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Phonon generation and phonon energy current fluctuation in quantum dot molecules

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Abstract
We study the phonon dynamics in a biased molecular junction with the interplay of electron–phonon coupling and Coulomb interaction. These interactions are taken into account within the self-consistent Born approximation and mean-field methods. It is found that the Coulomb interaction can enhance the nonequilibrium phonon generation. A general formula for the zero-frequency power spectral density of the phonon energy current fluctuation is presented in terms of the nonequilibrium phonon Green's functions.

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

1. Introduction

The advance in nano-technology has made it possible to have electrons transported through a single molecule. Compared with semiconductor devices, electrons in molecular conductors experience strong coupling to the vibrational degree of freedom [1–3]. This strong coupling not only modifies the electron behaviors such as the electronic spectrum and the conductance, but also excites nonequilibrium phonons (vibrational quanta of the conductor) in the conductor region accompanied by energy exchange between electrons and phonons. As great achievements have been made in the study of electronic transport behavior of molecular conductors with vibrational effects [4–14], an increasing interest has been attracted to the nonequilibrium phonon dynamics due to the electron–phonon coupling [2, 15, 16]. The study of phonon dynamics at the mesoscopic scale has its driving force from not only science but also technology. For example, when electronic devices become smaller and smaller, the generated heat, which is an unavoidable by-product of electron–phonon coupling in the devices, will lead to some consequences. It will increase the temperature in electronic devices, decrease the reliability and lead to undesired parametric changes. It is thus important to describe the performance of molecular devices by taking the full dynamics of both electrons and phonons on an equal footing.

The coupled transport of electrons and phonons in molecular devices has been studied by different theoretical approaches. However, most of these previous studies neglect the Coulomb interaction, which has been proven to be significant in small conductors [17]. The phonon dynamics will be complicated by the interplay of the vibrational effect and the Coulomb interaction. It is the aim of the present paper to study the nonequilibrium phonon dynamics in molecular devices mediated by Coulomb interaction. To take the full dynamics of electrons and phonons on an equal footing, we apply the well established self-consistent Born approximation (SCBA), which has the advantage of including Coulomb effects at the mean-field level. We use different mean-field techniques in a self-consistent manner to study the Coulomb interaction according to its interaction strength U. It is natural to distinguish the two regimes of molecular transport with Coulomb interaction [18]. One is for molecules with weak Coulomb strength, where the dominant energy scale is the contact coupling. For this weak interaction limit, the self-consistent Hartree–Fock mean-field approximation is appropriate. Another regime is for the cases where the Coulomb strength is the dominant energy scale. This happens when the molecule is very small. For example, the Coulomb charging energy of benzene is about 3 eV, which is much larger than the electrode coupling (<0.2 eV for benzene di-thiol on gold) [18]. For the small molecules with very large

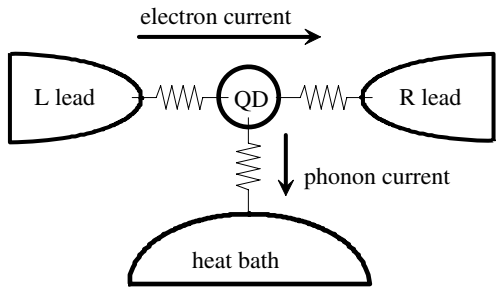

Figure 1. Schematic plot of the model system. The quantum dot is coupled to the electron leads and phonon bath. At nonzero voltage, the driven electron current generates the heat current due to the electron–phonon coupling in the dot.

charging energy, we may approximate the Coulomb strength to be infinity and the electron correlation effect must be taken into account. For this infinite-U strongly correlated regime, we adopt the slave-boson mean-field technique [19–23] to investigate the interplay of the strong Coulomb correlation and the electron–phonon coupling. Our numerical results show that the Coulomb interaction plays an important role in the phonon dynamics. In parallel to the electron current noise [24], previous studies have developed expressions of phonon energy current fluctuation with the Landauer scattering formalism for thermal transport [25–27]. However, in our model, the phonon energy current originates from the inelastic processes of electron–phonon interactions where the Landauer formalism is no longer valid. With the help of the nonequilibrium Green's function technique [28, 29], we present a general formula for the phonon energy current fluctuation with interaction effects.

2. Theoretical formalism

2.1. Model Hamiltonian

We consider a model of one single molecular level with spin degeneracy coupled to two ideal leads in thermal equilibrium. Electrons on this molecule are also coupled to the molecular vibrational degree of freedom. The generated phonon on the molecule can be dissipated by its coupling to the environment, which is represented by a bath of harmonic oscillators in thermal equilibrium. A schematic plot of the device is shown in figure 1. The system Hamiltonian is the sum of terms of electron and phonon parts as well as their mutual interactions. In second quantization, it is given by

$$\begin{aligned} H = &\sum_{\sigma} \epsilon_0 d_\sigma^\dagger d_\sigma + U d_\uparrow^\dagger d_\uparrow d_\downarrow^\dagger d_\downarrow \\ &+ \sum_{k\alpha\sigma} \epsilon_{k\alpha} c_{\sigma k\alpha}^\dagger c_{\sigma k\alpha} + \sum_{\sigma k\alpha} (V_{k\alpha} c_{\sigma k\alpha}^\dagger d_\sigma + \text{h.c.}) \\ &+ \hbar\omega_0 a^\dagger a + \sum_q \hbar\omega_q b_q^\dagger b_q + \sum_q V_q (b_q^\dagger + b_q)(a^\dagger + a) \\ &+ \sum_\sigma M (a^\dagger + a) d_\sigma^\dagger d_\sigma, \end{aligned} \tag{1}$$

