Transient currents of a single molecular junction with a vibrational mode

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Received 8 October 2015, revised 24 December 2015
Accepted for publication 4 January 2016
Published 21 January 2016

Abstract

By using a propagation scheme for current matrices and an auxiliary mode expansion method, we investigate the transient dynamics of a single molecular junction coupled with a vibrational mode. Our approach is based on the spinless Anderson–Holstein model and the dressed tunnelling approximation for the electronic self-energy in the polaronic regime. The time-dependent currents after a sudden switching on the tunnelling to leads, an abrupt upward step bias pulse and a step potential on the quantum dot are calculated. We show that the strong electron–phonon interaction greatly influences the nonlinear response properties of the system, and gives rise to interesting characteristics on the time traces of transient currents.

Keywords: single molecular junction, transient current, electron–phonon interaction

(Some figures may appear in colour only in the online journal)

1. Introduction

The charge transports through nanoscale physical systems, including semiconductor quantum dots [1], single molecular junctions (SMJ) [2], and carbon nanotubes [3], etc, are currently under active investigations in the field of nanoelectronics. During the electron tunnelling processes, the charging of a molecule or a nanotube quantum dot may cause deformations of its surrounding geometry and the concurrent emission or absorption of phonon(vibron) quanta, thus inelastic electron tunnelling effects due to the electron–phonon interaction (EPI) are ubiquitously observed in experiments [4–6]. It was shown that the EPI can give rise to a variety of physically interesting phenomena on the dc characteristics of SMJs or the suspended carbon nanotube quantum dots, e.g. strong current suppression at low bias voltage termed Franck–Condon blockade [4], side peaks of differential conductance [5], and negative differential conductance in the current–voltage characteristics [6].

The dynamical properties of nanoscale systems under time-dependent external potentials are also of great interest, and are important for their potential applications in future electronic devices. Some time-dependent potential related phenomena have been investigated for the systems in the linear response and nonlinear transport regimes, such as the ac response and current noise spectra [7–10], the quantum charge pumping [11–13] and the photon-assisted tunnelling [1, 14], etc. The theoretical investigation of time-dependent transport or transient properties of nanoscale electronic systems is still a challenging task, in particular for the systems with strong EPI or strong Coulomb interaction. For a noninteracting quantum dot, a general theoretical framework for time-dependent transport properties was given in the seminar work by Jauho et al [15] and the switching effects of currents in response to sharp step- and also square-shaped voltage pulses were analytically studied by Maciejko et al [16]. The transient currents through a quantum dot with weak Coulomb interaction were studied by Schmidt et al [17] within the Anderson impurity model by using a mean field theory approximation and also the nonequilibrium Monte Carlo simulation method. The nonequilibrium and transient properties of the Anderson impurity model in the Kondo regime were addressed by using various methods, including the time-dependent noncrossing approximation [18–20], the time-dependent NRG [21, 22]
or the time-dependent DMRG [23] and the functional RG [24]. Recently, we have also studied the time-dependent electron transport through a quantum dot with strong on-site Coulomb interaction [25] using a second-order quantum rate equation and a propagation scheme proposed by Croy and Saaßmann [26].

For the system with EPI, e.g. a SMJ or a carbon nanotube quantum dot coupled with vibrational mode, an archetypetype theoretical model is the Anderson–Holstein model. The transient effects of the Anderson–Holstein model in the weak EPI limit were investigated using perturbation theory [27]. For a fast phonon mode ($\omega_0 \gg \Gamma$) and in the case of sudden switching on the coupling to the leads, it was shown that the time-dependent total current exhibits plateau structures, which can be attributed to the effect of electron shuttling due to the EPI. The dynamics of the Anderson–Holstein model in the strong EPI region is more complicated, and a variety of theoretical methods were used to address this problem, e.g. nonequilibrium Green’s function techniques [28, 29], the polaron tunnelling approximation [30], the scattering states NRG method [31], functional RG approach [32], the diagrammatic Monte Carlo (diagMC) simulation [33–37], the multilayer multiconfiguration time-dependent Hartree method [37–42], the iterative summation of path integrals [43] or the stochastic Langevin equation [44, 45], etc. Bistability of steady state currents and the dot occupation number in the strong EPI case were shown for different initial preparations of the electronic or the phonon states of the system [27, 37, 40–42]. However, a general consensus about the dynamics in the strong coupling region and the condition for bistability in the Anderson–Holstein model haven’t been obtained [46].

