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# Time-dependent electron transport through molecular quantum dots in the presence of external irradiation

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**Abstract**

We present a fully nonequilibrium calculation of the low-temperature transport properties of a single molecular quantum dot coupled to the local phonon mode when an ac field is applied to the gate. The resonant behaviour is shown in the time-averaged differential conductance as the ac frequency matches the frequency of the local phonon mode, which is a direct consequence of the satellite-phonon-peak structure in the dot electron spectral function. Different step structure with and without the external irradiation is found in the $I$–$V$ curves, and oscillation behaviour is found in the step height as a function of the irradiation intensity.

(Some figures in this article are in colour only in the electronic version)

Recent advances in nanotechnology have allowed the fabrication of very small molecular quantum dots coupled to the macroscopic charge reservoirs (leads) [1]. In contrast to the semiconductor dot, which is quite rigid in space, the molecules involved in the electron tunnelling process naturally possess the vibrational degrees of freedom which will inevitably react to the transform of electrons through the molecular quantum dots [1]. It was first observed in a single $C_{60}$ molecule that quantum mechanical behaviour of the centre of mass oscillation of a $C_{60}$ can be excited by the electrons tunnelling from electrodes and the signs of vibrational sidebands were shown in transport through a single molecule [1]. Similar vibrational sideband structures were found in other more complicated molecules. In addition to the importance in molecular-scale electronics from the application point of view, these artificial, tunable devices are potentially important for understanding the basic physics including the many-body effect [2]. Theoretically, a lot of effort has been focused on the quantum conductance of molecular systems based on the kinetic equation approach [3], the rate equation approach [4], the correlation effects [5], the nonequilibrium quantum theory [6–9], the strong coupling to environment [10], the numerical renormalization group calculation [11], and more recently a new proposal to decouple the electron–phonon interaction from the total electron system [12].

So far, the stationary quantum transport through the molecular dots has been considered, while the influence of a time-dependent ac field on the current has not been well addressed.

Irradiation of a quantum dot with an ac field [13–15] offers a new way of affecting its dynamics, which enables one to study the effect of electron–phonon interaction on the transport phenomenon of molecular dots in an essentially nonequilibrium condition.

In this paper we use the Keldysh nonequilibrium Green function technique to study the nonlinear ac transport through a single-molecular quantum dot coupled to a local phonon mode with the external irradiation applied to the gate. After a canonical transformation, we obtain a formula for the time-dependent current in general terms of bias, temperature, the intensity and frequency of the external ac field, and the electron–phonon coupling. We show that the satellite-peak structure in the dot electron spectral function due to the electron–phonon interaction can be probed by imposing on top of the gate bias an ac bias voltage. The satellite-peak structure in the dot electron spectral function gives rise to resonant behaviour in the time-averaged current as the ac frequency matches the frequency of local phonon mode, which can be observed directly in experiments. The calculated $I$–$V$ curves show the different step structures with and without the external irradiation, respectively, and the step height shows the Bessel-type oscillation behaviour as a function of irradiation intensity.

To understand the basic physics of effect of electron–phonon interaction on the time-dependent transport through a single-molecular quantum dot, in this work we consider a simplest Holstein-type model with a single phonon mode is employed to address the vibrational degrees of freedom in the molecular dot. The single-molecular quantum dot is assumed to have only one single-particle energy level (measured with respect to the zero-bias Fermi energies of the two leads) connected to two noninteracting electronic reservoirs via the tunnelling barriers. The two reservoirs remain in local equilibrium with the Fermi distribution, and each has only one electronic channel. All other complexity of real molecular devices, apart from the interaction with the phonon mode, is ignored. Moreover, the electron–phonon interaction takes place solely in the central molecular quantum dot, while the high-order exchange between electrons and phonons and the damping effect of vibration by coupling to the environment [10] is neglected. We also assume that only the single-mode Einstein phonon is directly coupled to the electron in the single-molecular quantum dot. Moreover, we do not include the on-site electron–electron Coulomb interaction in quantum dots, which is crucially important to the Coulomb blockade and the many-body Kondo effect at low temperatures, to avoid further complications.

