Relaxation Process of Transient Current Through Nanoscale Systems; Density Matrix Calculations

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(Received 7 August 2008; Accepted 10 September 2008; Published 25 September 2008)

Transient current behavior through a nanoscale system responding to a sudden voltage application was investigated, based on the Liouville equation of density matrix. It was found that the transient current behavior is characterized with the oscillation and relaxation processes. The oscillation originates from the time-dependent electron transitions between the electrode and nanoscale system, and the period depends on the Fermi energy and the bandwidth of electrodes. On the other hand, the relaxation occurs due to the energy dissipation into a number of electronic states in electrodes, thus, the relaxation time depends on the density of states in electrode and the electron transfer energy between the electrode and nanoscale system. Furthermore, we clarified that the strong electron-electron interaction decreases the relaxation time. [DOI: 10.1380/ejssnt.2008.213]

Keywords: Models of non-equilibrium phenomena; Computer simulations; Electrical transport; Quantum effects; Quantum dots

I. INTRODUCTION

Recent developments in fabrication technology have enabled to produce various nanoscale systems such as semiconductor quantum-point contacts [1], quantum dots [2–4], and molecular bridges [5–7]. Electronic transport through these nanosystems has attracted many interests in these days, and the unique transport properties such as persistent and anisotropic currents, which originate from the quantum interference of electron wavefunctions, have been revealed [8–11]. Most of these studies are concerned with steady-state currents. To employ these systems as practical devices, it is essential to understand the behavior of transient currents when a sudden switching or a pulse voltage is applied to these nanosystems. In fact, real-time measurements of electronic current have been developed recently, where a definite deformation of an input voltage-pulse shape was observed when the rectangular pulse passes through the quantum-point contacts [12, 13]. However, the origin of such pulse deformation has never been clarified yet, and the behavior of transient current still remains in an unexplored area.

First theoretical study of time-dependent transport through nanosystems was performed by Wingreen et al. for the resonant-tunneling system under a finite bias voltage [14]. Since then, various methods have been developed to calculate time-dependent current through nanosystems and these are classified into three groups. One is the method using the time-dependent nonequilibrium Green’s function in the Keldysh formalism [14–16]. This method first prepares Green’s functions of electrodes and nanosystem separately and then connects them by using the Dyson-type equation with the electronic couplings between electrodes and nanosystem as the interactions. This formulation is easy to exactly include the interactions between electrodes and nanosystem. However, it is difficult to arrange arbitrary initial conditions, especially in case of nanosystems having many-body interactions [17, 18]. Alternative approach is based on the use of the time-dependent density matrix based on the Liouville equation [19–21]. This method evaluates the density matrix of nanosystem by eliminating the electrode freedoms from the density matrix of the total system using the projection operator. Since the projection is not compatible with the interactions between electrodes and nanosystem, we have to use the perturbation approximation to such interactions. However, we can easily arrange the arbitrary initial conditions to the nanosystem and include many-body effects in the nanosystem. Moreover, as shown later, it is easy to obtain the physical pictures. The third approach is the one using the time-dependent density functional theory [22]. This method can treat the electronic structures and interactions between electrode and nanosystem in realistic manner. However, it is still limited to the closed system, i.e., the microcanonical system, and to the linear response regime.

In most studies so far, the calculated transient current sometimes shows the relaxation and oscillation [23–26]. However, the origins of such transient behavior have not been understood well. To understand such origins, we employed the density-matrix method. Pedersen et al. used the density-matrix method to realize the steady-state current in case that the nanosystem has electron-electron interaction [19]. However, with respect to the transient behavior, they assumed the Markov limit and neglected the memory effect. Therefore, their result does not simulate the correct transient behavior. In our previous paper, we have already reported the characteristic behavior of transient current obtained by fully taking into account the memory effect, briefly [20, 21]. We have found that the transient current behavior is characterized with the oscillation and relaxation processes. The memory effect

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induces the oscillation of transient current. In this paper, we give the more detailed derivation of the transient current formula, and clarify the origins of the transient current behaviour. Furthermore, the electron-electron interaction effects on the transient current are studied. We clarify that the relaxation time is decreased by the strong electron-electron interactions.

The rest of this paper is organized as follows. In Sec. II, we present the model system to study the transient current through the nanosystem. Then, we explain our calculation method of time-dependent current, which includes the memory effect. In Sec. III, we show the calculated transient currents through the nanosystem and discuss the physical origins of the relaxation and oscillation. Then we consider how the transient current changes when the bandwidth and density of states of electrodes changes and when the electron-electron interaction is included in the nanosystem. In Sec. IV, we present our conclusions.