In this paper we will study the transient dynamics of a simplified version of the Anderson–Holstein model, in which the spin degree of freedom for electron is neglected. We study this spinless Anderson–Holstein model in the strong EPI region by using the Lang-Firsov unitary transformation and a recently proposed approximation for the electron self-energy termed the dressed tunnelling approximation (DTA) [47–49]. It was shown [48] that the DTA eliminates some pathologies of the single-particle approximation (SPA) or the polaron tunnelling approximation (PTA), and provides results for the current and the differential conductance in good agreement with that of the more elaborated methods such as the diagrammatic Monte Carlo simulation [33]. We consider the system in the polaronic regime with a fast phonon mode, and show that the time-dependent currents exhibit obvious signatures of EPI effect when the system is under a sudden switching on of the tunnel coupling to leads, an upward step-shaped bias voltage or a step potential on the quantum dot. Therefore, the measuring of transient currents can give us important information on the vibrational modes in this kind of SMJs.

This paper is organized as follows: in section 2 the model Hamiltonian and the self-energy terms in the presence of external time-dependent potentials are given. In section 3 within the propagation scheme and the auxiliary-mode expansion method, we derive equations of motion for the dot occupation number and the auxiliary current matrices. In section 4 numerical results and discussions on the transient currents are presented. In section 5 the summary and conclusion are given.

2. Model Hamiltonian and the self-energy terms

The Anderson–Holstein model can describe the electron transport between a molecular quantum dot (QD) and two metallic leads, where the molecular QD is assumed to have only one energy level (with energy $\epsilon_d$) involved in the electron tunnelling process, and is also coupled to a single vibrational mode (phonon) of the molecular with the frequency $\omega_0$ and the EPI strength $g_{ep}$. In this work, we will neglect the on-site Coulomb interaction in the molecular QD and only consider a spinless electron model. The Hamiltonian of the spinless Anderson–Holstein model is given as

$$H = \sum_{k,\eta} \epsilon_{k\eta}(t)c_{k\eta}^d d^d + \omega_0 d^d a^d a + g_{ep} d^d d(a^d + a)$$

$$+ \sum_{k,\eta} [\gamma_{\eta}(t)c_{k\eta}^d d^d a^d + H.c.]$$

(1)

where $\eta = L, R$ denotes the left and right leads, $\epsilon_{k\eta}(t) = \epsilon_d + \Delta_{\eta}(t)$ and $\epsilon_d = \epsilon_d + \Delta_0(t)$, with $\Delta_{\eta}(t)$ and $\Delta_0(t)$ being the time-dependent potentials applied to the lead $\eta$ and the dot region, respectively. $\gamma_{\eta}(t)$ describes the time-dependent tunnel-coupling matrix element between the QD and lead $\eta$, and takes the form of $\gamma_{\eta}(t) = \gamma_{\eta}(t)$, with $u(\eta)$ being a time-dependent function, e.g. in the case of sudden switching on the tunneling at the time $t = 0$, then $u(\eta) = \theta(t)$, with $\theta(t)$ denoting the Heaviside step function. We can also define the coupling strength between the leads and the QD as $\Gamma_{\eta}(\epsilon) = 2\pi \sum_{k} |\gamma_{\eta}(k)|^2 \delta(\epsilon - \epsilon_d)$. Applying the Lang-Firsov unitary transformation [50]: $H = e^{iH} e^{-H}$, with $S = g_{ep} d^d a^d - a$ and the dimensionless parameter $g = g_{ep}/\omega_0$. The Hamiltonian can be transformed to

$$\tilde{H} = \sum_{k,\eta} \epsilon_{k\eta}(t)c_{k\eta}^d X + \tilde{\epsilon}_d(t)c^d d^d a + \omega_0 d^d a$$

$$+ \sum_{k,\eta} [\gamma_{\eta}(t)c_{k\eta}^d dX + \gamma^*_{\eta}(t)X^d c_{k\eta}]$$

(2)

where the phonon shift operators $X = e^{i(\sigma a - a^d)}$, $X^d = e^{-i(\sigma a - a^d)}$ are introduced, and $\tilde{\epsilon}_d = \tilde{\epsilon}_d = \epsilon_d + \Delta_0(t)$ with the renormalized energy level $\tilde{\epsilon}_d = \epsilon_d - g_{ep}^2/\omega_0$. In the transformed Hamiltonian, the direct coupling term between the electron and the phonon is eliminated, but the dot-lead tunnelling amplitude is modified by the phonon shift operator $X$, which is responsible for the observation of the Franck-Condon steps in the dc current-voltage characteristics of the SMJs [4].