Then the system Hamiltonian can be written as

$$H = H_{\text{leads}} + H_{\text{X}} + H_{\text{D}} + H_{\text{T}}, \tag{1}$$

where the subscripts leads, X, D and T stand for leads, local phonon mode, dot and tunnelling coupling between dot and leads, respectively. Here,

$$\begin{aligned} H_{\text{leads}} &= \sum_{k,\eta,\sigma} \epsilon_{k\eta} c^{\dagger}_{k\eta\sigma} c_{k\eta\sigma}, \\ H_{\text{X}} &= \omega_0 a^{\dagger} a, \\ H_{\text{D}} &= \sum_{\alpha} \left[\epsilon_0(t) + \lambda \left(a + a^{\dagger}\right)\right] d^{\dagger}_{\alpha} d_{\alpha}, \\ H_{\text{T}} &= \sum_{k,\eta,\sigma,\alpha} \left[V^{\eta}_{k\sigma,\alpha} c^{\dagger}_{k\eta\sigma} d_{\alpha} + \text{H.C.}\right]. \end{aligned} \tag{2}$$

$c^{\dagger}_{k\eta\sigma}(c_{k\eta\sigma})$ are creation (annihilation) operators for the noninteracting electrons with momentum $k$ and spin index $\sigma$ in the left ($\eta = \text{L}$) or right ($\eta = \text{R}$) metallic leads. $\omega_0$ is the frequency of the single phonon mode, and $a^{\dagger}$ ($a$) is the phonon creation (annihilation) operator. $H_{\text{D}}$ describes the electron in the molecular quantum dot coupled to the local phonon mode with the coupling constant $\lambda$, where $d^{\dagger}_{\alpha}$ ($d_{\alpha}$) is the dot–electron creation (annihilation)

operator, and $\epsilon_0(t)$ is the single energy level of the dot, which can be tuned by the external irradiation, $\epsilon_0(t) = \epsilon_0 + V_r \cos(\omega_r t)$ for harmonic bias. Here we assume that the metallic leads are dc biased, neglecting the possible 'leakage' of the irradiating ac field to the leads. The generalization to the case of nonzero ac bias is straightforward. $H_T$ describes the tunnelling coupling between dot and leads, where the tunnelling matrix elements $V^{\eta}_{k\sigma,\alpha}$ transfer electrons through an insulating barrier out of the dot. We emphasize that when the inelastic processes involving phonon emission and absorption are considered when electron tunnelling through the single-molecular quantum dot, the nonequilibrium electron transport governed by Hamiltonian equation (1) becomes a strongly correlated many-body problem which cannot be solved exactly. Therefore, the usual perturbation theory, which has been successfully applied in studying the conventional transport problems in bulk materials, is invalid in treating the nonequilibrium electron transport in a single-molecular quantum dot with strong electron–phonon interaction.

Based on the Keldysh nonequilibrium Green function formalism [16], the time-dependent current from the $\eta$ lead to the dot is given by [13]

$$J_\eta(t) = -\frac{2e}{\hbar}\sum_\alpha \int_{-\infty}^{t} dt' \int \frac{d\epsilon}{2\pi} \mathrm{Im}\left\{e^{i\epsilon(t-t')}\Gamma^{\eta}_{\alpha}\left[G^{<}_{\alpha\alpha}(t,t') + f_\eta(\epsilon)\, G^{r}_{\alpha\alpha}(t,t')\right]\right\}, \tag{3}$$

where $f_{L(R)}(\epsilon)$ are the Fermi distribution functions of the left (right) leads, which have different chemical potentials upon a dc bias voltage $\mu_L - \mu_R = eV$. $\Gamma^{\eta}_{\alpha} = 2\pi\rho_\alpha(0)|V^{\eta}_{k\alpha,\alpha}|^2$ characterizes the tunnelling coupling between dot and leads, and $\rho_\alpha(0)$ is the spin-$\alpha$ band density of states in the leads. Here we assume that the leads give rise to a flat, energy independent, density of states (i.e., the wide-band limit). $G^{r(<)}$ is the retarded (Keldysh less) Green function of quantum dot. It is noted that the general formula of time-dependent current through central interacting dot coupled to two leads was derived by Jauho *et al* [13], who expressed the current in terms of the distribution functions in the leads and the local properties of the central dot, such as the occupation and density of states (i.e., via the retarded and lesser Green functions of dot electrons).

