

Dynamical responses of quantum dots by pulsed fields

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Tunneling currents of a quantum dot induced by pulsed fields are investigated theoretically using time-dependent tunneling rates. Taking into account nonadiabatic effects, we present simple analytic expressions for tunneling currents, which can be used to study the evolution of the occupation probabilities in the quantum dot as well as extract system parameters governing tunneling measurements.

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I. INTRODUCTION

There has been considerable interest in semiconductor quantum dots because of both fundamental and applied aspects. As well as for optoelectronic nanodevices,^{1,2} quantum dots are considered as potential building-blocks for quantum computation and information processing because electronic wave functions are easily tailored by patterning electrodes.^{3,4} For sophisticated implementation of such devices, it is essential to understand detailed electronic structure of quantum dots. Tunneling spectroscopy is one of the commonly performed methods to characterize the electronic structure of quantum dots.⁵⁻⁷ In this method, a quantum dot is placed between two adjacent macroscopic electrodes and separated by tunneling barriers. Then, since tunneling is strongly affected by energy levels in the quantum dot, barrier heights, and capacitances between the quantum dot and leads, currents through the system contain the information related to them.

Although, in most experimental work, tunneling currents are measured in static conditions, we examine responses of the quantum dot to time-dependent perturbing fields, especially to square pulsed form in this work. The reasons for this are twofold. The first is to obtain the information about the electronic structure of a quantum dot which may be missing in static cases. Namely, we search for nonadiabatic effects to give further information about the electronic structure. Along this aspect, several experimental works have been also reported.⁸ The other is the controllability of the occupation of electrons in the quantum dot by pulsed fields, which can be extended to basic operations to manipulate quantum bits in quantum computations.

We consider a semiconductor quantum dot with two closely-located-energy levels. This situation occurs in a doubly stacked-quantum-dot geometry, where a dot-dot interaction splits energy levels into bond and anti-bonding states with their energy difference of about a thermal energy or tunneling rates.⁹ This geometry is of prime interest because the bonding and antibonding states can be considered as a basis of a charge qubit and the occupancy of each state may be controlled by electrical pulses.⁷

For such a system, calculations of tunneling currents are complicated by both nonadiabatic effects from external time-dependent perturbations and charging effects of electrons in

the quantum dot. To do this, we use the formalism recently developed by the authors based on the reduced-density-operator theory.¹⁰ In the absence of the charging effects, a similar problem is investigated by Wingreen, Jauho, and Meir¹¹ on resonant-tunneling structures. Even though the energy band diagram of their system is very similar to ours (Fig. 1), detailed formula governing the occupancy in the central region is very different. The major difference between two approaches comes from the constraint about the total occupancy of two levels. In our case the total occupancy is restricted to be less than one due to a large charging energy while such a condition is not necessary if it were for charging effects. Along this aspect, our problem is rather similar to those studied by Refs. 12 and 13 and their results for the tunneling currents are also available to obtain our final forms. However, we adopt the approach of Ref. 10 because it provides rather explicit expressions for the tunneling currents and the occupancies as well as is easily extended to the case of more charged states.

The constraint about the total occupancy is automatically imposed in the master equation in our problem. According to the formalism in Ref. 10, the master equation is expressed in terms of time-dependent tunneling rates. Within a sequential tunneling regime, the tunneling rates take into account state-dependent tunneling including spins and provide the associated selection rules via overlap matrices between states. Here, we neglect the selection rules in order to focus merely on time-dependent properties of the system. This is equivalent to the situation that the quantum dot has less-

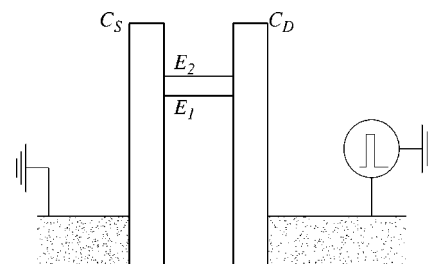


FIG. 1. A systematic drawing of a tunneling geometry is shown. Tunneling barriers are model with capacitance C_S and C_D , respectively, and a square pulsed field $V(t)$ is applied to the drain with respect to the source lead.