The electric current from the lead $\eta$ to the molecular dot $I_{\eta}(t) = -e \langle \frac{dN_{\eta}}{dt} \rangle$, is given by

$$I_{\eta}(t) = \frac{e}{\hbar} \sum_k \left[ \gamma^{*}_{\eta}(t)c_{k\eta}^d dX - \gamma_{\eta}(t)X^d c_{k\eta} \right]$$

$$= \frac{e}{\hbar} (\Pi_{\eta}(t) + \Pi^*_{\eta}(t))$$

(3)
where a current matrix $\Pi_q(t) = i \sum_k \gamma_q(t) e_i^{\dagger} DX_k$ is introduced. By using the equation of motion method, one can express the current matrix as an integral on the closed-time contour over the combination of the GFs of QD operator and its corresponding self-energy as follows

$$\Pi_q(t) = \int_{t-\infty}^{t+\infty} dt'[G_q(t',t')\Sigma_q(t',t) + G_q(t,t')\Sigma_q(t',t)]$$

$$= \int_{t-\infty}^{t+\infty} dt'[G_q(t',t')\Sigma_q(t',t) - G_q(t,t')\Sigma_q(t',t)].$$

(4)

Here $G_q(t,t') \equiv -i\langle T_0 d(t)d(t') \rangle$ denotes the GF of dot operator in the transformed Hamiltonian of (2), and it satisfies the Dyson equation on the closed-time contour

$$[i \frac{\partial}{\partial t} - \varepsilon_d(t)]G_q(t,t') = \delta(t,t') + \sum_\eta \int dt\Sigma_\eta(t,t)G_k(t,t').$$

(5)

Within the dressed tunnelling approximation [47–49], the self-energy term $\Sigma_q(t',t)$ is given by

$$\Sigma_q(t',t) = \sum_k \gamma_q(t') g_{qk}(t') \gamma_q(t) K(t',t),$$

(6)

where $g_{qk}(t',t)$ is the GF of lead $\eta$ in the presence of the time-dependent potential $\Delta_q(t)$, and $K(t',t)$ is the propagator of the phonon shift operator, $\sigma_q(t') \equiv (\varepsilon_d X(t)K(t'))$. By making comparison with the noninteracting resonant tunnelling model, one can see that here the self-energy due to tunnelling into the leads is dressed by a polaronic cloud term.

In this work we will consider the system with a fast phonon mode ($\omega_0 \gg \Gamma$), and a strong energy dissipation of the vibration mode to a thermal bath, e.g. a substrate or a back-gate. Then we can assume the phonons are always in equilibrium and have an equilibrium Bose distribution $n_B = 1/(e^{\beta \omega_0} - 1)$ at the temperature $T = 1/\beta$. The phonon shift operator GF $K(t,t')$ can be obtained by its equilibrium correlation function [47], and its greater and lesser parts are given explicitly as

$$K^>(t',t) = \exp[-\beta \gamma(t) \eta_B - 1 - e^{-\beta \omega_0(t-t')}]$$

and $K^<(t',t) = K^>(t',t)$. They can also expressed as sums of Fourier series

$$K^>(t',t) = \sum_{n=-\infty}^{\infty} w_n e^{-i\omega_n(t-t')}$$

$$K^<(t',t) = \sum_{n=-\infty}^{\infty} w_n e^{i\omega_n(t-t')}.$$  

(7)

(8)

The factor $w_n$ is the weighting factor describing the electronic tunnelling involving absorption or emission of $n$ phonons.

$$w_n = e^{-\beta \gamma(2\omega_n + 1)} I_{\omega}(2\sqrt{\gamma n_B (\eta_B + 1))}) e^{\gamma \omega_n/2},$$

where $I_{\omega}(x)$ is the $n$th Bessel function of complex argument. Substitute (8) into the self-energy expression (6), we obtain the greater and lesser parts of the self-energy as

$$\Sigma_q^>(t',t) = \sum_k \gamma_q^>(t') g_{qk}^>(t') \gamma_q(t) K^>(t',t')$$

$$= -i\nu(t) u(t) \sum_{n=-\infty}^{\infty} w_n$$

$$\times \int \frac{de}{2\pi} [1 - f_\eta(e)] \Gamma_\eta(e) e^{-i(\epsilon + \nu e_\eta(t') - \epsilon)} e^{i\int_0^{\tau} dr \Delta_r(t')},$$

(10a)

$$\Sigma_q^<(t',t) = \sum_k \gamma_q^<(t') g_{qk}^<(t') \gamma_q(t) K^<(t',t')$$

$$= i\nu(t) u(t) \sum_{n=-\infty}^{\infty} w_n$$

$$\times \int \frac{de}{2\pi} f_\eta(e) \Gamma_\eta(e) e^{-i(\epsilon + \nu e_\eta(t') - \epsilon)} e^{i\int_0^{\tau} dr \Delta_r(t')},$$

(10b)

Here $f_\eta(e) = 1/[e^{\beta(\epsilon - \mu_\eta)} + 1]$ is the Fermi distribution function, with $\mu_\eta$ being the chemical potential of lead $\eta$.

