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Nanoscale capacitance: A quantum tight-binding model

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ABSTRACT

Landauer–Buttiker formalism with the assumption of semi-infinite electrodes as reservoirs has been the standard approach in modeling steady electron transport through nanoscale devices. However, modeling dynamic electron transport properties, especially nanoscale capacitance, is a challenging problem because of dynamic contributions from electrodes, which is neglectable in modeling macroscopic capacitance and mesoscopic conductance. We implement a self-consistent quantum tight-binding model to calculate capacitance of a nano-gap system consisting of an electrode capacitance C' and an effective capacitance C_d of the middle device. From the calculations on a nano-gap made of carbon nanotube with a buckyball therein, we show that when the electrode length increases, the electrode capacitance C' moves up while the effective capacitance C_d converges to a value which is much smaller than the electrode capacitance C'. Our results reveal *the importance of electrodes in modeling nanoscale ac circuits*, and indicate that *the concepts of semi-infinite electrodes and reservoirs well-accepted in the steady electron transport theory may be not applicable in modeling dynamic transport properties.*

1. Introduction

The success of quantum mechanics can be reflected from the development of quantum chemistry and solid state physics. Quantum chemistry uses quantum mechanics to study finite structures like atoms and molecules, while solid state physics applies quantum mechanics to (infinite) bulk materials under periodic boundary conditions. For electronic structure calculations based on the density functional theory [1,2], commercial and academic software packages have been developed and widely used in both quantum chemistry and solid state physics. The development of nanoelectronics, however, introduces a new structure [3,4] consisting of two separated electrodes and a finite device in between [see Fig. 1(a)]. The two electrodes are usually bulk materials and assumed as semi-infinite. Obviously, this transport system is infinite but lacks periodicity. Both the methods from the quantum chemistry (for finite structures) and solid state physics (for bulk materials with a lattice periodicity) cannot be used in this new structure. It was Landauer and Buttiker, who established a standard model for nanoscale direct-current (dc) transport [5]. In their model each electrode is viewed as a reservoir with a given chemical potential, which is unaffected by the central device and the transport current. The difference between the chemical potentials in two electrodes is viewed as the bias voltage. The two-terminal conductance is then determined by the transmission of electrons from one electrode to the other.

Recently, this two-terminal transport system was proposed to detect the DNA sequence [6], where the middle device is taken as different DNA nucleotides [adenine (A), cytosine (C), guanine (G), and thymine (T)]. This proposal relies on the nucleotide-resolved conductance, which, however, depends exponentially on the effective distances between the nucleotide and electrodes [7]. To wash out this distance dependence of the conductance [8], thousands of measurements and a good statistics might be needed. As an alternative proposal on transport-based DNA sequencing, alternative-current (ac) capacitance measurement [9] may provide a better signal/noise ratio than the dc conductance measurement. The reason is that the capacitance results mainly from direct Coulomb interaction between the nucleotide and electrodes and is thus rather less sensitive to the nucleotide–electrode distance than the dc conductance.

It is believed that ac transport would provide faster and more energy-efficient nanoelectronic devices. Many concepts and algorithms on dc conductance of nanoscale structures have been generalized to calculate dynamic conductance [10–13] and capacitances [14–16]. In these models the transport properties due to the middle device are focused and the electrodes are usually simplified by

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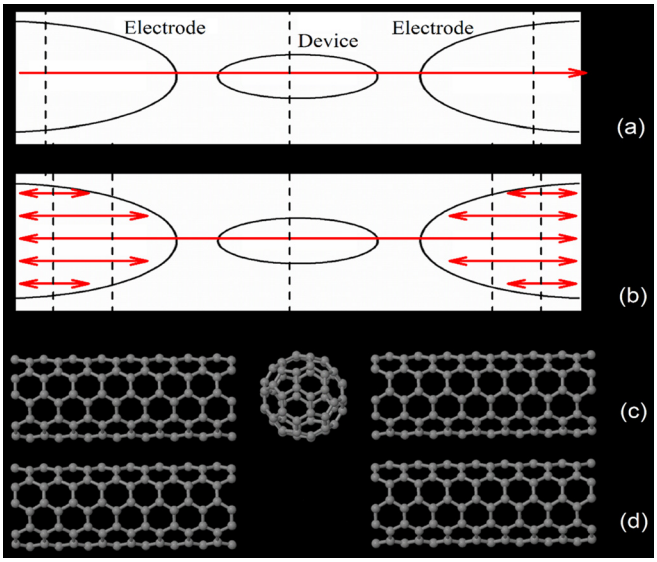

Fig. 1. (Color online.) (a) and (b) A typical two-terminal transport system consisting of two separated electrodes and a device on the nanoscale. The red arrows illustrate space distributions of (a) the steady direct currents and (b) the dynamic alternating currents. (c) and (d) A nano-gap made of carbon nanotubes, with and without a buckyball in between.

taking the *wide-band limit*. However, ac transport is distinct from dc transport even for a simple two-terminal system [see Figs. 1(a) and 1(b)]. In Fig. 1(a), the steady dc currents through any cross-area of the two electrodes are the same. This two-terminal current could be strongly affected by the middle device since it has to go through the middle device due to the current conservation. Thus the I–V curves measured in electrodes may reveal some properties of the middle device. In contrast, an ac current flowing back and forth in electrodes could bypass the middle device [Fig. 1(b)]. This indicates that the ac current amplitude in electrodes can be much bigger than that in the middle small device, for example, when the capacitance of the electrodes is much larger than that of the middle device. The dynamic I–V characters measured in electrodes may be irrelevant to the middle device. When ac transport properties of nanoscale circuits are concerned, we should notice that *the electrodes can contribute dominantly to the current and should be treated carefully*. To extract the dynamic properties of the middle device from the transport measurements, it is necessary to remove properly the contribution from the electrodes.