II. MODEL AND METHODOLOGY

In this paper, we consider a nanoscale system that is sandwiched by left and right metallic bulk electrodes. Hereafter, we call such nanoscale system as the center system (C-system). The schematic picture of this arrangement is shown in Fig. 1. First, we consider the simplest case where the electron is a spinless fermion and the C-system has a single electronic state with the energy, $\varepsilon_C$. The Hamiltonian of this system is written as

$$\hat{H} = \varepsilon_C \hat{a}^\dagger \hat{a} + \sum_{k,\alpha=L,R} \varepsilon_{k\alpha} \hat{a}_{k\alpha}^\dagger \hat{a}_{k\alpha}$$

$$+ \sum_{k,\alpha=L,R} v_{\alpha} (\hat{a}_{k\alpha}^\dagger \hat{a}_{k\alpha} + \hat{a}_{k\alpha} \hat{a}_{k\alpha}^\dagger),$$

where $\hat{a}^\dagger$ and $\hat{a}$ are creation and annihilation operators of an electron, respectively. The first term in the right-handed side of this equation corresponds to the energy of the C-system, while the second term to those of left ($\alpha=L$) and right ($\alpha=R$) electrodes that are characterized by the bulk wavenumber, $k$. The last term represents the electron-transfer energy between electronic states of the C-system and electrodes, where we adopt for simplicity that $v_{\alpha}$ does not depend on $k$.

We investigate the time evolution of this system based on the Liouville equation of density matrix; $\partial \hat{\rho}(t)/\partial t = i [\hat{L}, \hat{\rho}(t)]$, where $\hat{L}$ is the super operator called the Liouville operator and $\hat{\rho}(t)$ is the density matrix [21]. The Liouville operator is defined for any operators, $\bullet$, as $\hat{L} \bullet \equiv -1/i[\hat{L}, \bullet]$. The operators $\hat{L}_L$, $\hat{L}_C$ and $\hat{L}_R$ are Liouville operators, respectively corresponding to the $\alpha$-electrode, the C-system and the electron-transfer parts of the total Hamiltonian. Since we solve the equation of motion, some higher-order effects of $v_{\alpha}$ are automatically included in the time evolution. However, it should be noted here that such treatment neglects higher-order quantum effect of $v_{\alpha}$ in the electron transition between the C-system and electrodes, which is sometimes recognised as the Kondo effect [28].

In this section, we present the model system to study the transient current formula, and clarify the origins of the transient effects of the C-system on the electrode and is justified when the electronic coupling, $v_{\alpha}$, is sufficiently small as discussed later.

To incorporate the above assumption into the Liouville equation, we introduce the following super operators called the projection operators, $\hat{P}$ and $\hat{Q}$, that are defined by $\hat{P} \bullet = \hat{P}_L \hat{P}_R \hat{T}_L \hat{T}_R \bullet$ and $\hat{Q} = 1 - \hat{P}$, respectively [27]. In this case, the density matrix is projected as $\hat{P} \hat{\rho}(t) = \hat{P}_L \hat{P}_R \hat{\rho}_C(t)$, where $\hat{\rho}_C(t)$ is the density matrix of the C-system defined by $\hat{\rho}_C(t) = \hat{T}_L \hat{T}_R \hat{\rho}(t)$. Applying these projection operators to the Liouville equation and adopting the second-order approximation of $v_{\alpha}$ in the time-evolution equation, we obtain the following equation, which details are described in Appendix;

$$\frac{\partial \hat{\rho}_C(t)}{\partial t} = i \hat{L}_C \hat{\rho}_C(t)$$

$$+ \hat{T}_L \hat{T}_R i \hat{L}_C \int_{t_0}^{t} d\tau e^{i(\epsilon_C(\tau - \tau))} \hat{L}_C \hat{P}_L \hat{P}_R \hat{\rho}_C(\tau).$$

(2)

Here, $\hat{L}_0$ is defined by $\hat{L} - \hat{L}_0 = \hat{L}_C + \hat{L}_L + \hat{L}_R$. The operators $\hat{L}_C$, $\hat{L}_L$ and $\hat{L}_R$ are Liouville operators, respectively corresponding to the $\alpha$-electrode, the C-system and the electron-transfer parts of the total Hamiltonian. Since we solve the equation of motion, some higher-order effects of $v_{\alpha}$ are automatically included in the time evolution. However, it should be noted here that such treatment neglects higher-order quantum effect of $v_{\alpha}$ in the electron transition between the C-system and electrodes, which is sometimes recognised as the Kondo effect [28].

Transforming into the interaction representation defined by $\hat{O}_I(t) \equiv \exp[i\hat{H}_0(t-t_0)/\hbar] \hat{O} \exp[-i\hat{H}_0(t-t_0)/\hbar]$, we obtain

$$\frac{\partial \hat{\rho}_{IC}(t)}{\partial t} = -i \hat{L}_C e^{-i\epsilon_C t} \hat{\rho}_{IC}(t) + e^{-i\epsilon_C t} \frac{\partial \hat{\rho}_{IC}(t)}{\partial t},$$

