論文の内容の要旨

論文題目

Theoretical analysis of transient transport behaviors of nanomaterials

(ナノマテリアルにおける過渡的電子輸送現象の理論解析)

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1. Introduction

The rapid progress of nanotechnology now enables us to fabricate ultra-fast electronic devices with increasingly smaller size and higher performance. When the devices go down to nanoscale, quantum effects become obvious, which open new challenges not only in device engineering but also in fundamental science. Thus theoretical investigations of the electrical transport behaviors of nanomaterials, under quantum mechanics framework, is of great significance for giving guidance to experimental measurements of nanodevice characteristics and for realizing novel nanodevices that exploit quantum effects.

Recently, transient transport, especially the current dynamics immediately after applying the bias voltage, has attracted much attention in connection with investigations of high frequency applications of prototype nanoscopic ultrafast electrical devices. An experimental breakthrough is the fabrication of the quantum capacitor, a benchmark system for understanding ultrafast quantum dynamics of nanoscale devices [1]. Theoretical simulation based on a simple model [2] succeeded in reproducing the observed transient current dynamics. However, theoretical study, as well as our understanding, on the nanoscopic transient transport is yet insufficient, because only very simple models have been examined well so far and important effects on it, such as the carrier scattering by phonons and the resultant heating, have hardly been studied. In order to deepen our understanding on the relation between materials properties and their transient transport behaviors, in this dissertation, we aim at clarifying the following aspects closely related to device engineering in transient transport. Firstly, we examine the behaviors of elastic transient electrical currents of multi-level molecular systems. Next, the elastic transient energy currents as well as the energy balance within the nanoscale junctions is examined, which is an important step before calculating the heating effect in the nanojunction. Finally, the electron-phonon interaction is added to the scattering

region and the inelastic phonon scattering effect on transient current as well as the amount of heat generated due to such an effect is examined.

2. Model and method

In our study, the transient transport behaviors of nanostructures have been examined, taking single-level quantum dot and small molecules as examples. These nanostructures are described by the tight-binding Hamiltonians. As shown in Fig. 1, at time $t<0$, the nanostructure is symmetrically coupled to two electrodes, i.e. $\Gamma^L=\Gamma^R$, and the whole system is in the equilibrium state with the Fermi level ε_F set to zero. The bias voltage is applied oppositely on two electrodes at $t=0$ so that $eV^L=-eV^R$. The transient electrical currents as well as the energy currents are calculated using the Nonequilibrium Green's function method within the wide-band-limit approximation.

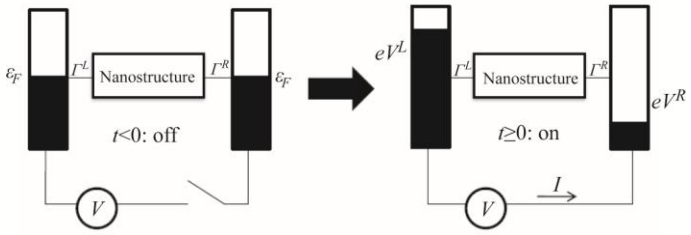

Fig. 1. Model systems of our calculation.

3. Transient current dynamics of multi-level molecular systems

The elastic transient current dynamics of multi-level molecular systems weakly coupled to two electrodes have been investigated. Noticing that the concept of molecular orbital is still valid for small coupling Γ, we show that the total current can be decomposed into nearly-independent current components, each corresponding to an eigen level, in case that no degenerate eigen levels exist in the molecule and the bias voltage is not large enough to excite transitions between different levels. By using such decomposition technique, we have clarified the transient current behavior of a hydrogen molecule (two-level system) and an octatetraene molecule (eight-level system) connected to two electrodes [3]. The transient current of the hydrogen molecular system is characterized by two current components with the same

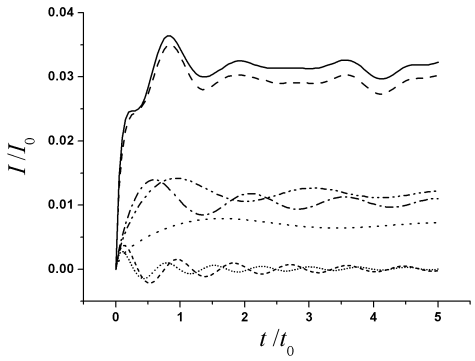

Fig. 2. Total transient current (solid line) and five dominate components (dotted, dash-dot-dotted, dash-dotted, short-dashed, and short-dotted lines) of the octatetraene molecular system. Dashed line denotes the current summation of the five dominant levels. The vertical axis is current scaled by $I_0=0.387\times10^{-4}$A. The horizontal axis is time scaled by $t_0=0.658\times10^{-15}$s, which is also applied to Figs. 3 and 4.

relaxation times and different oscillation periods. On the other hand, the current of octatetraene molecular system is decomposed into eight components. Five single levels, either sharing large coupling strengths or with larger initial electron densities, dominate the total current behavior at the initial stage (Fig. 2).

