

Theoretical study of current fluctuation in multi-contact molecular bridge systems

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Abstract

Current fluctuation caused by the multi-contact to electrode was studied for molecular bridge device system using the density-matrix simulation. It was shown that the correlation between contacts remarkably changes the magnitude of steady-state current and the response time of transient current to applied voltage. These results indicate that the control of electrode contacts is essential to realize the steady device operation.

1. Introduction

Molecular bridge system is the smallest electronic device in Nature, where a single molecule is sandwiched between metallic electrodes. Electron transport properties have intensively been studied in this decade for a number of molecular bridge systems and unique features such as enormous loop/transient currents have been elucidated [1,2].

In most of previous studies, the contact of molecule to respective electrode was assumed realized by a single chemical bond. However, since molecules often have wide shapes, the connection of the molecule to electrodes has a variety of structures as shown in Fig. 1(a), even to a single electrode, and such variety induces the fluctuation of current characteristics. To use molecular bridge systems as real devices, it is important to clarify such fluctuations. In this work, we study what fluctuations are observed in steady and transient currents of molecular bridge systems when the molecule is connected to electrode with plural number of chemical bonds, by using theoretical simulation.

2. Model and method of calculation

To simplify the multi-contact molecular bridge, we use the two-site model-molecule system shown in Fig. 1(b), where the molecule is connected to left source electrode with two bonds with transfer energies, v_{A1} and v_{B2} , while connected to right drain electrode with a transfer energy of

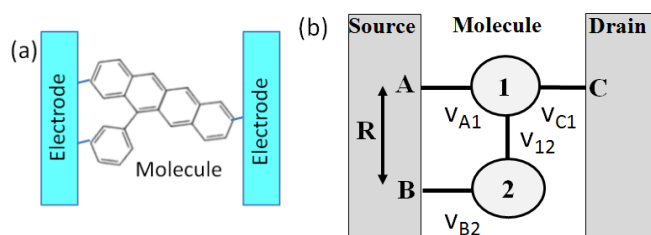


Fig.1. (a) Schematic picture of multi-contact molecular bridge system. (b) Model system adopted in this work. v_{A1} , v_{B2} , v_{C1} , and v_{12} are transfer energies. Molecule is connected to left electrode at A and B points separated by the distance, R .

v_{C1} . v_{12} is the intra-molecule transfer energy, which is often much larger than the other v 's.

Time evolution of electronic current through this molecular bridge system is calculated based on the following quantum Boltzmann equation for the density matrix of molecule, $\sigma(t)$, [1].

$$\frac{\partial \sigma(t)}{\partial t} = - \sum_{X,Y} \sum_{u,w} v_{uX} v_{wY} \int_0^t dt_1 \times \text{Tr} \left[a_X^\dagger(t) c_u(t), [c_w^\dagger(t_1) a_Y(t_1), \rho_e \sigma(t_1)] \right] + \text{h.c.}$$

Here, $a_X^\dagger(t)$ is the electron creation operator at X point in electrode ($X, Y=A, B$, or C), while $c_u^\dagger(t)$ is that at u point in molecule ($u, w=1$ or 2). Trace is calculated over all the freedoms of left and right electrodes, where we assume that they are in grand-canonical distributions, ρ_e . We solve this equation numerically.

When the A and B contact points is located nearby and after the electron is transferred from A to 1 points, the electron numbers not only at A but also at B points decrease due to the electronic correlation in left electrode. If we assume that the electrode is described by the free-electron system, there appears an electronic correlation between A and B points as

$$\langle a_A^\dagger a_B \rangle = \frac{1}{(2\pi)^d} \int d\mathbf{k} e^{-i\mathbf{k} \cdot \mathbf{R}} f_{\mathbf{k}},$$

where $f_{\mathbf{k}}$ is the Fermi distribution of electrons having the wave-number, \mathbf{k} , d denotes the dimension of electrode ($d=2, 3$), and \mathbf{R} is the distance between A and B contact points. This electronic correlation becomes the largest and zero for $\mathbf{R}=0$ and ∞ , respectively.

3. Results and Discussions

We first show typical transient behavior of the electronic current and the number of electrons in the present molecule

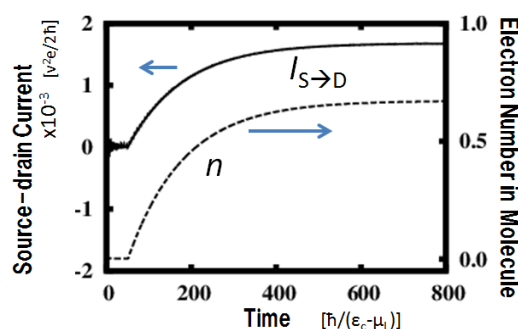


Fig.2. Typical transient behavior of the electronic current and the number of electrons in molecular bridge system.

bridge system in Fig.2. After switching-on the voltage (time=50), the source-drain current and electron number in molecule gradually increase and approach steady values.