(3)

where we use the following relation, $\hat{\rho}_{IC}(t) \equiv \exp[i\hat{H}_0(t)/\hbar] \hat{\rho}_{IC}(t) \exp[-i\hat{H}_0(t)/\hbar] = \exp(-i\epsilon_C t) \hat{\rho}_{IC}(t)$. $\hat{H}_0$ is the Hamiltonian compounded from the first and second terms of Eq. (1). Inserting Eq. (2) to Eq. (3), we obtain the time-evolution equation of the C-system in the interaction picture.
Using the relation, \( \exp(i\hat{L}_0t) = \exp(-i\hat{H}_0t/h) \exp(i\hat{H}_0t/h) \), the equation is written as

\[
\frac{\partial \hat{\rho}_{IC}(t)}{\partial t} = \text{Tr}_L\text{Tr}_R e^{-i\hat{L}_0t} i\hat{L}_v \int_0^t d\tau e^{i\hat{L}_0(t-\tau)} i\hat{L}_v \hat{\rho}_L \hat{\rho}_R \hat{\rho}_C(\tau) \cdot (4)
\]

Performing the simple commutation calculations, we obtain

\[
\frac{\partial \hat{\rho}_{IC}(t)}{\partial t} = \sum_{\alpha=L,R} \left( \frac{v_\alpha}{\hbar} \right)^2 \int_0^t d\tau \sum_k \sum_{k'} \left[ -\hat{a}_I(t)\hat{a}^\dagger_I(\tau)\hat{\rho}_{IC}(\tau)\text{Tr}_\alpha[\hat{\rho}_\alpha\hat{a}^\dagger_{I\alpha}(0)\hat{a}_{I\alpha}(\tau-t)] -\hat{a}^\dagger_I(t)\hat{a}_I(\tau)\hat{\rho}_{IC}(\tau)\text{Tr}_\alpha[\hat{\rho}_\alpha\hat{a}_{I\alpha}(0)\hat{a}^\dagger_{I\alpha}(\tau-t)] +\hat{a}_I(t)\hat{\rho}_{IC}(\tau)\hat{a}^\dagger_I(\tau)\text{Tr}_\alpha[\hat{\rho}_\alpha\hat{a}_{I\alpha}(0)\hat{a}^\dagger_{I\alpha}(\tau-t)] +\hat{a}^\dagger_I(t)\hat{\rho}_{IC}(\tau)\hat{a}_I(\tau)\text{Tr}_\alpha[\hat{\rho}_\alpha\hat{a}_{I\alpha}(0)\hat{a}_{I\alpha}(\tau-t)] -\hat{\rho}_{IC}(\tau)\hat{a}_I(t)\hat{a}^\dagger_I(\tau)\text{Tr}_\alpha[\hat{\rho}_\alpha\hat{a}_{I\alpha}(0)\hat{a}^\dagger_{I\alpha}(\tau-t)] -\hat{\rho}_{IC}(\tau)\hat{a}^\dagger_I(t)\hat{a}_I(\tau)\text{Tr}_\alpha[\hat{\rho}_\alpha\hat{a}_{I\alpha}(0)\hat{a}_{I\alpha}(\tau-t)] \right] . (6)
\]

In this equation, the traces over the eigenstates of electrodes are calculated as

\[
\text{Tr}_\alpha[\hat{\rho}_\alpha \hat{a}^\dagger_{I\alpha}(0) \hat{a}_{I\alpha}(\tau-t)] = e^{-i\epsilon_{k\alpha}(\tau-t)/\hbar} f^\alpha_k \delta_{kk'}, (7)
\]

where \( f^\alpha_k = 1/(e^{\beta(\epsilon_{k\alpha}-\mu_\alpha)} + 1) \) is the Fermi distribution function of electrodes.

Then, we consider the matrix elements of \( \hat{\rho}_{IC}(t) \). In the present model, the C-system has two bases; \([1] \) and \([0] \). The former corresponds to the electron occupied state, while the latter to the unoccupied state. Diagonal matrix elements, \( \rho_{C,1}(t) = \langle 1|\hat{\rho}_C(t)|1 \rangle \) and \( \rho_{C,0}(t) = \langle 0|\hat{\rho}_C(t)|0 \rangle \), represent the electron occupied and unoccupied probabilities of the C-system. Using Eqs. (6) and (7), the time evolution of \( \rho_{C,1}(t) \) is written in the form of charge-density conservation law as

\[
e^{-i\epsilon_{k\alpha}(\tau-t)/\hbar}\sum_k \cos((\epsilon_C - \epsilon_{k\alpha})(\tau-t)/\hbar) f^\alpha_k \rho_C, \, \alpha(\tau) \cdot (8)
\]

Here, \( I_\alpha(t) \) is the electronic current flowing through the contact from the \( \alpha \) electrode to the C-system, and is given by

\[
I_\alpha(t) = 2e \left( \frac{v_\alpha}{\hbar} \right)^2 \int_0^t d\tau \sum_k \cos((\epsilon_C - \epsilon_{k\alpha})(\tau-t)/\hbar) f^\alpha_k \rho_C, \, \alpha(\tau) - (1 - f^\alpha_k) \rho_{C,1}(\tau) ) . (9)
\]