To illustrate the role of electrodes in determining physical quantities related to ac transport, in this work we calculate capacitances of a nano-gap system as shown in Fig. 1(b) and define effective capacitances for the nanoscale device therein. The capacitances of the electrodes and device will play an important role in determining dynamic current in ac transport, but will not affect steady current in dc transport. The nano-gap system consists of two metallic electrodes at a distance of several nanometers. We adopt both a classical atomic charge model [17,18] and a quantum tight-binding model to calculate the static capacitance. For a nano-gap [shown in Figs. 1(c) and 1(d)] made of carbon nanotubes (CNTs), our calculations based on both models show that the inclusion of a nanoscale device like a buckyball (C_{60}) brings only a tiny change on the capacitance of the system. This indicates that when connecting in a nanoscale circuit, it is the electrodes rather than the middle device will contribute mainly to dynamic currents.

2. Models on capacitances of nano-gap systems

To calculate the capacitance of a nano-gap system schematically shown in Fig. 1(c), for each electrode we take a finite length N_C which starts from the electrode's terminal [19]. The length N_C is in unit of the size of a unit cell of electrodes and will be gradually increased in calculations so that each electrode can approach the corresponding semi-infinite electrode. The whole system $\boldsymbol{H_{2E+D}}$ in calculations is finite and consisting of two finite electrodes and a middle device. We assign a positive charge $+Q$ on the left electrode and a negative charge $-Q$ on the right one. The potential difference ΔV between the two electrodes can be calculated and the capacitance of this finite system is defined as $C = Q/\Delta V$. For the finite system $\boldsymbol{H_{2E}}$ including only the two electrodes, as shown in Fig. 1(d), we can similarly calculate its capacitance from $C' = Q/\Delta V'$. Then the contribution from the middle device can be calculated by $C_d = C - C'$ and defined as the effective capacitance of the device [20]. As shown below, the value of C_d at large length N_C is a constant and thus can be viewed as an intrinsic capacitance contribution of the middle device.

To calculate the potential difference ΔV, here we adopt a self-consistent tight-binding model. The finite system $\boldsymbol{H_{2E+D}}$ can be described by the Hamiltonian

$$H = \sum_{i=1}^{N}(E_i + V_i)C_i^+ C_i - \sum_{\langle i,j\rangle} t_{i,j}C_i^+ C_j \tag{1}$$

Here N is the total number of atoms, C_i is the annihilation operator of electrons on atom i, E_i and V_i are the zero-field onsite energy and electric potential energy at atom i, and $t_{i,j}$ is the hopping energy from atom j to atom i. Note that there is no hopping between atoms belonging to different parts (left electrode, right electrode, and device). We assume that the tight-binding parameters $t_{i,j}$ and E_i depend only on the materials. The electric potential energies V_i are solved in a self-consistent manner. For the initial guess values of electric potential energies, we use the results from classical calculations as described in the classical model below. We then diagonalize the Hamiltonian (1) to yield a set of energy levels and corresponding wave functions. For each electrode, the energy levels are occupied by electrons until reaching the assigned charge. From the wave functions of occupied energy levels, we can calculate the charge q_j of each atom j for the whole system. The charge distribution of each atom is then approximated by a Gaussian distribution. The charge density $\rho_j(\mathbf{r})$ of atom j is $\rho(\mathbf{r}; q_j) = \frac{q_j}{\pi^{3/2}R^3}\exp[-(\frac{|\mathbf{r}-\mathbf{r}_j|}{R})^2]$, where $\mathbf{r}_j$ is the position of the atom j and R is the width of the distributions. Then a new set of values of the electric potential energies can be calculated by $V_i = -e(\sum_{j=1}^{N} T_{ij}q_j + \chi_i)$, and be used as the new guess V_i in the Hamiltonian (1). Here e is the proton's charge, $T_{ij} = \mathrm{erf}(r_{ij}/\sqrt{2}R)/(4\pi\epsilon_0 r_{ij})$, and χ_i is the electron affinity of atom i. This procedure is repeated until a self-consistent solution of electric potential energies V_i is reached. Finally, one obtains the potential difference between the two electrodes $\Delta V = |\varepsilon_{maxL} - \varepsilon_{maxR}|/e$, where $\varepsilon_{maxL}(\varepsilon_{maxR})$ is the energy of highest occupied energy levels in the left (right) electrode.

We have also present results from a classical model for comparison. The classical charge q_j of the atom j satisfies the following equations:

$$\begin{cases} \sum_{j=1}^{N} T_{ij}q_j + \chi_i - V_L = 0; \quad i \in L \\ \sum_{j=1}^{N} T_{ij}q_j + \chi_i - V_R = 0; \quad i \in R \\ \sum_{j\in L} q_j = +Q\,; \\ \sum_{j\in R} q_j = -Q\,, \end{cases}$$