3. Auxiliary-mode expansion and the propagation scheme

In the above section we show that an energy integral over Fermi function is involved in the calculation of the lesser or greater self-energy. It will be a heavy burden on computation if multiple integrals are encountered in the study of transient dynamics. At a finite temperature, a well-known method for performing the energy integral is to use the residue theorem and represent the integral by the Matsubara summation [51], but this method suffers from slow convergence especially at the low temperature. Some highly accurate and more efficient methods have been developed in the literatures. In this work, we will use the auxiliary-mode expansion introduced by Croy and Saalman [26]. The Fermi function is expanded in a sum over simple poles

$$f_\eta(e) \approx \frac{1}{2} \sum_{\mu=1}^{N_p} \left( \frac{1}{e - \chi_{\eta\mu}^{-}} + \frac{1}{e - \chi_{\eta\mu}^{+}} \right),$$

(11)

where $\chi_{\eta\mu}^{\pm} = \mu_\eta \pm i\chi_\eta/\beta$ (with $\chi_\eta > 0$). Thus all poles $\chi_{\eta\mu}^{\pm}$ are in the upper (lower) complex plane (details for how to calculate these pole parameters can be found in the appendix of [26]). $N_p$ is the number of pole pairs. Therefore, we will replace the Fermi function in (10) by the above auxiliary-mode expansion.

Next, in order to take into account the finite bandwidth effect, we take the energy dependence of the linewidth function $\Gamma_\eta(e)$ to be Lorentzian

$$\Gamma_\eta(e) = \frac{\Gamma_\eta^0 W^2}{e^2 + W^2} = \frac{\Gamma_\eta^+}{e - iW} + \frac{\Gamma_\eta^-}{e + iW},$$

(12)

where $\Gamma_\eta^0$ is the linewidth constant and $W$ is the bandwidth. It indicates that this linewidth function has two simple poles at $\epsilon = \pm iW$ with residues $\Gamma_\eta^\pm = \mp i\Gamma_\eta^0 W/2$. Then equations (11) and (12) are plugged into the definition of the self-energies in
(10),\ and\ the\ energy\ integral\ is\ evaluated\ by\ the\ contour\ integration\ method.\ For\ \( t > t' \),\ we\ close\ the\ contour\ in\ the\ upper\ half\ plane\ and\ pick\ up\ the\ poles\ to\ obtain\}

\[
\Sigma_p^{\pm}(t', t) = u(t')u(t)\sum_n w_n \left\{ \frac{\Gamma_n^{\pm}}{4} [1 - f_n^{\pm}(iW)] e^{-i(n\omega_n + iW)(t' - t)} \right. \\
+ \frac{1}{\beta} \sum_{\rho = 1}^N \Gamma_{\rho}(\chi^{\pm}_{i\rho}) e^{-i(n\omega_n + iW)(t' - t)} \left\} e^{-i \int_{t'}^{t} dr' \Delta_n(r)},
\]

(13a)

\[
\Sigma_d(t', t) = -u(t')u(t)\sum_n w_n \left\{ \frac{\Gamma_n^{\pm}}{4} f_n^{\pm}(iW) e^{-i(n\omega_n + iW)(t' - t)} \right. \\
- \frac{1}{\beta} \sum_{\rho = 1}^N \Gamma_{\rho}(\chi^{\pm}_{i\rho}) e^{-i(n\omega_n + iW)(t' - t)} \left\} e^{-i \int_{t'}^{t} dr' \Delta_n(r)},
\]

(13b)

where \( f_n^{\pm}(iW) \) denotes that the Fermi function at the pole \( iW \) calculated by the expansion given in (11).\ To simplify\ the\ notation,\ one\ can\ introduce\ the\ coefficients\ and\ exponents\ as\ follows:

\[
\Gamma_n^{\pm} = \left\{ \pm w_n \Gamma_n^{\pm} [1 - f_n^{\pm}(\pm iW)], \pm \frac{1}{\beta} w_n \Gamma_n^{\pm}(\chi^{\pm}_{i\rho}) \right\},
\]