This equation have the clear physical meaning; the first term in the right-handed side, \( f^\alpha_k \rho_C, \, \alpha \), corresponds to the electronic transition from the \( k \)-th state in the \( \alpha \) electrode to the energy level in the C-system, while the second term, \( (1 - f^\alpha_k) \rho_{C,1} \), to that from the C-system to the \( \alpha \) electrode. The \( 2(v_\alpha/\hbar)^2 \) and cosine prefactors represent the time-dependent transition probability. Using the normalization relation of \( \rho_{C,1}(t) + \rho_{C,0}(t) = 1 \), this current can be written as

\[
I_\alpha(t) = 2e \left( \frac{v_\alpha}{\hbar} \right)^2 \int_0^t d\tau \sum_k \cos((\epsilon_C - \epsilon_{k\alpha})(\tau-t)/\hbar) \times \{ -\rho_{C,1}(\tau) + f^\alpha_k \} . (10)
\]

In this way, we obtained the coupled equations, Eqs. (8) and (10), for the density-matrix element, \( \rho_{C,1}(t) \), and the \( \alpha \)-electrode-side current, \( I_\alpha(t) \). These equations include only the \( \rho_{C,1}(t) \) element. This occurs because we adopt the second-order approximation of \( v_\alpha \) in the time-evolution equation.

We numerically solve the time-evolution of density matrix, i.e., Eqs. (8) and (10). In all simulations, we first prepare the C-system and two electrodes separately, then connect these systems at \( t_0 = 0 \). After the steady state is realized, we next apply the voltage to the left electrode at \( t = 30 \) (in the unit of \( \hbar/(\epsilon_C - \mu_R) \)). We adopted the first connection at \( t_0 = 0 \) experimentally because the most nano-contact current measurements are performed after connecting the C-system with electrodes and theoretically because such connection induces the quantum hybridization between energy states of the C-system and electrodes and promotes unintentional transient currents. On the other hand, the voltage application is realized by employing the time-dependent Fermi energy, \( \mu_L(t) \), in \( f^\alpha_k \) of the left electrode [29].

There are many parameters in the present model. We chose the energy difference between the energy level of the C-system and the Fermi energy of right electrode, \( \epsilon_C - \mu_R \),
as the unit of energy, and set $\varepsilon_C$ and $\mu_R$ as 1.0 and 0.0, respectively. Fermi energy of right electrode, $\mu_R$, keeps this constant value during the time evolution, while the Fermi energy of left electrode, $\mu_L$, changes from 0.0 to 2.0 at time $t = 30$ corresponding to the application of voltage. We characterize the transfer energies between C-system and electrodes by a single small transfer energy as $\varepsilon = \varepsilon_L = \varepsilon_R = 0.1$. As an initial condition, we assume that there is no electron in the C-system before the connection to electrodes is realized, i.e., $t < t_0 = 0$. However, since we apply the voltage at $t = 30$ after the steady state is realized by the connection, such initial electron occupancy does not affect the results obtained in this paper. Moreover, we assume the zero temperature for the grand-canonical ensembles of electrodes, which treatment simplifies the calculation.

III. RESULTS AND DISCUSSION

A. Origins of transient behavior

In this subsection, we first investigate the transient behavior of current by considering the simplest case; both electrodes have the same constant density of states, $D = D_L = D_R = 1.0$, and an infinite band width. In this case, we can replace the summation of $k$ in Eq. (10) by the simple integration as $I_{\alpha}(t) = \int d\varepsilon_\alpha \cdot D_{\alpha}$ and rewrite the current formula of Eq. (10) as [21]

$$I_{\alpha}(t) = \frac{e}{\hbar} 2\pi v^2 d_{\alpha} \{-\rho_{C,1}(t) + \Gamma(\mu_{\alpha} - \varepsilon_C; t - t_0)\},$$

where $\Gamma$ is given by

$$\Gamma(\mu_{\alpha} - \varepsilon_C; t - t_0) = \frac{1}{2} + \int_{t-t_0}^{t} ds \left(\frac{\sin((\mu_{\alpha}(s) - \varepsilon_C)s/\hbar)}{\pi s}\right).$$

First, we consider the steady-state values of current and electron number. Using the steady-state conditions that the time is infinity and $\rho_C(t)$ in Eqs. (8) and (11) has a constant value, the transition probabilities become independent of time. In this case, one obtain the balance equation of transition probability as $\nu_{R}^2 D_R \cdot \rho_{C,1} = \nu_{L}^2 D_L \cdot \rho_{C,0}$. Thus, when the energy level of the C-system is located between the Fermi energies of left and right electrodes, i.e., $\mu_L > \varepsilon_C > \mu_R$, the electron occupation is $\nu_L^2 D_L / (\nu_L^2 D_L + \nu_R^2 D_R)$, while both left and right-contact currents become $2\pi v^2 \hbar / (v_L^2 D_L + v_R^2 D_R)$. The steady-state current is consistent with that obtained from the Master equations [30, 31]. Especially, in the present case of the same transfer energy and the same density of states for left and right electrodes, the electron occupation and the current become a half and $2\pi v^2 \hbar / v^2 D / 2$, respectively. On the other hand, when $\varepsilon_C > \mu_L$ or $\mu_R > \varepsilon_C$, the electron number is zero or unity, respectively, while the current always becomes zero.