In order to compute the time-dependent current, one has to compute the dot electron Green functions in the presence of both the electron–phonon interaction and the tunnelling coupling between dot and leads. The problem described by the Hamiltonian equation (1) is a many-body problem involving the phonon emission and absorption when the electron tunnels through the central quantum dot. In the case of strong electron–phonon coupling, the usual perturbation theory is invalid when dealing with this nonequilibrium transport problem. The Green function can be calculated by performing the nonperturbative canonical transformation $\mathcal{S} = (\lambda/\omega_0)\sum_\alpha d^{\dagger}_{\alpha} d_\alpha (a^{\dagger} - a)$ [17], and then the dot level is renormalized to $\epsilon_0 - \Delta$, where $\Delta = \lambda^2/\omega_0$, and the tunnelling coupling term is also renormalized as $\overline{H}_T = \sum_{k,\eta,\sigma,\alpha}[V^{\eta}_{k\sigma,\alpha} c^{\dagger}_{k\eta\sigma} d_\alpha X +$ H.C.], where $X = \exp[-(\lambda/\omega_0)(a^{\dagger} - a)]$. Ignoring the effects of narrowing the bands of leads due to the phonons [18], the dot–electron Green function can be decoupled as $G^{r}_{\alpha\alpha'}(t,t') = \tilde{G}^{r}_{\alpha\alpha'}(t,t')\langle X(t)X^{\dagger}(t')\rangle_{ph}$, where $\tilde{G}^{r}_{\alpha\alpha'}(t,t') = -i\Theta(t-t')\langle\{\tilde{d}_\alpha(t), \tilde{d}^{\dagger}_{\alpha}(t')\}\rangle_{el}$, $\tilde{d}_\alpha(t) = e^{i\overline{H}_{el}t} d_\alpha e^{-i\overline{H}_{el}t}$, $\overline{H}_{el} = H_X$, and $X(t) = e^{i\overline{H}_{ph}t} X e^{-i\overline{H}_{ph}t}$. The renormalization factor due to the electron–phonon interaction is [17] $\langle X(t)X^{\dagger}(t')\rangle_{ph} = e^{-\Phi(t-t')}$, where $\Phi(t) = (\lambda/\omega_0)^2[N_{ph}(1 - e^{i\omega_0 t}) + (N_{ph}+1)(1 - e^{-i\omega_0 t})]$, and $N_{ph} = 1/[\exp(\beta\omega_0) - 1]$. The retarded Green function can be easily obtained by the standard Dyson equation approach [13], and the result is

$$G^{r}_{\alpha\alpha'}(t,t') = -i\delta_{\alpha\alpha'}\Theta(t-t')\, e^{-i\int_{t'}^{t} d\tau V_r \cos(\omega_r\tau)} \times e^{-i[\epsilon_0 - \Delta - \frac{i}{2}\Gamma_\alpha](t-t') - \Phi(t-t')}, \tag{4}$$

where $\Gamma_\alpha = \Gamma^{L}_{\alpha} + \Gamma^{R}_{\alpha}$ is the total tunnelling coupling to the leads. Following the operational rules [19] to the Dyson equation for the contour-ordered Green function, the Keldysh Green

function is found to be

$$G^{<}_{\alpha\alpha'}\left(t,t'\right)=\delta_{\alpha\alpha'}\int \mathrm{d}t_1\int \mathrm{d}t_2\,\tilde{G}^{\mathrm{r}}_{\alpha\alpha}\left(t,t_1\right)\Sigma^{<}_{\alpha}\left(t_1,t_2\right)\tilde{G}^{\mathrm{a}}_{\alpha\alpha}\left(t_2,t'\right), \tag{5}$$

