AC Conductance of DNA molecule at low temperature

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Abstract. Using nonequilibrium Green’s function technique, we have investigated the ac conductance of poly(C)-poly(G) DNA molecule at low temperature. Our results indicate that thermal fluctuations of DNA hopping energy will play a profound role for the ac conductance. It smoothes out the conductance curves at high ac signal frequency while does not suppress it substantially. The dependences of the ac conductance on the density of states in the contacts, ac signal frequency and temperature are also investigated.

1. Introduction
The DNA molecule has recently caught much attention for their potential use as nanoelectronic devices, both as templates for assembling nanocircuits and as basic elements of such circuits¹. There are several properties distinguishing DNA from other nanoelectronic materials. First, the basic pairs of DNA follow a strict pairing rule: adenine always pairs with thymine, while cytosine always pairs with guanine. This behavior, combined with self-assembly property of DNA can be used to build complex nanowire geometrical arrangements. Furthermore, the interbase hybridization of \(\pi\) orbital in neighboring nucleobases is believed to lead to conducting behavior.

If DNA is ever going to be used as components of electronic circuitry, theoretical knowledge on ac conductance of DNA molecule is essential. In this work, we present calculations for ac conductance of poly(C)-poly(G) DNA molecule by using nonequilibrium Green’s function technique. Our model for the DNA molecule is based on a simple tight-binding model. It is our purpose to uncover the qualitative and generic transport physics rather than the quantitative details at the present stage. With simple model parameterized by experimental or \textit{ab initio} data, we can get insight into some basic problems, such as how will the finite temperature influence the ac conductance, the problem resulting from the fact that there exists several conflicting effects, such as thermal excitations of carriers and thermal expansion and fluctuations of the bonds between neighboring nucleobases, in the system, and what is the dependence of inductive or capacitive behavior of DNA on Fermi energy and ac signal frequency? In this paper, we try to answer these questions by investigating ac transport through a poly(C)-poly(G) DNA molecule.

2. Model
We consider the following tight-binding Hamiltonian to describe the model DNA molecules²,³:

\[
H = \sum_l [-t_0 \cos \theta_{l,l+1} (c_l^+ c_{l+1} + c_{l+1}^+ c_l) + \varepsilon_l c_l^+ c_l].
\] (1)

Here \(c_l^+\) is the creation operator for an electron at site \(l\); \(\varepsilon_l\) is the on-site energy which depends on the details of the molecule; \(t_0\) is the hopping energy at zero temperature. \(\theta_{l,l+1}\) are twisted angles between...
neighboring nucleobases. These angles are independent random variables following a Gaussian distribution at finite temperature\(^1\): \(<\theta_{i\neq j}> = 0, <\theta_{ji}^2> = T/250\) K. For the on-site energy \(\varepsilon_i\) there are four types of nucleobases, A, G, C, T, with \(\varepsilon_A = 7.75\) eV, \(\varepsilon_G = 8.87\) eV, \(\varepsilon_C = 8.24\) eV and \(\varepsilon_T = 9.14\) eV. In our calculation, \(t_0\) is set to be 1 eV. For simplicity, we further assume the model DNA molecule is connected to the outside world through type-G DNA nucleobase\(^3\). Thereby the surface Green’s functions of the leads are written as follows:

\[
g_a(\varepsilon) = e^{-\varepsilon_G} - 2i \frac{\varepsilon}{|t_G|^2} \frac{\varepsilon - \varepsilon_G}{2|t_G|^2}.
\]

(2)

Thus only in the energy range \(\varepsilon < \varepsilon_G - 2|t_G| \leq \varepsilon \leq \varepsilon_G + 2|t_G|\) will the electron transmission happens. The hopping energy \(t_0\) between electrodes and scattering region is also set to be 1 eV.

We start our calculation of ac current from a general formula for the charge currents at electrode \(\alpha\) in mesoscopic systems\(^4,5\) (let \(\hbar = 1\)):

\[
I_\alpha(\omega) = e \int_{-\infty}^{\infty} d\varepsilon \int_{-\infty}^{\infty} d\varepsilon_g Tr\left[ G'(\varepsilon + \omega, \varepsilon_g) \Sigma^{\alpha}_{\omega}(\varepsilon, \varepsilon_g) + G^<(\varepsilon + \omega, \varepsilon_g) \Sigma^{\alpha}_{\omega}(\varepsilon, \varepsilon_g) \right] - \Sigma^{\alpha}_{\omega}(\varepsilon + \omega, \varepsilon_g) G^{\alpha \dagger}(\varepsilon, \varepsilon_g) \Sigma^{\alpha}_{\omega}(\varepsilon + \omega, \varepsilon_g),
\]

(3)

where \(G'(\varepsilon, \omega)\) are the full Green’s functions of the poly(C)-poly(G) DNA molecule, and \(\Sigma^{\alpha}_{\omega}(\varepsilon, \omega)\) are the self-energies of electrode \(\alpha\). For small ac signals, we linearize \(G'(\varepsilon, \omega)\) based on its value at the steady-state, that is, \(G'_{\omega}(\varepsilon, \omega) = G'_{\omega}(\varepsilon + \omega, \varepsilon) \delta(\varepsilon + \omega, \varepsilon)\). Similar treatments are applied to calculate the self-energies, \(\Sigma^{\alpha}_{\omega}(\varepsilon + \omega, \varepsilon) = \Sigma^{\alpha}_{\omega}(\varepsilon, \varepsilon) + \sigma^{\alpha}_{\omega}(\varepsilon + \omega, \varepsilon)\). In previous expressions, \(G\) and \(\Sigma\) at the right-hand side denote the Green’s functions and self-energies at the steady state, while \(g\) and \(\sigma\) denote those caused by the small-signals. Then, we obtain the expression for the charge-current conductance as follows\(^6\):

\[
G_{\omega\omega}(\omega) = -q \int_{-\infty}^{\infty} d\varepsilon \left\{ \Sigma^{\omega}_{\omega} - \Sigma^{\omega}_{\omega} + g^{\omega}_{\omega} \sum^{\omega}_{\omega} + (G'_{\omega} \sigma^{\omega}_{\omega} - \sigma^{\omega}_{\omega} G^{\omega} + G'_{\omega} \sigma^{\omega}_{\omega} - \sigma^{\omega}_{\omega} G^{\omega} ) \right\}.
\]