symmetrical shape to give the finite and nearly same overlaps between many-body states as well as has an even number of electrons in a ground state (we call it the vacuum state in the following section). By the even number of electrons we mean that the excitation by a single-electron tunneling is independent of spins and, therefore, releases the Kondo effects in our problem.¹⁴ By introducing simple forms of the tunneling rates associated with pulsed fields and separating them into adiabatic and nonadiabatic contributions, we derive analytic expressions for the evolution of the electron occupancy and the tunneling currents. Similar to Refs. 11–13, the nonadiabatic contribution gives rise to ringing currents which are resulted from the mismatch between incoming-electron energy and an energy level in the quantum dot. For easy comparison with experimental measurements we also propose a simple analytic form for a static component of the tunneling currents as a function of pulse amplitude and durations. It is found that the expression is appropriate to reveal detailed electronic structure of the system and its dynamical behavior such as the time evolution of the electron occupancy. For convincing ones of our results, numerical examples concerning about a typical situation are given.

II. THE MODEL AND CALCULATIONAL METHODS

As shown in Fig. 1, we consider a small quantum dot weakly coupled via tunnel barriers with capacitances C_D and C_S to electron reservoirs, called source and drain. By a small quantum dot we mean well resolved single-particle states in it due to the electronic confinement. Then, for not too large perturbations, it is a good approximation to consider several lowest energy states for tunneling. We denote energies of the single-particle states by $E_k(k=1, 2, \dots)$ which are measured relative to the chemical potentials of the reservoirs at equilibrium. We also assume that these states are located at high positions over thermal ($k_B T = 1/\beta$) and tunneling broadening ($\hbar\gamma_D, \hbar\gamma_S$), where T is a temperature and γ_D (γ_S) bare tunneling rates of the source (drain) lead. Consequently, tunneling through these state is completely blocked in the absence of external perturbations.

As for external perturbations, we consider square pulses. Although it is expected that, in experiments, a shape of a pulse is distorted due to stray resistances and capacitances of external circuits, we assume here that an ideal square shape can be generated and transferred to the system, i.e., a voltage pulse $V(t)$ is modeled by a Heaviside step function $\theta(t)$

$$V(t) = V_0 \theta(t) \theta(\tau_p - t), \quad (1)$$

where V_0 is its amplitude and τ_p duration, respectively. The voltage pulse $V(t)$ is applied to the drain with a positive amplitude, so that at the resonant condition, the chemical potential of the source coincides to one of energy levels in the quantum dot for a pulse duration of τ_p .

Due to the rapid variation of external perturbation $V(t)$ beyond typical characteristic times of the system, for instance, inverse of the bare tunnel rates, usual tunneling

formula based on the orthodox theory are not adequate to analyze its time-dependent properties. Instead, tunneling rates incorporating nonadiabatic effects should be adopted. To do this, we use the formalism in Ref. 10 (the nonequilibrium approach done by Wingreen *et al.*¹¹ is also available, however with modified Green's functions in the central region). According to the formalism, the tunneling rate $\Gamma_k^{\alpha\pm}(t)$ of a barrier α through an energy level $|k\rangle$ is given by

$$\Gamma_k^{\alpha\pm}(t) = \gamma_\alpha \int_{-\infty}^{\infty} d\epsilon \left\{ 1 \mp \tanh \frac{\beta}{2} (\epsilon + E_k) \right\} \frac{\text{Re}}{2\pi\hbar} \times \int_{-\infty}^0 d\tau e^{\gamma_\alpha \tau - (ie/\hbar)\tau - (ie/\hbar)\int_{t+\tau}^t dt' v_\alpha(t')} \quad (2)$$

where superscripts \pm represent tunneling into (+) and from (−) the quantum dot, respectively. Here, a time-dependent function of $v_\alpha(t)$ is a voltage difference between the quantum dot and the lead α . In the case of Fig. 1, $v_\alpha(t)$ are given by,

$$\begin{pmatrix} v_D(t) \\ v_S(t) \end{pmatrix} = \begin{pmatrix} \frac{C_S}{C_S + C_D} \\ -\frac{C_D}{C_S + C_D} \end{pmatrix} V(t) = \begin{pmatrix} V_D \\ V_S \end{pmatrix} \theta(t) \theta(\tau_p - t). \quad (3)$$

