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Temperature dependence of transport behavior of a short DNA molecule

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Abstract

DNA molecules may work as novel devices due to their interesting electronic transport properties. We here propose a theoretical method to deal with the temperature dependence of the transport behavior of a short DNA molecule, taking into account Coulomb interaction of electrons and the coupling between electrons and the two-level system in the DNA molecule. The nonlinear current–voltage curves are derived by using the Landauer formulae. We find that the voltage gap of the current–voltage curves is sensitive to the parameters of the two-level system. We also find that Coulomb blockade peaks can be controlled by varying the temperature, which relates to particular features of the DNA molecule.

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Owing to the advance in molecular electronics, DNA is expected to become one of the promising molecular-wire candidates. Recently, poly(G)-poly(C) DNA has exhibited large-bandgap semiconducting behavior in electronic transport [1–3] and these experimental observations have been explained in theoretic studies [4,5]. However, understanding of the strong temperature dependence of the gap has been elusive up to now [6]. On the other hand, a small DNA molecule that combines the characteristics of typical quantum systems (the Kondo effect and Coulomb blockade) with those of DNA molecules, such as the thermal vibrations of the base clusters, may be used for extremely novel devices. It is therefore interesting to explore possible new effects in electronic transport in few-electron DNA molecules.

There are many factors that influence conductivity for a short DNA molecule with two semi-infinite leads, such as metal work functions, length of the DNA molecule, the role of structure and environment et al. The structural disorder of the DNA plays a significant role in the electronic transport in dry DNA, which comes from thermal vibrations and twist of the base molecules. In principle, inelastic electron–phonon scattering can occur in the presence of thermal vibrations of the base molecules. However, Hjort and Stafström suggested that inelastic scattering has a minor effect on the conductance since the electron–phonon coupling is very weak, but the electronic level shifting due to frozen phonon modes of the base molecules is more important to the conductance [4]. As indicated in the experiments, the phosphate groups carry full negative charges on the backbone, the root-mean-square vibrational displacement of a base pair in DNA at room temperature is estimated to be about 0.3–0.4 Å [7], which is an order of magnitude higher than in crystals at room temperature, thus the electronic level gets split depending on the particular charge polarization. Lee et al. suggest that poly(G)-poly(C) DNA has the characteristic behavior of a *p*-type semiconductor using oxygen adsorption experiments [8]. Furthermore, fluorescence polarization anisotropy have been one of the most common methods for studying short-time DNA dynamics [9]. The method relies on a probe molecule that has a large change in dipole moment in the excited state. The probe in the interior of the helix is sensitive to the motion of many nearby charged groups. Hence, we propose that the vibrations and twists of the base molecules induce dipole moments that play an essential role for electronic transport. In the pioneering work of Cuniberti et al. [10] this dipole viewpoint has been put

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forward by considering the hybridization of the π stack with the backbone states.

In this paper we investigate the temperature dependence of the transport behavior for electrons through a small DNA molecule. A two-level system is adopted to describe the two polar states which come from frozen vibrations and twist modes in the DNA molecule. We concentrate on the effect of coupling between the electron and the TLS, and assume an electron having short-range interaction with a single TLS at each base pair. We adopt the equivalent multi-channel network (EMCN) method [11] to investigate the transport properties of the system. The advantage of the method is the description of the tunneling electrons in terms of the combination of many-body states within the DNA molecule and single-electron states in the leads. From the many-body description within the molecule, Coulomb blockade and the coupling between electrons and TLS can be properly considered, separately from the single-electron states in the leads. The transport properties can be then easily calculated by the Landauer formulae, since the conduction electrons interact with the TLS by electrostatic interaction.

We consider that a DNA molecule consists of N base pairs (double-stranded poly(G)-poly(C)) with an effective TLS coupled to each base pair, and ignore the interaction among the TLS's. The Hamiltonian of the isolated DNA molecule can be written as $H_M = H_{\mathrm{GC}} + \sum_j H_{\mathrm{TLS}}^{(j)} + \sum_j H_{\mathrm{in}}^{(j)}$. H_{GC} describes the motion of electrons in the DNA molecule, and can be written as $H_{\mathrm{GC}} = \sum_{\sigma,j=1}^{N} (t_j c_{j+1,\sigma}^{\dagger} c_{j,\sigma} + \mathrm{H.c.}) + \sum_{\sigma,j=1}^{N} \varepsilon_0 c_{j,\sigma}^{\dagger} c_{j,\sigma}$, where $c_{j,\sigma}$ is the annihilation operator of electrons with spin σ in the jth GC pair, t_j is the hopping integral of π orbitals between two nearest-neighbor GC pairs, and ε_0 is the energy level controlled by the applied gate potential. $H_{\mathrm{TLS}}^{(j)}$ is the coupling energy between the TLS and the jth GC pair, which can be expressed in terms of two states of vibrations $|j_1\rangle$ and $|j_2\rangle$, $H_{\mathrm{TLS}}^{(j)} = \Delta_0(|j_1\rangle\langle j_1| - |j_2\rangle\langle j_2|) + \Delta|j_2\rangle\langle j_1| + \Delta^*|j_1\rangle\langle j_2|$, where Δ_0 describes the energy splitting between the two states and Δ stands for the overlap. By diagonalization, $H_{\mathrm{TLS}}^{(j)}$ can be rewritten as $H_{\mathrm{TLS}}^{(j)} = \sqrt{\Delta_0^2 + \Delta^2}(|j_+\rangle\langle j_+| - |j_-\rangle\langle j_-|)$, where $|j_+\rangle = \mu|j_1\rangle + \gamma|j_2\rangle$ and $|j_-\rangle = \gamma|j_1\rangle - \mu|j_2\rangle$, with $\mu = \left(\frac{1+\Delta_0/(\Delta_0^2+\Delta^2)^{1/2}}{2}\right)^{1/2}$, $\gamma = \left(\frac{1-\Delta_0/(\Delta_0^2+\Delta^2)^{1/2}}{2}\right)^{1/2}$. The coupling energy between the TLS and the electrons is $H_{\mathrm{in}}^{(j)} = g c_{j,\sigma}^{\dagger} c_{j,\sigma} \cdot (|j_1\rangle\langle j_1| - |j_2\rangle\langle j_2|)$, where g is the strength of the coupling. With the diagonalized picture of TLS, the distribution probability in equilibrium of the TLS can be written as $F_{\mathrm{TLS}}^{(\pm)}(T) = \frac{1}{Z}\exp\left(\mp\frac{\sqrt{\Delta_0^2+\Delta^2}}{k_B T}\right)$ with $Z = \exp\left(-\frac{\sqrt{\Delta_0^2+\Delta^2}}{k_B T}\right) + \exp\left(-\frac{-\sqrt{\Delta_0^2+\Delta^2}}{k_B T}\right)$.

