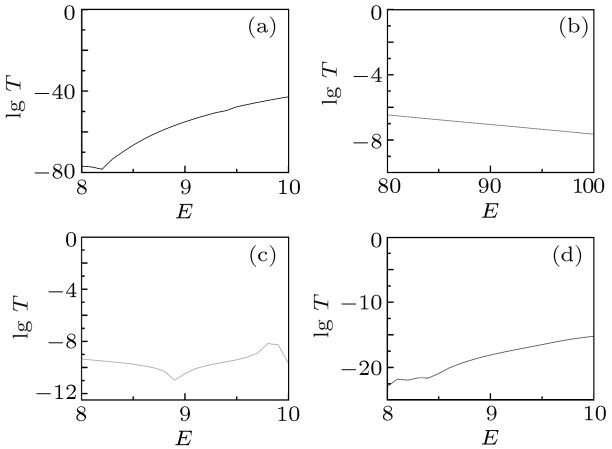


Fig. 6. Transmission versus the electron energy: (a) $N = 30$, (b) $N = 1$, (c) $N = 2$, (d) $N = 7$.

4. Conclusion

Using the TBA model and the Green function technique, we have explored the charge and spin trans-

fer through a DNA type molecule. The distance-dependent transport shows a crossover point N_c when the transmission probability is a function of the base pairs number. N_c is dependent on the interstrand and intrastrand coupling interaction between the bases in the duplex. The transmission probability decays exponentially with the length of the molecule; when $N < N_c$, the curve drops very sharply and when $N > N_c$, the curve will be more flat, i.e., the superexchange between the base pairs will help the transfer of the charge and spin, stronger interstrand coupling will lead to higher transfer efficiency and lower crossover point (temperature). The distance-dependent crossover is based on the topological variation from the one-dimensional to the two-dimensional model as we switch on the interstrand coupling. When there exist N base pairs, a new base pair will present $N-1$ extrachannels for the spin and the charge, long transfer distance means more base pairs in the duplex and more extra channels will be present. This makes the decay of the transmission more slowly than the one-dimensional model and will improve the stability of the quantum transport. By comparing the test spectrum with the standard spectrum, we can make a verdict of the change in the on-site energy and the hopping integral. Calculation also indicates that the intrastrand coupling could enhance the transmission. Spin and charge transfer through DNA molecule is consistent with the quantum tunneling barrier, and we can obtain different spin currents under suitable parameters for these two right leads. This means that the brand of the DNA can transfer different spin components to the corresponding end.

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