(14a)

\[
\Gamma_n^{\pm} = \left\{ \mp w_n \Gamma_n^{\pm} + f_n^{\pm}(\pm iW), \pm \frac{1}{\beta} w_n \Gamma_n^{\pm}(\chi^{\pm}_{i\rho}) \right\},
\]

(14b)

\[
\chi^{\pm}_{i\rho} = \left\{ \pm iW + n\omega_n, \chi^{\pm}_{i\rho} + n\omega_n \right\},
\]

(14c)

Then\ the\ self-energies\ can\ be\ written\ in\ a\ compact\ form:

\[
\Sigma_p^{\pm}(t', t) = u(t)\sum_{n, \lambda} \Sigma_{p,n}^{\pm}(t', t),
\]

(15)

in\ which\ for\ \( t > t' \),

\[
\Sigma_{p,n}^{\pm}(t', t) = u(t')\Gamma_n^{\pm} e^{-i\chi_n^{\pm}(t' - t)} e^{-i \int_{t'}^{t} dr' \Delta_n(r)},
\]

(16)

and\ for\ \( t < t' \),\ we\ have

\[
\Sigma_{p,n}^{\pm}(t', t) = u(t')\Gamma_n^{\pm} e^{-i\chi_n^{\pm}(t' - t)} e^{-i \int_{t}^{t'} dr' \Delta_n(r)}.
\]

(17)

The\ current\ matrix\ \( \Pi_{\rho}(t) \)\ in\ (4)\ can\ be\ written\ in\ terms\ of\ auxiliary-mode\ expansion\ current\ matrices\ \( \Pi_{\rho,n}(t) \)

\[
\Pi_{\rho}(t) = u(t)\sum_{n, \lambda} \Pi_{\rho,n}(t),
\]

(18)

with

\[
\Pi_{\rho,n}(t) = \int_{-\infty}^{t} dt'[G^{\rho}_{\chi}(t', t) \Sigma_{p,n}^{\rho}\chi^{\rho}(t', t - t') - G^{\rho}_{\chi}(t', t) \Sigma_{p,n}^{\rho}(t', t)].
\]

(19)

By\ differentiating\ the\ above\ equation\ with\ respect\ to\ the\ time\ variable\ \( t \),\ and\ following\ the\ same\ procedure\ as\ in\ [26],\ we\ obtain\ the\ equation\ of\ motion\ for\ the\ auxiliary\ current\ matrices\ as

\[
i \frac{\partial}{\partial t} \Pi_{\rho,n}(t) = u(t) \left[ \Gamma_{\rho}^{\pm} + \eta_n(t)(\Gamma_{\rho}^{\pm} - \Gamma_{\rho}^{\pm}) \right] \\
+ \left[ \chi_n + \Delta_{\chi}(t) - (\chi_{\rho,\chi}(t) + \Delta_{\rho}(t)) \right] \Pi_{\rho,n}(t) + u(t) \sum_{\eta, \lambda} \Omega_{\rho,\eta,\lambda}(\epsilon_{\eta,\chi}^{\rho})(t),
\]

(20)

where\ a\ new\ quantity\ \( \Omega_{\rho,\eta,\lambda}(\epsilon_{\eta,\chi}^{\rho})(t) \)\ is\ introduced,\ which\ satisfies\ the\ following\ equation\ of\ motion:

\[
\frac{\partial}{\partial t} \Omega_{\rho,\eta,\lambda}(\epsilon_{\eta,\chi}^{\rho})(t) = u(t)[\Pi_{\rho,\eta,\lambda}(t)(\Gamma_{\rho}^{\rho} - \Gamma_{\rho}^{\rho}) + (\Gamma_{\rho}^{\rho} - \Gamma_{\rho}^{\rho})\Pi_{\rho,\lambda}(t)]
\]

(21)

From\ the\ current\ conservation\ condition\ in\ the\ QD\ region,\ it\ is\ easy\ to\ see\ that\ the\ equation\ of\ motion\ for\ the\ QD\ occupation\ number\ \( n_\eta(t) \)\ is\ given\ by

\[
\frac{\partial}{\partial t} n_\eta(t) = \sum_{\eta, \lambda} [\Pi_{\eta,\lambda}(t) + \Pi_{\rho,\lambda}(t)].
\]

(22)

Thus\ equations\ (20)–(22)\ form\ a\ closed\ set\ of\ first\ order\ differential\ equations\ for\ current\ matrices\ in\ this\ auxiliary-mode\ expansion\ method,\ and\ give\ the\ basis\ of\ a\ propagation\ scheme\ for\ the\ transient\ dynamics\ of\ this\ EPI\ system.