Next, we consider the transient behavior of currents and electron number. Figure 2(a) shows the calculated transient current flowing at the left and right contacts as a function of time, while Fig. 2(b) the corresponding electron density in the C-system. The current is displayed in units of $2\pi v^2 D e / \hbar$, while the time is in units of $\hbar / (\varepsilon_C - \mu_R)$. It is seen that the left-contact current suddenly starts to flow at $t = 30$ and approaches a steady-state value of 0.5 with exploring the oscillation. On the other hand, both the right-contact current and the electron number gradually increase and approach the steady-state values with also exhibiting the oscillation. Since the physical origins of transient behavior are same for the left and right-contact currents, we concentrate on the left-contact current for a while.

We first consider the relaxation of transient current to the steady state. The relaxation occurs due to the first term in Eq. (11) because this term acts as a friction-like term in Eq. (8) for the electron-number change in the C-system. Accordingly, the relaxation time of transient current to the steady state reflects the effective coupling magnitude between electrode and the C-system as $\tau = \hbar / 2\pi v^2 D$. In our previous paper, we have investigated the relaxation of transient current under the transfer energy $v = 0.2$, which is twice the magnitude of that used in this paper [21]. It is confirmed that the relaxation time becomes shorter when the transfer energy, $v$, becomes larger. Such coupling dependence is physically reasonable because this term appears reflecting the process that the electron in the C-system goes back to diffuse into a number of electronic states in the electrodes. In this sense, the relaxation of transient current represents the electronic energy dissipation of the C-system into electrodes.
On the other hand, the oscillation of transient current occurs due to the second term of Eq. (11), i.e., \( \Gamma(t) \) in Eq. (12). This term appears due to the time-dependent transitions of electrons between various energy levels of electrodes and the energy level of the C-system and corresponds to the memory-function term in the Mori’s density-matrix formalism [32]. In fact, as seen in Eq. (10), these transitions have various frequencies such as \((\varepsilon_{\alpha} - \varepsilon_{\alpha\alpha})/h\). However, the sum of such transitions over the occupied \( k \) states in electrodes results in the oscillation term in Eq. (12) with a single frequency, \((\mu_{\alpha} - \varepsilon_{C})/h\), thus depending on only the energy difference between the Fermi energy of the electrode and the discrete energy level in the C-system. In this sense, one can assign the oscillation to the Fermi-energy-edge effect, which reflects the Fermion property of electrons. As seen in the prefactor of \( \Gamma(t) \) in Eq. (11), the oscillation amplitude is also proportional to the effective coupling, \( v^{2}D \), thus the amplitude increasing with increasing the coupling.

Then, we consider why such oscillation damps down as the time proceeds. There are two origins to induce the damping. One is the first term in Eq. (11), i.e., the friction-like term corresponding to the electron dissipation into electronic states in electrodes. This term always acts to prevent the change of current. The other origin exists in the time-evolution nature of \( \Gamma(t) \). \( \Gamma(t) \) shows the oscillation for a small value of \( t \). However, as the time proceeds, \( \Gamma(t) \) shows a gradual decrease of oscillation amplitude and converges to a constant value, i.e., unity and zero in cases of left and right contacts, respectively. Such change of \( \Gamma(t) \) occurs physically because the sum of time-dependent electron transitions over the elapse time gradually loses the interference of transitions.

Then, we consider the different behavior of transient current between left and right contacts. Such difference originates from the initial electron occupancy in the C-system. Since there is no electron in the C-system before \( t = 30 \), the left-contact current is initially amplified to quickly realize the electron occupation, while the right-contact current starts only after the electron occupancy is realized in the C-system. For example, the amplified current is observed at the left contact in the experiment [12]. It is noted here that, because of the time delay in the right contact, the oscillation phase of right-contact current does not coincide with that of left-current. As a result, the electron number shows a simple increase with a small oscillation amplitude compared to those of currents.

Finally, we evaluate typical values of the relaxation time and the oscillation period for the semiconductor quantum-dot system with two-dimensional electron electrodes as an example. When the electrode is made of n-doped GaAs, which has the effective mass of conduction-band electron around \( m = 0.07 \) [13], the density of states in electrodes is estimated as \( D = (m/\pi h^{2}) \cdot (\pi \lambda^{2}/2) \approx 0.46 \text{ eV}^{-1} \). Here, since the coupling of the electrode and the C-system is like a point-contact, we assumed that the electronic states within a half circle of the radius, \( \lambda = 1 \text{ nm} \), effectively contribute to the contact and evaluated the density of states. Therefore, when the transfer energy is \( v = 0.1 \text{ eV} \), the relaxation time is evaluated as \( \tau = h/2\pi v^{2}D = 22.8 \text{ fs} \). On the other hand, when the bias voltage is applied between the left and right electrodes such as \( |\mu_{\alpha} - \varepsilon_{C}| = 1 \text{ eV} \), the oscillation periods due to the Fermi-energy-edge effect is evaluated as \( 4.1 \text{ fs} \) independent of the effective coupling \( v^{2}D \), while the band-edge effect shown in the next subsection gives the similar oscillation period. As seen in these estimates, the weak coupling between the C-system and electrodes is the key quantity to enable the experimental observation of the relaxation and the oscillation.