The expression of Eq. (2) is derived by assuming sequential tunneling process resulted from very opaque tunneling barriers. So, for only static voltages applied, the tunneling rates are reduced to the widely used form,

$$\Gamma_k^{\alpha\pm}(t) = \gamma_\alpha f_{\text{FD}}^\pm(E_k + ev_\alpha + i\hbar\gamma_\alpha), \\ = \gamma_\alpha \left\{ \frac{1}{2} \mp \frac{1}{\pi} \text{Im} \psi_0 \left(\frac{1}{2} - \frac{\beta}{2\pi i} (E_k + ev_\alpha - i\hbar\gamma_\alpha) \right) \right\} \quad (4)$$

where $f_{\text{FD}}^\pm(E_k)$ are broaden Fermi-Dirac distribution functions for particles (+) and holes (−), respectively, and ψ_0 is a digamma function. Whereas, when a pulsed perturbation such as Eq. (1) is applied, the tunneling rates from Eqs. (2) and (3) become,

$$\Gamma_s^{\alpha\pm}(t) = \gamma_\alpha f_{\text{FD}}^\pm(E_k + ev_\alpha(t) - i\hbar\gamma_\alpha) \\ \pm \gamma_\alpha F(E_k + eV_\alpha - i\hbar\gamma_\alpha, E_k - i\hbar\gamma_\alpha, t) \quad (5)$$

where $F(z_1, z_2, t)$ is defined by,

$$F(z_1, z_2, t) = \frac{2}{\beta} \text{Re} \sum_{n=0}^{\infty} \frac{z_2 - z_1}{(U_n + z_1)(U_n + z_2)} D_n(z_1, z_2, t), \quad (6)$$

$$D_n(z_1, z_2, t) = \begin{cases} 0 & \text{for } t < 0 \\ e^{(U_n+z_1)t/i\hbar} & \text{for } 0 \leq t < \tau_p \\ e^{(U_n+z_2)(t-\tau_p)/i\hbar} (e^{-i(U_n+z_1)\tau_p/i\hbar} - 1) & \text{for } t \geq \tau_p \end{cases} \quad (7)$$

with $U_n = \pi(2n+1)/i\beta$. In the first term of Eq. (5), energy levels in the quantum dot are simply modulated with the applied bias voltage $v_\alpha(t)$ and thus the tunneling rates follow the applied voltages adiabatically. On the other hand, the second term is the newly generated contribution from the rapid time variation of applied voltages, say, nonadiabatic contribution. It is interesting to note that F is an oscillating function of time with a frequency $(E_k + eV_\alpha)/\hbar$ for $0 \leq t < \tau_p$ (i.e., a pulse is on) and E_k/\hbar for $t \geq \tau_p$ (i.e., a pulse is off), respectively. In addition, in each time section, these oscillations decay with about a rate of $\gamma_\alpha + \pi/\hbar\beta$, and thus F is saturated exponentially to a steady value. The oscillatory behavior is resulted from the mismatch between incoming-electron energy and a energy level in the quantum dot, and is a central property representing nonadiabatic effects (a “ringing” current found in Refs. 11–13 is also responsible for this oscillation).

In usual experimental situations, since energy levels E_k are enough high not to be occupied when a pulse is off, we restrict ourselves to the case of $E_k \gg \hbar\gamma_\alpha, 1/\beta$. Then, as soon as a pulse is off ($t \geq \tau_p$), F shows rapid oscillating behavior of a frequency of E_k/\hbar compared to a time scale of $1/(\gamma_\alpha + \pi/\hbar\beta)$, and therefore we may neglect its effects. As a result, the nonadiabatic contribution F has a nonzero value only when a pulse is on. In other words, for a positive pulse amplitude, tunneling into (from) the quantum dot through the drain barrier is not (always) possible. Then, the tunneling rate at the drain is simplified as,

$$\Gamma_k^{D+}(t) = 0, \quad \Gamma_k^{D-}(t) = \gamma_D \quad (8)$$

irrelevant of time.