4. Results and discussions

By\ solving\ the\ above\ set\ of\ partial\ differential\ equations\ with\ the\ finite\ difference\ method,\ we\ can\ obtain\ the\ numerical\ results\ of\ the\ transient\ current\ for\ various\ initial\ conditions.\ For\ simplicity,\ we\ consider\ the\ system\ with\ symmetric\ tunnelling\ coupling\ to\ leads,\ with\ \( \Gamma_L^0 = \Gamma_R^0 = 1.0 \),\ and\ \( \Gamma = (\Gamma_L^0 + \Gamma_R^0)/2 \)\ is\ taken\ as\ the\ unit\ of\ energy.\ The\ phonon\ frequency\ is\ taken\ as\ \( \omega_0 = 10.0 \),\ and\ the\ Fermi\ level\ of\ leads\ at\ equilibrium\ \( \mu_L = \mu_R = 0 \)\ gives\ the\ reference\ point\ of\ energy.\ In\ our\ calculation,\ the\ system\ is\ assumed\ at\ a\ finite\ temperature\ \( T = 0.5 \).\ The\ number\ of\ poles\ \( N_\rho = 80 \),\ which\ ensures\ the\ convergence\ of\ the\ numerical\ results,\ is\ used\ for\ the\ expansion\ of\ Fermi\ distribution\ function.\ We\ study\ the\ properties\ of\ the\ transient\ current,\ which\ can\ be\ divided\ into\ two\ terms:\ the\ total\ current\ \( I(t) = [I_L(t) - I_R(t)]/2 \),\ which\ measures\ the\ charge\ transported\ through\ the\ system,\ and\ the\ displacement\ current\ \( I_{\text{disp}}(t) = I_L(t) + I_R(t) \),\ which\ reflects\ the\ change\ of\ the\ occupation\ number\ in\ the\ dot\ region\ \( I_{\text{disp}}(t) = n_\eta(t) \) [27].

4.1. Sudden switching on the coupling to leads

We\ first\ consider\ transient\ behavior\ of\ currents\ when\ the\ coupling\ of\ the\ quantum\ dot\ to\ leads\ is\ suddenly\ switched\ on\ at\ an\ initial\ time\ \( t_0 = 0 \).\ In\ this\ case\ the\ time-dependent\ function\ \( u(t) = \theta(t) \),\ and\ we\ assume\ the\ quantum\ dot\ initially\ to\ be\ empty.\ In\ figure\ 1\ the\ currents\ \( I_L \)\ and\ \( I_R \)\ from\ the\ left\ and\
the right leads and also the total current $I(t)$ are plotted for the system with a bias voltage $\Delta \mu = 2.0$. Because the finite bandwidth (we take $W = 50$) of leads has been taken into account in our calculation, the currents $I_L$ and $I_R$ are zero at the initial time $t = 0$, then increase to the maximal values at a very short time-scale of $t \approx 1/W$. It is well known that in the infinitely wide band approximation, an unphysical current jump to a finite value immediately at the initial time is obtained \cite{17}. Therefore, the finite bandwidth approach adopted in this work gives a more accurate description of the transient current behavior. The time evolution of currents in figure 1 shows that the system will reach a steady state in the long time limit, however a much longer time is needed for the system with the strong EPI. One can attribute this phenomenon to the strong renormalization of the tunnelling coupling between the leads and the quantum dot because of the EPI. For the system without EPI ($g = 0$) as plotted in figure 1(a), the time traces of currents show smooth curves, which is in good agreement with the results obtained in \cite{17}. In a moderate strength of EPI case ($g = 1.0$), the time-dependence of currents have some irregularities as an indication of EPI effect. For the strong EPI case ($g = 2.0$) in figure 1(c), more complex structures of current evolutions are clearly observed in the short time region.