with the lesser self-energy

$$\Sigma^{<}_{\alpha}\left(t_1,t_2\right)=\mathrm{i}\sum_{\eta}\int\frac{\mathrm{d}\epsilon}{2\pi}\Gamma^{\eta}_{\alpha}\mathrm{e}^{-\Phi[-(t_1-t_2)]}f_{\eta}\left(\epsilon\right)\mathrm{e}^{-\mathrm{i}\epsilon(t_1-t_2)}. \tag{6}$$

Without the electron–phonon interaction, the above result fully agrees with that for time-dependent transport through a noninteracting quantum dot [13].

Substitution of the Green function equations (4)–(6) into equation (3) gives

$$J_{\eta}(t)=-\frac{e}{\hbar}\sum_{\alpha}\Gamma^{\eta}_{\alpha}\int\frac{\mathrm{d}\omega}{2\pi}\left\{2f_{\eta}(\omega)\,\mathrm{Im}\left[A_{\alpha}\left(\omega,t\right)\right]-2\left(\sum_{\eta}\Gamma^{\eta}_{\alpha}f_{\eta}(\omega)\right)\right.$$
$$\left.\times\int_{-\infty}^{t}\mathrm{d}t_1\mathrm{e}^{-\Gamma_{\alpha}(t-t_1)}\,\mathrm{Im}\left[A_{\alpha}\left(\omega,t_1\right)\right]\right\}, \tag{7}$$

with $A_{\alpha}(\omega,t)=\int_{-\infty}^{t}\mathrm{d}t'\mathrm{e}^{\mathrm{i}\omega(t-t')}G^{\mathrm{r}}_{\alpha\alpha}(t,t')$. Obviously, in the time-independent case, $A_{\alpha}(\omega)$ is just the Fourier transform of the retarded Green function $G^{\mathrm{r}}_{\alpha\alpha}(\omega)$. After some algebra, we find that for this model

$$A_{\alpha}\left(\omega,t\right)=\mathrm{e}^{-g\left(2N_{\mathrm{ph}}+1\right)}\sum_{l,m,n}\left(-1\right)^{l+m}\mathrm{e}^{\mathrm{i}(l-m)\omega_{\mathrm{r}}t}\mathrm{e}^{n\omega_0\beta/2}$$
$$\times J_l\left(\frac{V_{\mathrm{r}}}{\omega_{\mathrm{r}}}\right)J_m\left(\frac{V_{\mathrm{r}}}{\omega_{\mathrm{r}}}\right)I_n\left\{2g\left[N_{\mathrm{ph}}\left(N_{\mathrm{ph}}+1\right)\right]^{1/2}\right\}$$
$$\times\frac{1}{\omega-\left(\epsilon_0-\Delta\right)+m\omega_{\mathrm{r}}-n\omega_0+\frac{\mathrm{i}}{2}\Gamma_{\alpha}}, \tag{8}$$

where parameter $g=(\lambda/\omega_0)^2$, and $J_m(z)$ is the Bessel function of the $m$th order, $I_n(z)$ is the Bessel function of complex argument, and $l$, $m$, $n=0,\pm1,\pm2,\ldots$. Equation (8) together with the current expression equation (7) provides the complete solution to the time-dependent transport of the molecular quantum dot coupled to the local phonon mode in the presence of ac field. Although equation (7) looks similar to the result of time-dependent current equation (65) in [13], which is valid only in the case of proportional tunnelling coupling (i.e., $\Gamma^{\mathrm{L}}=\lambda\Gamma^{\mathrm{R}}$, where $\lambda$ is a constant), we remark that equation (7), which includes the contributions from both the retarded (equation (4)) and lesser (equations (5) and (6)) Green functions, is the general formula for time-dependent current in quantum dot with electron–phonon interaction in the presence of ac field. This general time-dependent current formula can be applied in the much more complicated case where the proportional coupling between the dot and the leads is not justified. It is easy to show that for the noninteracting case the above expression is just the Landauer–Büttiker formalism developed based on the scattering matrix theory. The connection between these two formalisms was first established by Jauho *et al* [13].