4. Transient energy transport

The elastic transient energy currents in a single-level quantum dot system have been examined for four different coupling strengths. The results show that the transient energy currents have the same relaxation times and oscillation behaviors as the corresponding electrical currents, and their physical meaning is explained as the power carried by electrons in the deep insides of electrodes. In contrast to the steady state, the transient energy currents in the two electrodes do not balance each other, and their sum reflects the real-time electron redistribution in energy domain in the region between deep insides of two electrodes (Fig. 3). In addition, the amplitude of the energy change, which is the time integral of the sum of energy currents, does not vary monotonically with the coupling strength between the dot and the electrode, in contrast to the relaxation time, which is inversely proportional to the coupling strength. The transfer Hamiltonian, which is the interaction between the quantum dot and the electrode, plays an important role in understanding the transient energy transport. Its time derivative, which is equal to zero in the steady state, is understood as the change of the energies of the electrons in time domain.

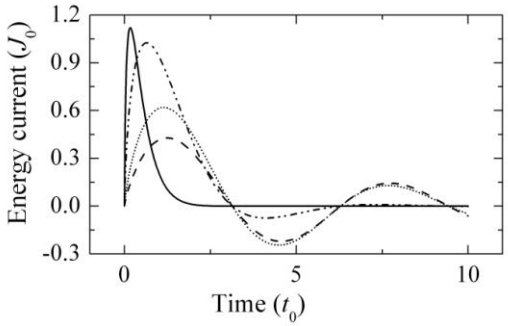

Fig. 3: Time-dependent dynamics of the sum of energy currents in two electrodes for four different coupling strengths: 2.5 eV (solid), 0.5 eV (dash-dot-dotted), 0.1 eV (short-dotted), and 0.05 eV (dashed). The unit of energy current is J_0=0.242×10^{15} eV/s.

5. Inelastic transient transport and phonon heating

We have obtained the time-dependent power transfer between electrons and a single phonon mode in a single-level quantum dot system as the sum of inelastic part of the energy currents in two electrodes, subtracting the change of electron energy due to the time-dependent bias voltage. The subtraction is done using two methods, that is, the extraction of the energy conserved part of electron-phonon scattering term corresponding to the equilibrium state (t<0) and steady state ($t\rightarrow\infty$), respectively. The time-dependent phonon number, which determines the system temperature and the heating effect on inelastic current, is calculated using a phenomenological method employing

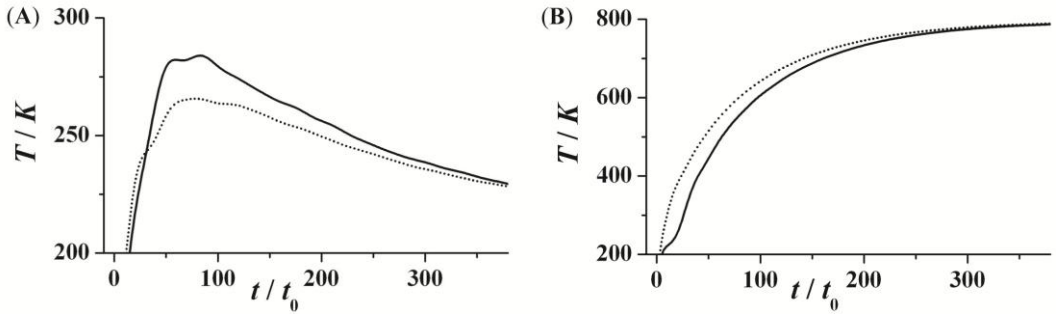

Fig. 4: Real-time temperature of the single-level quantum dot system in case of (A) voltage not larger than the phonon energy and (B) voltage larger than the phonon energy. The solid and dotted line denote the temperature estimated by two methods.

the time-dependent power transfer. We have shown that the two methods provide qualitatively similar dynamical behavior of system temperature, which can be classified into two classes: when the energy corresponding to the applied bias voltage is smaller than or equal to the phonon energy, temperature first increases due to the phonon emission, and then decreases due to the phonon absorption process (Fig. 4(A)); On the other hand in the case in which the energy corresponding to the bias voltage is larger than the phonon energy, temperature increases continuously until reaching its highest value in the steady state (Fig. 4(B)). The total electrical current is suppressed by the phonon heating, while the heat transfer between the dot and environment helps decrease the system temperature and relieve the current suppression.

6. Concluding remark

In the present dissertation, the followings have been achieved on nanoscopic transient transport: (1) the current decomposition method have been proposed, which is useful in understanding the complex behaviors of the transient electrical currents in multi-level molecular systems; (2) the elastic transient energy currents in a single-level quantum dot system have been examined, with keeping the energy conservation of the whole nanocircuit; (3) the real-time phonon heating and its effect on the inelastic electrical current in a single-level quantum dot system has been clarified. We believe that the results obtained in this dissertation provide important guidance to the experimental studies, and expect that they will be confirmed in the near future.

Reference

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[2] K. Sasaoka, et al., Appl. Phys. Lett. **96**, 102105 (2010)

[3]. W. Liu, et al., *J. Appl. Phys.* **109**, 123705 (2011)