In figure 2(a) we examine the total current in the short time region for the systems with strong EPI in more details. Three different dot level positions are considered for the SMJ under a fixed bias voltage ($\Delta \mu = 2.0$). As the dot level $\varepsilon_d = 0$, which is located in the transport window ($\mu_R < \varepsilon_d < \mu_L$), the steady total current in the long time limit has a relatively high value. On the contrary, the steady total current is very small when the dot level $\varepsilon_d$ is far below or above the Fermi levels of the leads. It shows that in the short time region the total current exhibits Fano lineshape in the time domain with peak and dip structures at the time $t \approx 2 \pi n/\omega_0$, where $n$ is an integer. Similar current oscillating behaviors were also found previously by the time-dependent APTA and the diagMC simulation \cite{30}, and were attributed to the electron shuttling due to the EPI. It was also shown \cite{29} that the amplitudes of the oscillating current and current spikes increase significantly if the charging image effects in the metallic leads are taken into account. These
results indicate that experimental measuring of the transient total current through SMJs can provide useful information about the vibration modes of the molecule between the leads. In order to estimate the quality of the approximation used in our calculation, we have made a comparison between our results with that of the diagMC simulation [49] in figure 2(b). It is noted that our result for the total current in the short time region agrees well with the diagMC result for the initially occupied quantum dot. For the initially empty quantum dot, the amplitudes of total currents display somewhat large discrepancies, but the positions of dip or peak structures of the total current still coincide. The discrepancy might be due to

the limitation of the approximation made in our calculation for the self-energy of the dot electron. Since the diagMC simulation also suffers from the so-called sign problem [30], we believe that further investigations are needed to give conclusive results for transient current properties of this spinless Anderson–Holstein model.

The displacement current $I_{\text{disp}}(t)$ which reflects the charge accumulation process in the dot is plotted in figure 3. The displacement currents for three different dot levels all exhibit sharp current peaks initially, because the dot to leads coupling is suddenly switched on. For the dot level well above the Fermi level, negative value of $I_{\text{disp}}$ is found immediately after
the initial current peak, which indicates that transient charge depletion process in the center region can also exist in this sudden switching on case. At the time $t \approx t_n^2/\omega_0$, there are also some peak or dip structures at the displacement current, which manifest the EPI effect.

4.2. Currents driven by an upward step pulse

We consider another situation with the quantum dot initially coupled with left and right leads, and the system is in an equilibrium state with zero bias voltage. Then a finite bias applied symmetrically between leads ($\mu_L = -\Delta \mu/2$) is suddenly turned on at the time $t_0 = 0$, and a transient current is driven through the quantum dot. In figure 4 the time-dependent left, right and total current $I_L(t)$, $I_R(t)$ and $I(t)$ after an upward step pulse are plotted for the systems with different EPI constants. In all of figures 4(a)–(c) a sharp rising of current amplitude after turning on the bias voltage is observed. For the noninteracting or the moderate EPI cases shown in figures 4(a) and (b), all of the left, right and total current have one broad peak initially, and then some smooth current oscillations as the system evolves to a steady state. The magnitude of the left current $I_L(t)$ for electron tunnelling into the quantum dot is not equal to that of the tunnelling out current $-I_R(t)$, which indicates that there are charge accumulating and depletion processes in the centre region. For the system with strong EPI in figure 4(c), the first peak for the current becomes quite sharp, and the current amplitude also decreases drastically compared with the weak EPI case, which can be attributed to the strong renormalization of the tunnelling coupling strength $\Gamma$ by EPI. In the time evolution of the currents, some periodic structures with a time-period of $2\pi/\omega_0$ are clearly observed, as a result of the EPI effect in the system.

Next we study the total current under different step voltage pulses in the strong EPI case. Figures 5(a) and (b) correspond to the systems with different dot levels, respectively. In figure 5(a) when the dot level $\bar{\varepsilon}_d$ is well above the Fermi energy ($\bar{\varepsilon}_F = 0$), significant increasing of the steady currents in the long-time limit is observed with increasing the bias voltage. Whereas, for the dot level located around the Fermi energy shown in figure 5(b), the steady current values are almost independent of the bias voltages. It is easy to understand this
Figure 5. The time-dependent total current $I(t)$ for the quantum dot system under different upward step bias voltages. The dot levels are: (a) $\tilde{\epsilon}_d = 5.0$; (b) $\tilde{\epsilon}_d = 0.0$. The EPI constant is taken as $g = 2.0$.

Figure 6. Left, right and total current $I_L(t)$, $I_R(t)$ and $I(t)$ as a function of time after applying a step potential to the QD. The panels (a)–(c) correspond to different EPI constants. The bias voltage is fixed at $\Delta \mu = 2.0$ and the frequency of vibration mode $\omega_0 = 10.0$. The initial dot level is $\tilde{\epsilon}_d = 5.0$, and the QD is in an almost empty state.
behavior since in the later case the dot level is already located in the transport window even for a small bias voltage. Sharp initial current peaks are observed in both cases of different dot level positions. Broad current peaks following the sharp peaks are exhibited for the system with $\bar{\varepsilon} = \varepsilon_0d$, indicating a relatively larger current flow in this system after switching on the bias voltage. As the bias voltage increases, peak or dip structures of the total current arising from EPI interaction at the time $\pi \omega_0 t_n^2$ become more significant.