### B. Effect of finite band width

Here, we investigate how the transient current changes when the electronic bands in electrodes have a constant density of states, \( D \), but a finite bandwidth ranging from \( \varepsilon_{\alpha}^{\text{bottom}} \) to \( \varepsilon_{\alpha}^{\text{top}} \). In this case, the time-dependent current is evaluated as [21]

\[
I_{\alpha}(t) = \frac{e}{h} 2\pi v^{2}D_{\alpha} \left[ -\int_{0}^{t-t_{0}} ds \left\{ \frac{\sin((\varepsilon_{\alpha}^{\text{top}} - \varepsilon_{C})s/h)}{\pi s} - \frac{\sin((\varepsilon_{\alpha}^{\text{bottom}} - \varepsilon_{C})s/h)}{\pi s} \right\} \rho_{C,1}(t-s) + \Gamma(t-t_{0}) \right], \tag{13}
\]

where \( \Gamma(t) \) is given by

\[
\Gamma(t-t_{0}) = \int_{0}^{t-t_{0}} ds \left\{ \frac{\sin((\mu_{\alpha} - \varepsilon_{C})s/h)}{\pi s} - \frac{\sin((\varepsilon_{\alpha}^{\text{bottom}} - \varepsilon_{C})s/h)}{\pi s} \right\}. \tag{14}
\]

The first and second terms in the integrand of Eq. (13) correspond to those in Eq. (11), respectively.

Figures 3(a) and 3(b) show the calculated transient currents in cases of different bandwidths. Here, we adopted \( \varepsilon_{\alpha}^{\text{top}} = 3 \) and \( \varepsilon_{\alpha}^{\text{bottom}} = -3 \) in Fig. 3(a), while \( \varepsilon_{\alpha}^{\text{top}} = 6 \) and \( \varepsilon_{\alpha}^{\text{bottom}} = -6 \) in Fig. 3(b). We note that the damping speed of the oscillation becomes slow as the bandwidth of electrode decreases. This is because, as the bandwidth and thus the number of electronic states in electrode decreases, the electron dissipation into such electronic states is suppressed and the interference loss due to the sum of various transitions over the elapse time decreases, thus diminishing both two major origins that promote the damping. This situation is easily recognized by seeing Eqs. (13) to (14); the integration in the first term of Eq. (13) provides a small relaxation term compared to that in Eq. (11), while the second oscillating term given in Eq. (14) slightly prolongs the oscillation compared to the case of
The current is displayed in units of $2\pi v^2 De/h$, where $v$ is the velocity of electrons, $D$ is the density of states, and $E$ is the energy. The times are in units of $h/(\varepsilon_C - \mu_R)$. Figure 3(a) shows the transient currents at the left and right contacts, where $I_L$ and $-I_R$, when both left and right electrodes have the finite band width ranging between (a) $\varepsilon^{\text{top}}_a = 3$ and $\varepsilon^{\text{bottom}}_a = -3$, and (b) $\varepsilon^{\text{top}}_a = 6$ and $\varepsilon^{\text{bottom}}_a = -6$. The current is displayed in units of $2\pi v^2 De/h$, while the time is in units of $h/(\varepsilon_C - \mu_R)$.

Moreover, we can see in Fig. 3(b) that a new oscillation having shorter period is added to the original oscillation. This new oscillation occurs due to the second term in Eq. (14), which reflects the finiteness of electrode bandwidth and has the frequency corresponding to the energy difference between the bottom of the band in electrode and the discrete energy level of the C-system. In this sense, we can assign this new oscillation to the band-edge effect. When the periods of both oscillations become close to each other, both oscillations cooperate with each other and one can see the large oscillation as displayed in Fig. 3(a).

C. Effect of electron-electron interaction

In this subsection, we consider what changes are induced in the transient current when the C-system has a single site with the electron-electron interaction. For this purpose, we consider the spin freedom of electrons and adopt the single-site Hubbard model as the simplest model, whose Hamiltonian is written by

$$\hat{H}_C = \varepsilon_C (\hat{a}_1^\dagger \hat{a}_1 + \hat{a}_1^\dagger \hat{a}_1) + U \hat{a}_1^\dagger \hat{a}_1 \hat{a}_1^\dagger \hat{a}_1,$$  (15)

where $\varepsilon_C$ and $U$ are a single-site energy and an electron-electron repulsive energy, respectively. Thus, the C-system is represented by four bases, $|0\rangle$, $|\uparrow\rangle$, $|\downarrow\rangle$ and $|\uparrow\downarrow\rangle$. In this case, the density matrix becomes a $4 \times 4$ matrix with diagonal elements such as $\rho_{C,0}(t) = \langle 0 | \hat{\rho}(t) | 0 \rangle$ and $\rho_{C,1}(t) = \langle \uparrow | \hat{\rho}(t) | \uparrow \rangle$.