Eigenvalues and eigenfunctions of H_M can be obtained by numerical diagonalization. As local excitations in DNA molecules reflect the effects of the electron-TLS coupling, they can be used to calculate the conduction of the system. In actual calculations, the DNA molecule contains 32 GC pairs. The hopping strength between the nearest-neighbor GC pairs $t_j = 0.1$ eV [12]. In the presence of electron-TLS coupling, the parameters used in the calculation are $\Delta_0 = 0.06$ eV, $\Delta = 0.23$ eV, and $g = 0.3$ eV. In Fig. 1, the lowest level E_1 as functions of the parameters of TLS are shown. E_1 increases monotonically with increasing the Δ and g, but decreases rapidly with increasing Δ_0 for relatively small values of Δ_0 and saturates for larger Δ_0. In the experiment of Porath et al. [1], the temperature dependence of the voltage gap in the I–V curves shows a linear relation at relatively high temperatures, but deviates from the linear behavior at lower temperature. In association with the TLS, the values of g, Δ_0, and Δ usually increase with increasing temperature [13]. Thus, in the lower temperature region, the increase of the energy splitting Δ_0 leads to a nonlinear relation of the voltage gap versus the temperature. On the other hand, Δ and g become dominant over the temperature dependence of the voltage gap in the higher temperature region, so a linear relation can prevail.

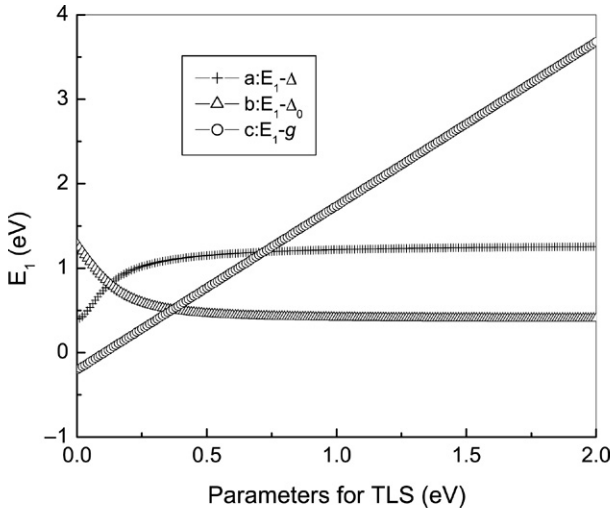

Fig. 1. The lowest energy level as a function of (a) the tunneling strength Δ, (b) the energy splitting Δ_0, and (c) the coupling strength g of the TLS. Other parameters are: (a) $\Delta_0 = 0.06$ eV, $g = 0.3$ eV; (b) $\Delta = 0.23$ eV, $g = 0.3$ eV; (c) $\Delta_0 = 0.06$ eV, $\Delta = 0.23$ eV.

Now we turn to calculating the I–V curves for the DNA molecule connecting to two semi-infinite chains. The total Hamiltonian of the system can be written as

$$H = H_{\mathrm{lead}} + H_M + H_T + H_{\mathrm{Coulomb}}, \tag{1}$$

where H_{lead}, H_T, and H_{Coulomb} represent the motion of electrons in noninteracting leads, the tunneling between the DNA molecule and the leads, and Coulomb interaction of electrons within the molecule, respectively. In a tight-binding description for the leads, $H_{\mathrm{lead}} = t_0 \sum_\sigma \sum_{m<-1 \text{ or } m\geq 1} (a_{m,\sigma}^{\dagger} a_{m+1,\sigma} + \mathrm{H.c.}) + \sum_\sigma \sum_{m\leq -1 \text{ or } m\geq 1} \varepsilon_m a_{m,\sigma}^{\dagger} a_{m,\sigma}$, where $a_{m,\sigma}$ is the annihilation operator of an electron at the mth site of the leads, ε_m and t_0 are site energy and nearest-neighbor hopping energy, respectively. With the eigenfunctions, H_M can be rewritten as $H_M = \sum_{\sigma,i=1}^{N} (E_i + V_g) d_{i,\sigma}^{\dagger} d_{i,\sigma}$, where $d_{i,\sigma}$ is the annihilation operator of electrons in the ith eigenfunctions, and