4.3. Step potential applied to the quantum dot

Now we study the transient behaviour of a fast switching of the gate potential on the quantum dot. In the initial state, the dot level is assumed to $\bar{\varepsilon}_d = 5.0$, which is well above the Fermi levels of the leads, and the quantum dot is almost in an empty state. Then a suddenly switching on of a step potential $V_0(t) = V_0\delta(t)$, with $V_0 = -5.0$, which pushes the dot level to the transport window, and leads to transient currents.

In figure 6 the left and right currents $I_L(t)$ and $I_R(t)$ and the total current $I(t)$ are plotted for the systems under fixed bias voltage ($\Delta \mu = 2.0$) and with different EPI constants. In the noninteracting case with $g = 0$, the transient currents have relatively large amplitudes, and show smooth curves with respect to the time variable. The currents also reach a steady state in a short time period. With increasing the EPI constant $g$, the phonon effect on the currents becomes visible at moderate electron–phonon coupling ($g \approx 1.0$), and is more evident for the systems with strong EPI strength in the short time regime as shown in figure 6(c). It is also observed that for the system with strong EPI constant a longer time is needed to get the steady state compared with the noninteracting system.

The total current for the system under different bias voltages after the sudden switching of the dot level are shown in figure 7. We consider the system in the strong EPI case with $g = 2$. We find that the increasing of the total current in the short time regime exhibits quasi-step structures with a time period of $2\pi \omega_0$. By increasing the bias voltage $\Delta \mu$, the jump amplitude between current steps also increases. It should be noted that here the EPI effects are observed for the system with the bias voltage $\Delta \mu$ lesser than the phonon energy $\omega_0$, therefore the phonon mode are excited by the abrupt gate potential change instead of the bias voltage. It is well known that in the study of the steady current, inelastic tunnelling effects are observed only when the bias voltage $\Delta \mu$ is larger than the phonon frequency $\omega_0$ [2, 9]. Therefore, the transient currents can provide information on high frequency vibrational modes even if the system is under a low bias voltage.

5. Conclusions

We have investigated transient effects in the electron tunnelling through a SMJ coupled with a vibrational mode by using the nonequilibrium GF method and a propagation scheme for current matrices. We mainly focus on the system with the phonon mode frequency $\omega_0 \gg \Gamma$ and in the strong EPI region. When the tunnelling to the leads is suddenly switched on, or the dot level is suddenly tuned to the transport window, the total current gives evident EPI effects, such as the emergence of plateau structures with a time period of $2\pi / \omega_0$, which reveals the frequency of vibrational mode in the molecular junction. The long time for the system to reach a steady state represents a strong renormalization of the tunnelling coupling between leads and the quantum dot by the EPI. As for the system under a upward step bias pulse, sharp initial current peaks are observed, but the oscillation of the total current is significantly suppressed in the strong EPI case. EPI effects are also noticeable in the time trace of the total current by changing the bias voltage or tuning the energy level of the quantum dot. It should be noted that the theoretical results obtained in the present work is based on the dressed tunnelling approximation of the dot level self-energy, and also

![Figure 7. Time-dependent total current $I(t)$ after a step potential applied to the QD. The EPI constant $g = 2.0$, and the bias voltage $\Delta \mu = 1.0, 2.0, 4.0$. The initial dot level is $\bar{\varepsilon}_d = 5.0$.](image-url)
the assumption of an equilibrium phonon distribution at zero temperature. This kind of approximations might be appropriate for the system with phonon frequency $\omega_0 \gg \Gamma$, and in the low bias voltage regime, where the emitted and absorbed phonons during the electron tunneling processes are not of a large number, hence the nonequilibrium phonon effects can be neglected. Since the spinless Anderson–Holstein model is considered, the Coulomb interaction between electrons is not taken into account. One may expect that the strong Coulomb interaction can give rise to interesting effects on the transient dynamics of SMJs, but it is beyond the present work. We expect that the main features of transient currents shown in this work can be testified experimentally in corresponding SMJs using ultrafast spectroscopy techniques.

Acknowledgments

This work was supported by Projects of the National Basic Research Program of China (973 Program) under Grant No. 2011CB925603, the National Natural Science Foundation of China (Grant Nos.91121021 and 11074166), and Shanghai Natural Science Foundation (Grant No. 12ZR1413300).

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