With the time-dependent tunneling rates of Eq. (5), the occupation of electrons in the quantum dot is determined by the master equation,

$$\begin{aligned} \frac{dP_1(t)}{dt} &= \sum_\alpha \{ \Gamma_1^{\alpha+}(t)P_0(t) - \Gamma_1^{\alpha-}(t)P_1(t) \} + wP_2(t), \\ \frac{dP_2(t)}{dt} &= \sum_\alpha \{ \Gamma_2^{\alpha+}(t)P_0(t) - \Gamma_2^{\alpha-}(t)P_2(t) \} - wP_2(t), \end{aligned} \quad (9)$$

where $P_k(t)$ and P_0 are the occupation probabilities to a state $|k\rangle$ and the vacuum state $|0\rangle$, respectively. Here, terms in the parenthesis describe usual changes of the probability $P_k(t)$ by the tunneling process.^{15–19} The last terms are responsible for the relaxation of electronic states due to the electron-phonon interaction from the state $|2\rangle$ to $|1\rangle$, in which we model its relaxation rate by a constant value, w for simplicity.⁸ Using the fact that $P_0(t) + P_1(t) + P_2(t) = 1$ and $\Gamma_k^{\alpha+}(t) + \Gamma_k^{\alpha-}(t) = \gamma_\alpha$ from Eq. (2), the master equation is further simplified as

$$\frac{dP_1(t)}{dt} = \sum_\alpha \Gamma_1^{\alpha+}(t)(1 - P_2) - \gamma P_1 + wP_2,$$

$$\frac{dP_2(t)}{dt} = \sum_\alpha \Gamma_2^{\alpha+}(t)(1 - P_1) - \gamma P_2 - wP_2, \quad (10)$$

with $\gamma = \gamma_D + \gamma_S$.

These coupled differential equations do not give analytic solutions generally, therefore, we solve them in a numerical way as in the following section. However, approximated solutions can be inferred. To do this, we divide the solutions into two parts from the adiabatic tunneling rates (P_k^A) and the nonadiabatic ones (P_k^N) of Eq. (5); $P_k(t) = P_k^A(t) + P_k^N(t)$. Then, the solution of $P_k^A(t)$ is given by, while a pulse is on,

$$P_k^A(t) = P_k^0 + \frac{1}{\gamma_0} (\Gamma_k^0 - \gamma_p P_k^0) e^{-\gamma_m t} - \frac{1}{\gamma_0} (\Gamma_k^0 - \gamma_m P_k^0) e^{-\gamma_p t}, \quad (11)$$

where $\Gamma_k^0 = \gamma_S \text{Re } f_{\text{FD}}^+(E_k + eV_S - i\hbar\gamma_S)$ is the tunneling rate into the quantum dot through the source with a static voltage equal to a pulse amplitude, and P_k^0 is corresponding occupation probabilities,

$$P_1^0 = \frac{\Gamma_1^0(\gamma + w - \Gamma_2^0) + w\Gamma_2^0}{\gamma_m \gamma_p}, \quad P_2^0 = \frac{\Gamma_2^0(\gamma - \Gamma_1^0)}{\gamma_m \gamma_p} \quad (12)$$

together with $\gamma_p = \gamma + (w + \gamma_0)/2$, $\gamma_m = \gamma + (w - \gamma_0)/2$, and $\gamma_0 = \sqrt{4\Gamma_1^0\Gamma_2^0 - 4\Gamma_2^0w + w^2}$. According to this result, the adiabatic part of the occupation $P_k^A(t)$ starts with a zero at a time $t = 0$ and exponentially approaches to a static value of P_k^0 as time elapses. By the presence of the phonon relaxation ($w \neq 0$), the lower state becomes more rapidly saturated to a larger value while the occupation to the upper is more reduced. On the other hand, the nonadiabatic contribution of $P_k^N(t)$ can be obtained with a simple perturbed method because the associated tunneling rates have appreciable value of Γ_k^0 only in the small range of time ($0 < t \ll 1/\gamma$) and thus it is expected to have a small value much less than one. Thus, by substituting $P_k^A(t)$ instead of $P_k(t)$ in the right-hand side of Eq. (10), we obtain,

$$P_1^N(t) = \gamma_S \int_0^t dt' F(E_1 + eV_S - i\hbar\gamma_S, E_1 - i\hbar\gamma_S, t') \{1 - P_2^A(t')\},$$

$$P_2^N(t) = \gamma_S \int_0^t dt' F(E_2 + eV_S - i\hbar\gamma_S, E_2 - i\hbar\gamma_S, t') \{1 - P_1^A(t')\}. \quad (13)$$