Using the same approximations as the case of the non-interacting single-site model used in the previous subsection, one can derive the time-evolution equations of diagonal elements as

$$\frac{\partial \rho_{C,0}(t)}{\partial t} = \sum_{\alpha=L,R} \frac{2v_\alpha^2}{h} \int_0^t d\tau \sum_k \cos((\varepsilon_C - \varepsilon_{\alpha \uparrow})(\tau - t)/h)\{-2f_k^{\alpha \uparrow} \rho_{C,0}(\tau) + (1 - f_k^{\alpha \uparrow})\rho_{C,1}(\tau) + \rho_{C,1}(\tau)\},$$

$$\frac{\partial \rho_{C,1}(t)}{\partial t} = \sum_{\alpha=L,R} \frac{2v_\alpha^2}{h} \int_0^t d\tau \sum_k \left[ \cos((\varepsilon_C - \varepsilon_{\alpha \uparrow} + U)(\tau - t)/h)\{-2f_k^{\alpha \uparrow} \rho_{C,1}(\tau) + (1 - f_k^{\alpha \uparrow})\rho_{C,1}(\tau) \right. + \cos((\varepsilon_C - \varepsilon_{\alpha \uparrow})(\tau - t)/h)f_k^{\alpha \uparrow} \rho_{C,0}(\tau) - (1 - f_k^{\alpha \uparrow})\rho_{C,1}(\tau) \right].$$  (16)

The equation of $\rho_{C,1}(t)$ is obtained by exchanging $\uparrow$ and $\downarrow$ in the latter equation, while that of $\rho_{C,1}(t)$ is derived using the probability conservation, $\rho_{C,0}(t) + \rho_{C,1}(t) + \rho_{C,1}(t) + \rho_{C,1}(t) = 1$.

Figures 4(a) and 4(b) show the calculated transient currents and electron numbers, respectively, for various values of $U$ as a function of time. The schematic energy diagram is displayed in the inset. We first consider the steady state. Figure 5(a) shows steady-state values of the left-contact current and the electron number as a function of $U$. When $U$ is small such that both the lower and upper Hubbard levels are located between Fermi energies of left and right electrodes as $\mu_L > \varepsilon_C + U > \varepsilon_C > \mu_R$, these values coincide with those in the case of the C-system without the electron-electron interaction. On the other hand, when $U$ increases and the upper Hubbard level becomes higher than the Fermi energy of the lower electrode as $\varepsilon_C + U > \mu_L$, the steady current and electron number quickly change to small values. It is noteworthy that these small values are larger than the halves of those of smaller $U$ cases in spite that only a single lower Hubbard level is located between Fermi energies of left and right electrodes. Such enhancement of electron occupation for a large $U$ is known as the prohibition effect of two-spin occupation in the steady state. In fact, when the two-spin occupation is forbidden as $\rho_{C,11} = 0$, the steady
FIG. 4: (a) Calculated transient currents at left contact for various values of U as a function of time. The inset shows the schematic energy diagram of the system. (b) Time-dependent behavior of electron numbers in the C-system for various values of U.

state is realized with the balance of other probabilities as \( \rho_{C,1} = \rho_{C,4} = \rho_{C,0} = 1/3 \). This is because the transition probabilities between these probabilities have the same values as shown in Fig. 6. Therefore, in the steady-state cases with large U, the electron number and the accompanying current become exactly two third of the cases with smaller U.

Then, we consider the relaxation of transient current. Figure 5(b) shows the calculated relaxation time as a function of U value. Here we defined the relaxation time as the time when the current magnitude is reduced to \( e^{-3} \) of the first peak. When U becomes large and enter into the prohibition region of two-spin state, the relaxation time sharply decreases. This might also occur because the two-spin state is forbidden and thus the number of transition paths decreases as shown in Fig. 6.

IV. CONCLUSIONS

We studied the time-dependent current flowing through a nanoscale system sandwiched by two bulk electrodes, based on the Liouville equation of density matrix, when a sudden voltage is applied between electrodes. It was shown that the transient current is characterized with the relaxation and the oscillation. We clarified that the relaxation is caused by the energy dissipation of nanoscale system into a number of electronic states with various energies in the electrode. Thus, the relaxation time depends on the effective coupling between the electrode and the nanoscale system, which is represented by the density of states in electrode and the electron transfer energy, and becomes longer when the bandwidth of electrode decreases. On the other hand, the oscillation occurs due to the time-dependent electronic transitions between the energy levels of electrode and that of nanoscale system, thus reflecting the memory effect. It was clarified that the oscillation period depends on the Fermi energy and the bandwidth of the electrode. Furthermore, we investigated the effects of electron-electron interaction on the transient current and clarified that, when the electron-electron repulsive energy becomes large and enter into the prohibition region of two-spin state, the relaxation time decrease due to the decrease of quantum transition paths.
Acknowledgments

This work was supported by the Next Generation Super Computing Project, Nanoscience Program, the Ministry of Education, Culture, Sports, Science and Technology, Japan, the 21COE program of Chiba University, and ISSP, University of Tokyo.