Experimentally, what is interesting is the current on a timescale long compared to $2\pi/\omega_{\mathrm{r}}$. Here we discuss the time-averaged current $\langle J(t)\rangle$, which could be directly relevant to experiment. For this model, we then obtain

$$\langle J_{\mathrm{L}}(t)\rangle=-\langle J_{\mathrm{R}}(t)\rangle=\frac{2e}{\hbar}\left(\frac{1}{2}\right)\mathrm{e}^{-g\left(2N_{\mathrm{ph}}+1\right)}\sum_{\alpha}\Gamma^{\mathrm{L}}_{\alpha}\Gamma^{\mathrm{R}}_{\alpha}\int\frac{\mathrm{d}\omega}{2\pi}$$
$$\times\left[f_{\mathrm{L}}(\omega)-f_{\mathrm{R}}(\omega)\right]\sum_{m,n}J^2_m\left(\frac{V_{\mathrm{r}}}{\omega_{\mathrm{r}}}\right)\times I_n\left(2g\left[N_{\mathrm{ph}}\left(N_{\mathrm{ph}}+1\right)\right]^{1/2}\right)\mathrm{e}^{n\omega_0\beta/2}$$
$$\times\frac{1}{\left[\omega-\left(\epsilon_0-\Delta\right)+m\omega_{\mathrm{r}}-n\omega_0\right]^2+\left(\frac{\Gamma_{\alpha}}{2}\right)^2}. \tag{9}$$

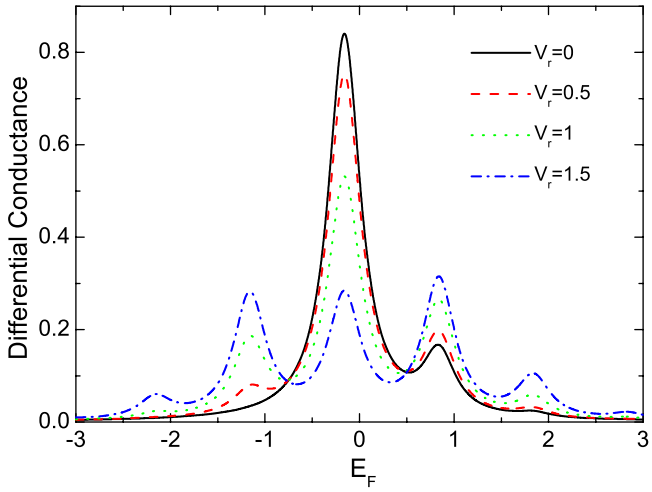


**Figure 1.** The time-averaged differential conductance, in units of $2e^2/h$ through the molecular quantum dot as a function of the Fermi energy $E_{\rm F}$ measured relative to the single level of the dot in the absence ($V_{\rm r} = 0$), and in the presence of the ac field with the frequency $\omega_{\rm r} = \omega_0$ and three different values of irradiation intensities $V_{\rm r} = 0.5\omega_0$, $\omega_0$, and $1.5\omega_0$, respectively. The energy is measured in units of the frequency of the phonon mode $\omega_0$, $\lambda = 0.4\omega_0$, $\epsilon_0 = 0$, and $\Gamma = 0.2\omega_0$.

In the absence of external irradiation, i.e., $V_{\rm r} = 0$, equation (9) fully agrees with the result of dc current in [8, 9]. The contribution of the external irradiation to the transport becomes significant when the argument of the Bessel function, $V_{\rm r}/\omega_{\rm r}$, is of the order of unity. Another important consequence is that the time-averaged current under irradiation is proportional to $J_m^2$. As a result, one should expect the change in current due to the external irradiation to depend on the intensity of irradiation.