Further simplification of these occupations can be made using Eqs. (7) and (11), however, still in a series form. It is found that the nonadiabatic contribution $P_k^N(t)$ exhibits weak oscillatory behavior near $t=0$ and has a small saturated value much less than one. This fact is easily deduced if one recalls oscillatory and decaying properties of $F(z_1, z_2, t)$ together with its integrals of Eq. (13).

When a pulse off, the occupation probabilities have a simple solution because, in this time section, particles in the quantum dot just escape from there to both leads. That is, $\Gamma_k^{\alpha+} = 0$ and $\Gamma_k^{\alpha-} = \gamma_\alpha$. Thus, the probabilities become

$$P_1(t) = e^{\gamma(\tau_p - t)} \{P_1(\tau_p) + P_2(\tau_p)(1 - e^{w(\tau_p - t)})\} \\ P_2(t) = e^{(\gamma+w)(\tau_p - t)} P_2(\tau_p), \quad (14)$$

to exhibit decaying behavior as time elapses.

As for tunneling currents, since an external perturbation is time-dependent, it consists of two components; displacement and tunneling currents. If a positive current at each lead is defined to flow into the quantum dot, currents $I_\alpha(t)$ flowing in the lead α are given by,⁴

$$\begin{pmatrix} I_D(t) \\ I_S(t) \end{pmatrix} = \frac{C_D C_S}{C_D + C_S} \frac{\partial V(t)}{\partial t} \begin{pmatrix} 1 \\ -1 \end{pmatrix} - \frac{I_D^t + I_S^t}{C_D + C_S} \begin{pmatrix} C_D \\ C_S \end{pmatrix} + \begin{pmatrix} I_D^t \\ I_S^t \end{pmatrix}. \quad (15)$$

Here, the first two terms are the displacement currents induced by external perturbations and tunneling processes, respectively, whereas the last term is the tunneling contribution. Due to the charge conservation, the total currents are conserved irrelevant of time, i.e., the sum of total currents ($I_D + I_S$) is zero. A tunneling part $I_\alpha^t(t)$ represents the average number of electrons tunneled from the quantum dot per an unit time through the barrier α , which are given by, in terms of the tunneling rates and the occupation probabilities,

$$I_\alpha^t(t) = e \sum_s \{\Gamma_s^{\alpha-}(t) P_s(t) - \Gamma_s^{\alpha+}(t) P_0(t)\}. \quad (16)$$

As in the orthodox theory,¹⁵⁻¹⁹ each term describes the total number of electrons tunneling out of or into the quantum dot.

Now, we focus on a static component of currents because it is easier to measure in experiments compared to alternating components. If a pulse repetition time is τ_r , from Eq. (15) the dc current is obtained as,

$$I_{dc} = \frac{1}{\tau_r(C_S + C_D)} \int_0^{\tau_r} dt \{C_S I_D^t(t) - C_D I_S^t(t)\} \equiv e \langle N \rangle / \tau_r, \quad (17)$$

where we assume a pulse sequence with a long pulse-off region compared to the inverse of tunneling rates, so that the system is always in equilibrium before another pulse arrives. In other words, this means each pulse is considered as independent one and $\langle N \rangle$ can be interpreted as the average num-

ber of electrons transferred from the source to the drain per a single pulse.