(4)

Considering the current conservation and the gauge invariance condition, we have to take the displacement currents into account for ac conductance. Following the formalism in [6], we write the full ac conductance as:

\[
G_{\omega\omega}(\omega) = G_{\omega\omega}^{d}(\omega) + G_{\omega\omega}^{c}(\omega),
\]

(5)

where the displacement-current conductance is given by

\[
G_{\omega\omega}^{d}(\omega) = -\sum_{\alpha} G_{\omega\alpha}(\omega) \sum_{\beta} g_{\alpha\beta}(\omega) / \sum_{\nu} g_{\nu\omega}(\omega).
\]

(6)

3. Numerical results and discussions

First we restrict our numerical calculations to the low temperature region since the quantum interference will be destroyed at high temperature. In addition, we have averaged the conductance over a number of different configurations\(^1\). The reason is that measurements are carried out within a finite time interval which potentially samples many structure configurations.

Figure 1 plots the transmission probabilities at different temperatures. The solid lines are obtained by averaging over 100,000 configurations with thermal fluctuations of the hopping energy, while the dash lines obtained by neglecting the thermally fluctuations. In the latter case, we replace \(t_0 \cos(\theta_t)\) with the root-mean-square value \(t_0 \sqrt{<\theta_t^2>}\) at that temperature. It is quite clear that there exists an energy gap between \(\varepsilon_G = 7.75\) eV and \(\varepsilon_C = 8.87\) eV for the poly(C)-poly(G) DNA molecules.

Another obvious phenomenon is that both the transmission probabilities and the energy range within which the transmission occurs shrink quickly with the increasing temperature. By comparing the transmission curves with and without thermal fluctuations of the hopping energy, we can see that the shifts of the transmission peaks are due to the smaller mean hopping energy between neighboring nucleobases at higher temperature, while thermal fluctuations will only cause the suppression of the transmission amplitudes.
However, it is quite interesting to note that for ac transport thermal fluctuations of the hopping energy (round-symbol lines) will not suppress the full ac conductance $G_{LR}$ at higher frequency. Instead, the average of thermal fluctuations will just smooth out the oscillating conductance curves (solid lines), as is seen in figure 2. Careful analysis indicates that this behavior originates from the quite different densities of states (DOS) in the molecule due to thermal fluctuations.

From figure 2, we also find that for input ac signals with very low frequencies, the imaginary parts of the ac conductance, $\text{Im}(G_{LR})$ and $\text{Im}(G_{cLR})$, quickly go to zero. It means that there is neither capacitive nor inductive behavior of the conductance. This behavior is as expected for very low frequency ac transport.

**Figure 3.** Conductance versus Fermi energy at $T=0$ K for various ac signal frequencies.

Figure 3 plots the ac conductance versus Fermi energy for several ac signal frequencies at $T=0$ K. We can see that at small ac signal frequency ($\hbar \omega = 10^{-6}$ eV) the real part of charge conductance $G_{cLR}$ is nearly the same as that of total conductance $G_{LR}$, while the imaginary part of $G_{LR}$ is quite small compared with the real part. These behaviors are quite reasonable since the ac conductance at low frequency limit should approach the dc case. At higher ac signal frequency ($\hbar \omega = 10^{-2}$ eV), we can see from figure 3(b) that the real part of $G_{LR}$ differs much more obviously from that of $G_{cLR}$. Meanwhile, the imaginary parts of the conductance become much larger, which denotes clear inductive or capacitive behavior.
In figure 4, the ac conductance versus Fermi energy at finite temperature is plotted. We can see that there is no sharp peak in $G_{LR}$ or $G'_{LR}$ versus Fermi energy curves. Besides, by comparing the conductance curves obtained with (symbol lines) and without thermal fluctuations (solid lines), we can see at this ac frequency, the amplitude of the conductance is strongly suppressed by the thermal fluctuations.

We have also investigated the temperature dependence of the ac conductance, as in figure 5. If thermal fluctuations of the hopping energy (solid lines) are neglected, the dependence of $G_{LR}$ and $G'_{LR}$ on temperature are mainly determined by two conflicting thermal effects: the thermal excitation of electrons within several $k_B T$ around the Fermi energy, and the thermal expansion of the twist angles $\theta_{l,l+1}$ between neighboring nucleobases. The former will enhance the conductance as the temperature increases, while the latter will suppress it due to the shrink of hopping energies. In our interested temperature range ($T \leq 120$ K), the conductance is increasing with the temperature at the beginning, then, it seems to become saturated at higher temperature. This implies the latter effect will dominate at higher temperature. On the other side, if thermal fluctuations of the hopping energy are taken into account, the low-frequency ac conductance gets significantly suppressed by this effect after $T$ goes beyond 25 K.

4. Conclusion

In summary, we have demonstrated the ac conductance of poly(C)-poly(G) DNA at low temperatures. Our results show that among several thermal effects, thermal fluctuations of the hopping energy play the most prominent role in the ac conductance. Not only do thermal fluctuations enhance the impedance significantly as the temperature increases, they can also invert the inductive behavior to the capacitive behavior at low ac signal frequencies. These results may help us evaluate the future DNA-based molecule devices.

References

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