For simplicity, we consider the tunnelling coupling between the molecular dot and the two leads to be symmetric and independent of the spin index, i.e., $\Gamma^{\rm L}_{\uparrow} = \Gamma^{\rm L}_{\downarrow} = \Gamma^{\rm R}_{\uparrow} = \Gamma^{\rm R}_{\downarrow} = \Gamma$. In figure 1, we plot the zero-temperature differential conductance as a function of the Fermi energy measured relative to the single level $\epsilon_0$ of the dot in the external ac field with frequency $\omega_{\rm r} = \omega_0$ and different values of irradiation intensities. For comparison, we also plot the differential conductance in the absence of ac field. At zero ac bias voltage, our result agrees well with that in [8, 9], where the electron–phonon coupling can lead to the satellite resonant peaks. Figure 1 shows that the ac field with frequency $\omega_{\rm r} = \omega_0$ can lead to the enhancement of satellite resonant peaks in the positive energy region and the appearance of new peaks in the negative energy region. One can also see that the main peak is suppressed by the irradiation while increasing the intensity. In figure 2, we plot the zero-temperature differential conductance as a function of the frequency of ac field for different values of irradiation intensities by fixing the Fermi energy of leads $E_{\rm F}$ as $E_{\rm F} - (\epsilon_0 - \Delta) = 0$ to avoid unnecessary complications. As the intensity of the ac field increases, resonant signals are clearly shown when the frequency of the ac field satisfies $m\omega_{\rm r} = n\omega_0$. The satellite-phonon-peak structure in the dot electron spectral function, as shown in [8, 9] and also in figure 1 of this paper, gives rise to resonant behaviour in the conductance as the irradiating ac frequency matches the frequency of local phonon mode, and can be observed directly in experiments. Figure 2 also shows that more resonant signals can be observed when increasing the irradiation intensity.

Figure 3 shows the calculated zero-temperature current–voltage curves with and without the external ac irradiation, respectively. Here we assume the leads to be symmetrically

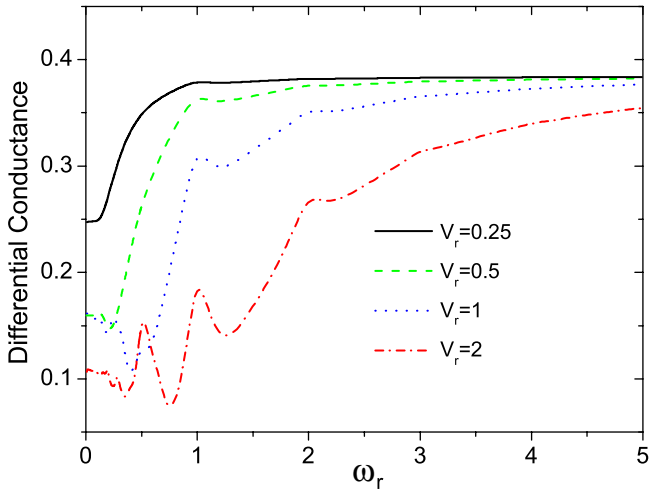


**Figure 2.** The time-averaged differential conductance, in units of $2e^2/h$ as a function of the frequency of irradiating ac field for different intensities of ac field with $V_r/\omega_0 = 0.25$, 0.5, 1, and 2, respectively. Here $\lambda/\omega_0 = 1$, $\epsilon_0 = 0$, $\Gamma = 0.2\omega_0$, and $E_F = (-1)\omega_0$.