In order to analyze dynamical behavior of the system, it is more instructive to measure the dc current as a function of a pulse duration and amplitude, and consider another dc current defined by,

$$i_{dc}(\tau_p) = e \frac{d\langle N \rangle}{d\tau_p} = \frac{dI_{dc}\tau_r}{d\tau_p}, \quad (18)$$

which is found to give a simpler expression for tunneling currents than Eq. (17) does. To obtain a final result, we separate time into pulse-on and -off regions in Eq. (17) and then apply the derivative with respect to the pulse duration to get,

$$i_{dc}(\tau_p) = \frac{1}{C_D + C_S} \{C_S I_D^t(t) - C_D I_S^t(t)\}_{t=\tau_p} \\ + \frac{\gamma_D C_S - \gamma_S C_D}{\gamma(C_S + C_D)} \frac{d}{dt} \{P_1 + P_2\}_{t=\tau_p}, \quad (19)$$

where a sufficiently long pulse-off region is taken into account. This relation is further simplified by using Eqs. (8), (10), and (16), and finally we obtain

$$i_{dc}(\tau_p) = \frac{e\gamma_D}{\gamma} \{\Gamma_1^{S+}(\tau_p)[1 - P_2(\tau_p)] + \Gamma_2^{S+}(\tau_p)[1 - P_1(\tau_p)]\}. \quad (20)$$

This is a main result in our work for pulsed responses of quantum dots. For a large pulse duration, the above expression gives the same results as those in a static case because tunneling rates and occupation probabilities approach to their static values, Γ_k^0 and P_k^0 , respectively. However, it is noted that for a small pulse duration ($\tau_p \ll \gamma$) the current $i_{dc}(\tau_p)$ shows different behavior from that in the static case. Let us assume a small pulse duration in which the occupation probabilities are negligible. Then, the current $i_{dc}(\tau_p)$ depends on tunneling rates through a source directly,

$$i_{dc}(\tau_p) = \frac{e\gamma_D}{\gamma} \{\Gamma_1^{S+}(\tau_p) + \Gamma_2^{S+}(\tau_p)\}. \quad (21)$$

This expression also holds for small pulse heights well below the lowest level which may give negligible occupations. From this result, one can estimate time-dependent tunneling rates of each energy level through the source by adjusting a pulse amplitude. For a pulse duration longer than the bare tunneling rate γ , the time-dependent tunneling rates become their static values of Γ_k^0 and the adiabatic contribution of the occupation probabilities is dominant. So, in this case the current i_{dc} becomes,

$$i_{dc}(\tau_p) = \frac{e\gamma_D}{\gamma} \{\Gamma_1^0[1 - P_2^A(\tau_p)] + \Gamma_2^0[1 - P_1^A(\tau_p)]\}. \quad (22)$$

Using this, one can see that the current as a function of time directly reflect the evolution of the occupation probabilities in the quantum dot. So, by fitting data measured in the whole range of a pulse duration to the above current expressions, it is possible to estimate the occupation probability of each level. Furthermore, comparing this evolution with tunneling

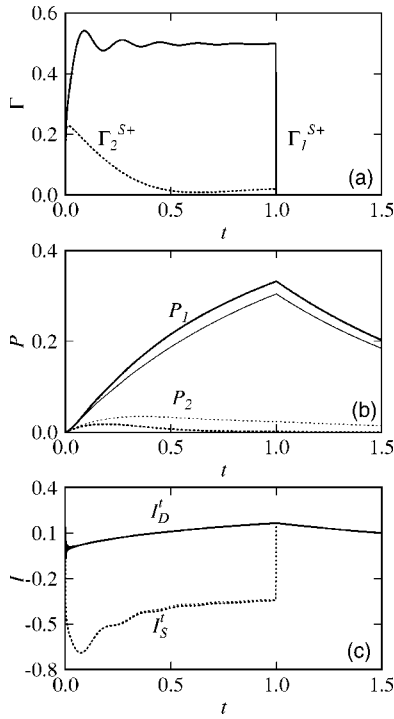


FIG. 2. For $\gamma_D = \gamma_S = 0.5\gamma$ and $V_0 = 2070\hbar\gamma/e$, we plot the tunneling rates available to each level in (a). For electron-phonon relaxation rates of $w=0$ (thin), 5γ (thick line), occupation probabilities and tunneling currents are shown as a function of time in (b) and (c), respectively. Here, a pulse width τ_p are assumed to be $1/\gamma$.

rates obtained from measured data for short pulse durations, one may determine the electron-phonon relaxation rate w of the system.