Appendix

In this Appendix, we show the derivation of time-evolution equation of the C-system shown in Eq. (2). We follow the Nakajima-Zwanzig projection operator formalism [27, 33, 34]. Applying the projection operators, \( \hat{P} \) and \( \hat{Q} \), to the Liouville equation, we obtain the following coupled equations:

\[
\begin{align*}
\hat{P} \frac{\partial \hat{\rho}(t)}{\partial t} &= \hat{P}i\hat{L}\hat{\rho}(t) + \hat{P}i\hat{Q}\hat{\rho}(t), \quad (17) \\
\hat{Q} \frac{\partial \hat{\rho}(t)}{\partial t} &= \hat{Q}i\hat{L}\hat{\rho}(t) + \hat{Q}i\hat{Q}\hat{\rho}(t). \quad (18)
\end{align*}
\]

Here, we use the rule that the right-side operator acts at first in the calculation. For example, \( \hat{P}i\hat{L}\hat{\rho} \) means \( \hat{P}(i\hat{\mathcal{L}}(\hat{P}\hat{\rho})) \). If the C-system is not connected to the electrodes at the initial time, \( t_0 \), the latter equation is exactly solved as

\[
\hat{Q}\hat{\rho}(t) = \int_{t_0}^{t} d\tau e^{(t-\tau)\hat{Q}i\hat{L}}\hat{Q}\hat{\rho}(\tau). \quad (19)
\]

Insertion of Eq. (19) into Eq. (17) gives the time-evolution equation of projected density matrix;

\[
\frac{\partial \hat{P}\hat{\rho}(t)}{\partial t} = \hat{P}i\hat{L}\hat{\rho}(t) + \hat{P}i\hat{L}\hat{\rho}(t)
\]

By using the following identities for any operators, \( \bullet; \)

\[
\begin{align*}
\hat{iL}_{\alpha}\hat{\rho}_{\alpha} &= 0, \quad \text{Tr}_{\alpha}[\hat{L}_{\alpha}\hat{\rho}_{\alpha}] = 0, \quad \hat{iL}_{\alpha}\hat{P} \bullet = 0, \quad \hat{P}i\hat{L}_{\alpha}\hat{P} \bullet = 0, \quad \text{and} \quad \hat{P}i\hat{L}_{\bullet} = \hat{P}i\hat{L}_{\bullet}, \quad \text{the first term of this equation can be written as}
\end{align*}
\]

\[
\hat{P}i\hat{L}\hat{\rho}(t) = \hat{P}i\hat{L}\hat{\rho}(t), \quad (21)
\]

while the second term as

\[
\hat{P}i\hat{L}\int_{t_0}^{t} d\tau e^{(t-\tau)\hat{Q}i\hat{L}}\hat{L}\hat{\rho}(\tau) = \hat{P}i\hat{L}_{\alpha}\hat{\rho}(t) \quad (22)
\]

Here, \( \hat{L}_{\alpha}, \hat{L}_{C} \) and \( \hat{L}_{o} \) are Liouville operators, respectively corresponding to the \( \alpha \)-electrode, the C-system and the electron-transfer parts of the total Hamiltonian. As a result, we obtain the following time-evolution equation of the C-system;

\[
\frac{\partial \hat{\rho}_{C}(t)}{\partial t} = i\hat{L}_{C}\hat{\rho}_{C}(t) + \text{Tr}_{L}\text{Tr}_{R}i\hat{L}_{\alpha}\int_{t_0}^{t} d\tau e^{(t-\tau)\hat{Q}i\hat{L}}\hat{L}\hat{\rho}_{R}\hat{\rho}_{C}(\tau). \quad (23)
\]

This equation of motion is exact and represents the reversible process of the total system. However, when the bulk electrodes have the macroscopic degree of freedom and the coupling between the electrode and C-system is sufficiently weak, the electronic states in electrodes change very slowly compared to the electronic state in the C-system. Therefore, we can assume that the electronic states of electrodes always follow the time-independent grand canonical ensemble and approximate \( \hat{L} \) in the exponent of the second term of Eq. (23) by \( \hat{L}_0 \), where \( \hat{L}_0 = \hat{L} - \hat{L}_{e} = \hat{L}_{C} + \hat{L}_{L} + \hat{L}_{R} \), thus the exponent being replaced as

\[
\text{exp} \left( (t-\tau)\hat{Q}i\hat{L} \right) \to \text{exp} \left( (t-\tau)\hat{Q}i\hat{L}_0 \right) = \text{exp} \left( (t-\tau)i\hat{L}_0 \right). \quad (24)
\]

This approximate treatment induces the information loss about the time evolution of electronic states in electrodes and promotes the irreversible energy-dissipation process for the C-system. We obtain the time-evolution equation of the C-system shown in Eq. (2) in the second-order approximation of \( v_{o} \).

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