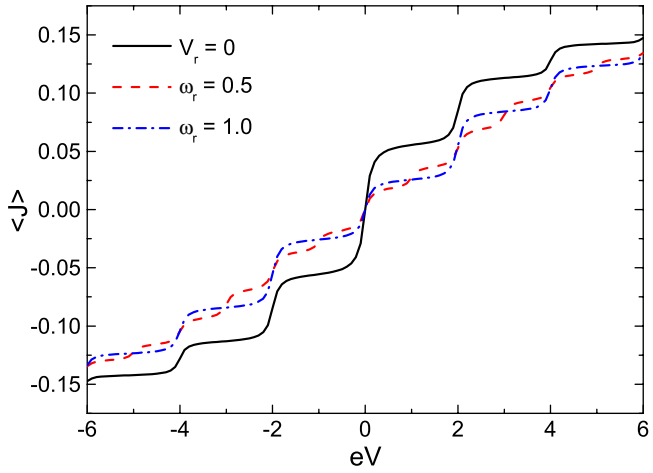


**Figure 3.** Current–voltage characteristics, in units of $2e/h$, without ($V_r = 0$) and with the ac field for the different irradiative frequencies: $\omega_r = 0.5\omega_0$ and $1\omega_0$. Here, $\lambda/\omega_0 = 1$, $E_F/\omega_0 = -1$, $\epsilon_0 = 0$, $V_r/\omega_0 = 2$, and $\Gamma = 0.05\omega_0$.

voltage biased, i.e., $+V/2$ on the left lead and $-V/2$ on the right one, to avoid unnecessary complications. In the absence of external ac field, clear steps appear at roughly $2\omega_0$ intervals in the weak tunnelling coupling limit, corresponding to $eV/2 = \pm n\omega_0$, with $n = 0, 1, 2, \ldots$, and the height of the $N$th step decreases with $N$, which can be easily understood from equation (10) (see below) by taking $V_r = 0$ for small electron–phonon interaction. In the presence of ac field with frequency $\omega_r = \omega_0$, steps appear at the same intervals $2\omega_0$ as those in the zero ac field, while the step height is modulated by the Bessel function due to the irradiation (see equation (10) below). Figure 3 also shows that more steps appear at the intervals $\omega_0$ in the case of $\omega_r = 0.5\omega_0$, corresponding to $eV/2 = m\omega_r - n\omega_0$ with $m = 0, \pm 1, \pm 2, \ldots$. The zero-temperature $N$th step's height in an external ac field with frequency $\omega_r = \omega_0$ can be

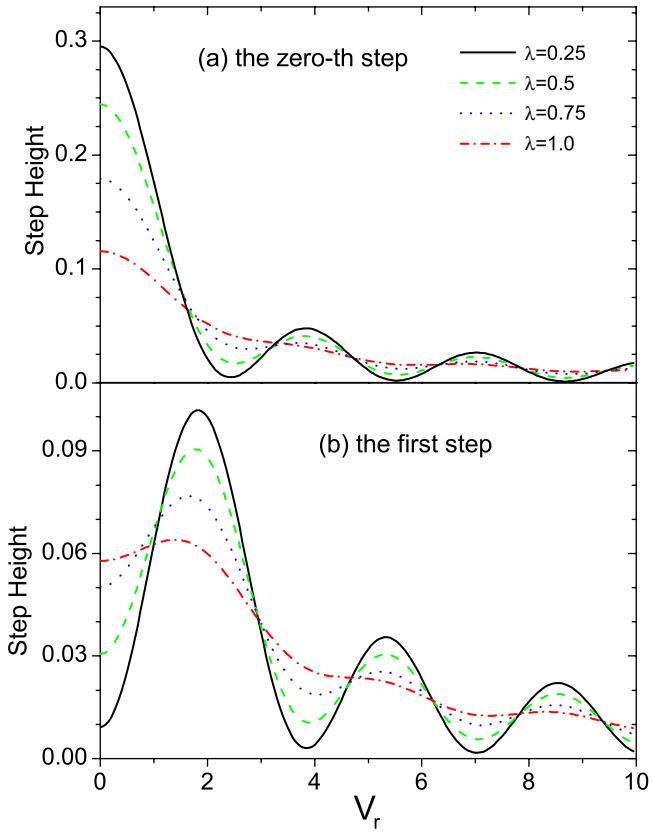


**Figure 4.** The height of the zeroth ($N = 0$) (a) and the first ($N = 1$) (b) step, in units of $2e/h$, as a function of the irradiation intensity by fixing the Fermi energy of the leads $E_F$ as $E_F - (\epsilon_0 - \Delta) = 0$. Here, $\omega_r = \omega_0$, and $\Gamma = 0.05\omega_0$.