III. NUMERICAL RESULTS AND DISCUSSION

For better understanding of our results, we now examine tunneling currents numerically. For this, we consider a typical case of the system satisfying conditions addressed in the previous section; in the units of tunneling rate, $\gamma = \gamma_D + \gamma_S$, two energy levels in the quantum dot are assumed to be located at $E_1 = 1000\hbar\gamma$ and $E_2 = 1040\hbar\gamma$, respectively, at a temperature of $T = \hbar\gamma/k_B$. Then, if capacitances of barriers are equal to each other ($C_D = C_S$), for pulse heights of $V_0 = 2000$, and $2080\hbar\gamma/e$, the chemical potential of the source coincide with the lower and upper energy level, respectively, while a pulse is on. (The above numbers should be taken as suggestive estimates; our scheme is general and does not depend on these specific values.)

First, in Fig. 2(a), we plot the tunneling rates of Eq. (5) as a function of time when a square pulse with a duration of $\tau_p = 1/\gamma$ and a amplitude of $V_0 = 2070\hbar\gamma/e$ is applied at $t=0$. Similar to the shape of an applied pulse, the tunneling rates start with a zero and have an infinite slope at $t=0$. As expected in the previous section, while a pulse is on, they shows oscillating behavior about an adiabatic value of $\gamma_\alpha \text{Re} f_{\text{FD}}^+(E_k + eV_\alpha - i\hbar\gamma_\alpha)$ with a frequency of $(E_k + eV_\alpha)/\hbar$. Simultaneously the oscillations are decayed with a rate of $\gamma_\alpha + \pi/\hbar\beta$, so that in the inverse of this rate the oscillating behavior is nearly disappeared.

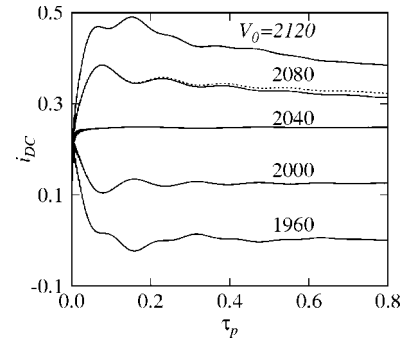


FIG. 3. For a symmetric geometry $C_D = C_S$ and $\gamma_D = \gamma_S$, the currents i_{dc} (defined in the text) are plotted as a function of pulse duration for several pulse amplitudes. The dotted lines represent results in the presence of the energy relaxation of $w=5\gamma$, however, in the most cases they are not resolved from those of $w=0$ except for $V_0=2080$. Here, currents and tunneling rates are measured in units of $e\gamma$ and γ , respectively.

The occupations are monotonically increasing functions while a pulse is on and then decayed in the absence of a pulse as shown Fig. 2(b). Overall patterns are mainly resulted from the adiabatic contributions of Eq. (11) and the contribution from the nonadiabatic term is found to be negligibly small. Effects of the electron-phonon relaxation from $|2\rangle$ to $|1\rangle$ are well resolved in the occupation probabilities. As indicated in Eqs. (11), (12), and (14), more rapid increase of the occupation $P_1(t)$ is found compared with those for $w=0$ while $P_2(t)$ shows slower increase and a smaller saturated value as shown with a thick-dotted line in Fig. 2(b).

In Fig. 2(c), we plot tunneling currents at the drain and source as a function of time. It is found that time-dependent behavior is largely different at the source and drain when a pulse is on although their total currents of Eq. (15) are equal to each other. This result is caused by different time-dependence of tunneling rates at the source and drain. In fact, the detuning energies of $eV_\alpha + E_k$ are very different between both electrodes. In the case of the drain, since the detuning energy is very large, the nonadiabatic contribution to its tunneling rate can be neglected and thus is independent of time. So, its time-dependent behavior comes from the variation of the occupation probabilities of electrons in the quantum dot. Whereas, at the source, since the detuning energy is relatively small, the nonadiabatic contribution of the tunneling rates is appreciable. Thus the time-dependence of tunneling currents is determined from both the tunneling rates and the occupation probabilities, in which, however, ringing behavior is mainly responsible for the tunneling rates. When a pulse is off, tunneling currents decrease exponentially because only electrons escaping from the quantum dot contribute to the currents. Since the same tunneling rate of $\gamma_D = \gamma_S$ are used in Fig. 2, the tunneling currents at the drain and source are calculated to be equal to each other in this time region.