Next, we consider how the steady-state current changes with varying the distance, R , between A and B contact points. Figure 3 shows the source-drain current magnitude as a function of the contact distance, R , in cases of two and three-dimensional electrodes. It is seen that the current is considerably small when R is near zero, while it shows the oscillation and approaches the constant value with increasing R . In addition, this oscillation becomes clear as the dimension of electrode decreases.

The oscillation is closely related to the electronic correlation in electrodes. The electrode is characterized by the Fermi wave-number, k_F . Thus, similar to the case of Friedel oscillation, there appears a correlation length, $R_F=1/k_F$, in electrode. Reflecting this R_F , the magnitude of molecular bridge current shows the oscillation behavior with changing the distance R in cases of multi-contacts. Moreover, it is well known that such Fermi-wave-number singularity becomes remarkable as the dimension of the system decreases, which is the reason why larger oscillation of current occurs in case of two-dimensional electrode.

Then, we consider why the current is so small when the distance R between contact points is small. Figure 4(a) shows the currents between molecule sites and between

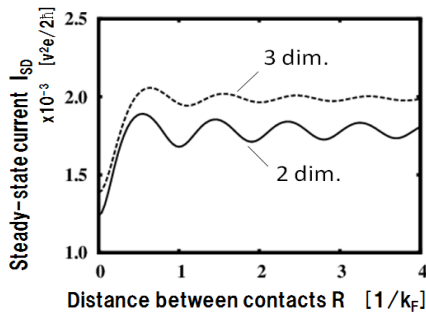


Fig.3. Calculated steady-state source-drain current magnitude as a function of the distance, R , between two contact points, in cases of 2 and 3-dimensional electrodes.

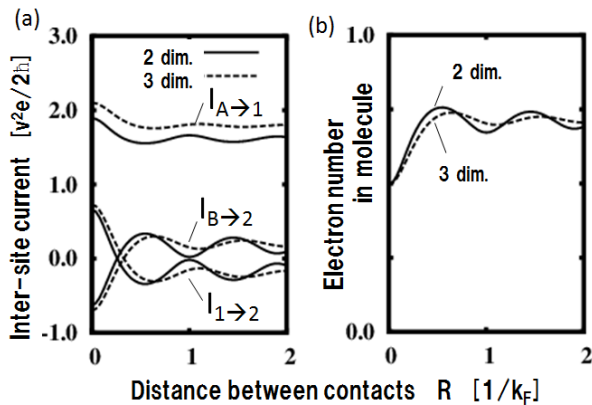


Fig.4. (a) Steady-state inter-site currents and (b) electron number in molecules, as a function of the distance, R , between two contacts, for 2 and 3-dimensional electrodes.

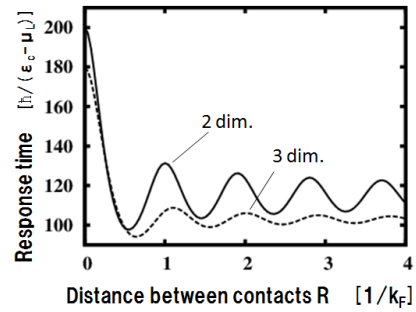


Fig.5. Calculated response time of transient current to applied voltage as a function of the distance, R , between two contact points.

electrode and molecule as a function of the distance R , while Fig. 4(b) shows the electron number in molecule. It is seen that, when R is large, the current flows as $A \rightarrow 1$, $B \rightarrow 2$, $2 \rightarrow 1$, and $1 \rightarrow C$ as normally expected. On the other hand, when R is small, the current directions change as $A \rightarrow 1$, $2 \rightarrow B$, $1 \rightarrow 2$, and $1 \rightarrow C$. This indicates that some of electrons that enters the molecule through A-1 bond go back to the electrode through 2-B bond and there appears a loop current, $A \rightarrow 1 \rightarrow 2 \rightarrow B \rightarrow A$, around the molecule-electrode contact. Correspondingly, the total source-drain current (Fig.3) and the number of electrons in molecule (Fig. 4(b)) become small when the distance R is small. It should be emphasized that the electron correlation (multi-electron coherence) in electrodes promotes these singular phenomena.

Finally, we show how the response time of current to applied voltage changes with varying the distance between contacts, R , the result being displayed in Fig. 5. Due to the correlation in electrodes, we can also see the oscillation and enormous elongation of response time.

4. Conclusions

Current fluctuation in molecular bridge system was studied when the molecule has multi-contacts to electrodes, by the density-matrix quantum calculation. We found that the electronic correlation between two contact points remarkably changes the magnitude of steady-state current and the response time of transient current to applied voltage, reflecting the Fermi-wave-number singularity and the dimensionality of electrodes. These results indicate that the control of molecule-electrode contacts is essential to realize the steady device operation.

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