analytically obtained from equation (9) at the fixed Fermi energy $E_F$ as $E_F - (\epsilon_0 - \Delta) = 0$,

$$\Delta J_N = \pi \left(\frac{2e}{h}\right) e^{-g} \sum_{\alpha} \frac{\Gamma_\alpha^L \Gamma_\alpha^R}{\Gamma_\alpha^L + \Gamma_\alpha^R} \sum_{n=0}^{\infty} \frac{g^n}{n!} \left[ J_{n+N}^2 \left(\frac{V_r}{\omega_r}\right) + J_{n-N}^2 \left(\frac{V_r}{\omega_r}\right) \right]. \tag{10}$$

In figure 4, we plot the height of the $N$th step, where $N = 0$ and 1, as a function of the irradiation intensity for different values of the electron–phonon coupling constant. The oscillation behaviour of the step height is clearly observed for the small electron–phonon interaction due to the external irradiation (see equation (10)), while this oscillation smears out for large electron–phonon interaction. The large electron–phonon interaction enhances the processes of absorption and emission of many ($n > 1$) phonons when the electron tunnels through the dot, and then the summation of Bessel function with large values of indices $n \pm N$ results in the smearing of the oscillation behaviour (see equation (10)).

For various molecular mesoscopic systems studied in experiments, a considerably wide range of phonon energy has been estimated from 0.01 to 10 meV [1, 2]. To experimentally investigate the resonant behaviour in time-dependent transport through molecular quantum dots, we suggest that the device is subject to a microwave irradiation field with the frequency ($\omega_r$) in the region of GHz to THz and the intensity with the same order ($V_r \sim \omega_r$). More recently, the photon-induced Kondo satellites have been observed experimentally in a single-

electron transistor irradiated with microwaves [15]. Experimental effort on a single-molecular transistor along this direction is in progress.

Summarizing, using the Keldysh nonequilibrium Green function technique, we have studied the time-dependent transport through a single-molecular quantum dot coupled to a local phonon mode in the presence of an external ac field. As an example, we have considered resonant electron tunnelling through a single level coupled to a single phonon mode with the external irradiation applied to the gate. This model without the ac irradiation field, although quite simple, is popular in connection with electron transport through a single molecule with vibrational mode. We have shown that the external irradiation provides another important experimental tool where both the equilibrium and out of equilibrium transport phenomenon can be probed. In particular, resonant behaviour as the ac frequency matches the frequency of local phonon mode is shown to exist as a result of the satellite-phonon-peak structure in the dot electron density of states. The nonlinear $I$–$V$ curves exhibit new structure caused by the external irradiation, which can be investigated by the recent experimental techniques. We have, as mentioned in the introduction, neglected the on-site electron–electron Coulomb interaction which is important to the many-body Kondo effect at low temperatures. It is, however, an interesting question to ask how the Kondo effect is influenced by the electron–phonon coupling, and how the interplay between the electron–electron interaction and electron–phonon coupling affects the time-dependent electron transport properties in a single-molecular quantum dot. The Kondo effect has also been observed in $C_{60}$ single molecules coupled to metallic [2] and ferromagnetic [20] electrodes. Several theoretical works have focused on the electron transport through molecular quantum dots coupled to a single vibrational mode in the Kondo regime in the absence of irradiation field based on the nonperturbative diagrammatic real-time technique [21], the numerical renormalization group method [11], a generalized Schrieffer–Wolff transformation [22], and the equation-of-motion approach with the improved truncation scheme [23]. It was found that the Kondo resonant peaks will break up into a series of vibron sidebands. It could be predicted that in the presence of microwave irradiation field, the resonant behaviour will show in the time-average transmission and the nonlinear differential conductance when the irradiation frequency matches the Kondo sidebands. The derived time-dependent current can be applied to a more complicated system which includes the on-site electron–electron Coulomb interaction. Work along this line is still in progress.

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