In Fig. 3, we plot the current i_{dc} as a function of a pulse duration for several amplitudes. As expected in the previous section, the currents i_{dc} shows characteristic oscillating behavior resulted from the time-dependence of tunneling rates, where frequencies of the oscillations are equal to the detun-

ing energy of the source, $eV_S + E_k$ and thus the oscillations are modulated with two frequencies of $(E_1 + eV_S)/\hbar$ and $(E_2 + eV_S)/\hbar$ for a given amplitude of a pulse. As special cases, when one of the detuning energy becomes zero (at $V_0 = 2000$ and 2080 in Fig. 3) the chemical potential of the source coincides with the lower or upper level, and the current i_{dc} oscillates a single frequency of $(E_2 - E_1)/\hbar$. In addition, when the chemical potential is located at the middle of two levels (at $V_0 = 2040$ in Fig. 3), no oscillations are found and the current i_{dc} is rapidly saturated to its static value. This is because the tunneling rates of Γ_1^{S+} and Γ_2^{S+} oscillate out of phase to each other.

We also examine the dc current i_{dc} for a finite electron-phonon relaxation rate w . The dotted lines in Fig. 3 show results of the current i_{dc} when the relaxation rate w is much larger than the tunneling rate ($w = 5\gamma$), so that the occupation of the upper level is largely suppressed. It is found that the largest modification occurs at the pulse height where the chemical potential of the source lies at the upper level (at $V_0 = 2080$ in Fig. 3). Otherwise, the relaxation rate w gives rise to a small modification to i_{dc} .

To model the currents i_{dc} , we compare in Fig. 4 the calculated results with simplified expressions of Eqs. (21) and (22). For a small pulse height such as $V_0 = 1960$ in Fig. 4(a), it is well fitted to Eq. (21) (its difference is not resolved in the figure) because the occupation probabilities are nearly zero. However, for the case of $V_0 = 2080$, it is more convenient to use Eq. (22) in the range of the long pulse duration [the dotted line in Fig. 4(b)]. Consequently, from two complementary fitting results we expect that it is possible to obtain a bare tunneling rate γ_α at each barrier and the electron-phonon relaxation rate w , as well as dynamical properties of electrons in the quantum dot including energy levels. For general cases such as an asymmetric barrier geometry and more energy levels, similar arguments can be made and it is expected that measured data can be fitted to.

In summary, we investigate time-dependent responses of a quantum dot coupled weakly to metallic leads by pulsed

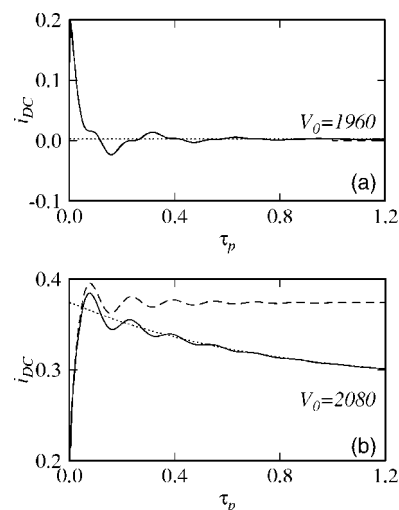


FIG. 4. We compare calculated results (solid lines) of Fig. 3 with the current expressions of Eqs. (21) and (22). For pulse amplitudes of $V_0 = 2080, 1960\hbar\gamma/e$ in (a) and (b), respectively, approximated currents are represented by dashed and dotted lines. In the case of (a), the difference between solid and dashed lines are not resolved.

fields. Treating the problem in a non-adiabatic regime, we show that dc tunneling currents as a function of a pulse duration contain detailed dynamical behavior of the system. We also present simple expressions for the dc current, by which one can get particle occupations as a function of time as well as various system parameters by fitting experimental data to them.

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