where the first two terms on the right-hand side (RHS) of equation (1) describe the electrons in the isolated molecule with Coulomb strength U. The third term represents the electrons in the αth ($\alpha = \text{L}, \text{R}$) lead. $d_\sigma^\dagger$ (d_σ) and $c_{\sigma k\alpha}^\dagger$ ($c_{\sigma k\alpha}$) create (destroy) electrons with spin index σ in the molecule with energy ϵ_0 and the state in the αth lead characterized by the quantum number k and energy $\epsilon_{k\alpha}$, respectively. The fourth term on the RHS of equation (1) corresponds to the coupling between the molecule state and states in ideal leads by the hopping matrix element $V_{k\alpha}$. The fifth and sixth terms on the RHS of equation (1) represent the vibrational modes of the molecule and its environment, i.e. the heat bath. $a^\dagger$ (a) and $b_q^\dagger$ (b_q) are the boson creation (annihilation) operators of the molecular vibrational mode with frequency ω_0 and the qth mode in the heat bath with energy ω_q, respectively. The seventh term is the interaction between these phonon modes with coupling matrix element V_q. The last term corresponds to inelastic interaction with the vibrational mode of the molecule when electrons are tunneling through the device. M is the electron–phonon coupling element.

2.2. Formula for the phonon energy current and its fluctuation

We are interested in the phonon dynamics in the molecular junction, which can be generated by the electron current via electron–phonon coupling. To describe the full dynamics of electrons and phonons, we use the nonequilibrium Green's function technique. The definition of the electron and phonon Green's function can be found in standard textbooks [29]. The lesser ($D^<$) as well as the retarded (D^R) phonon Green's function satisfy, respectively, the Keldysh and Dyson equations as

$$\begin{aligned} D^<(\epsilon) &= D^\text{R}(\epsilon)\Pi^<(\epsilon)D^\text{A}(\epsilon) \\ D^\text{R}(\epsilon) &= \{(D_0^\text{R}(\epsilon))^{-1} - \Pi^\text{R}(\epsilon)\}^{-1}, \end{aligned} \tag{2}$$

where $\Pi^{\text{A(R)}}$ is the advanced (retarded) phonon self-energy, D_0^R is the retarded Green's function for the phonon without interaction, and D^A is the advanced phonon Green's functions, which can be found from the conjugate of the retarded phonon Green's function D^R. The total phonon self-energy Π contains not only the coupling between the vibrating mode and the environment Π_ph, but also terms Π_el arising from the electron–phonon interaction. For the coupling between the molecular phonon mode and those in the environment, the corresponding retarded and lesser self-energies can be given as $\Pi_\text{ph}^\text{R}(\epsilon) = -\frac{\text{i}}{2}\text{sgn}(\epsilon)\gamma$ and $\Pi_\text{ph}^<(\epsilon) = -\text{i}\gamma F(\epsilon)$, where γ is the energy-independent coupling strength and $F(\epsilon) = f_\text{ph}(\epsilon)$ for $\epsilon > 0$ and $F(\epsilon) = 1 + f_\text{ph}(-\epsilon)$ for $\epsilon < 0$. f_ph is the Bose distribution function of the phonon. The electron–phonon interaction is investigated within the SCBA [8–10, 31]; the retarded Π_el^R and lesser $\Pi_\text{el}^<$ phonon self-energies due to electron–phonon coupling are obtained as

$$\begin{aligned} \Pi_\text{el}^\text{R}(\epsilon) = &-\text{i}\sum_\sigma M^2 \\ &\times \int \frac{\text{d}\epsilon'}{2\pi} [G_\sigma^<(\epsilon')G_\sigma^\text{A}(\epsilon' - \epsilon) + G_\sigma^\text{R}(\epsilon')G_\sigma^<(\epsilon' - \epsilon)] \end{aligned} \tag{3}$$

$$\Pi_\text{el}^<(\epsilon) = -\text{i}\sum_\sigma M^2 \int \frac{\text{d}\epsilon'}{2\pi} G_\sigma^<(\epsilon')G_\sigma^>(\epsilon' - \epsilon), \tag{4}$$

where $G^{\text{R(A)}}$ and $G^{<(>)}$ are the retarded (advanced) and lesser (greater) electron Green's functions. Equations for electron

Green's functions can be found following equations (2) by replacing D by G and Π by Σ, respectively, where Σ is the electron self-energy which arises from the dot–lead coupling $\Sigma_{\rm el}$ and the electron–phonon interaction $\Sigma_{\rm ph}$. For the sake of simplicity, the electron retarded and lesser self-energies associated with the dot–lead coupling are taken into account in the symmetric wide band approximation as $\Sigma^{\rm R}_{{\rm L/R},\sigma}(\epsilon) = -\frac{\rm i}{2}\Gamma$ and $\Sigma^{<}_{{\rm L/R},\sigma}(\epsilon) = {\rm i}\Gamma f_{\rm L/R}(\epsilon)$, where Γ is assumed constant and f_α is the Fermi distribution function in the αth lead. In the spirit of SCBA and after omitting the Hartree terms, the retarded ($\Sigma^{\rm R}_{\rm ph}$) and lesser ($\Sigma^{<}_{\rm ph}$) self-energies are given by

$$\Sigma^{\rm R}_{{\rm ph},\sigma}(\epsilon) = {\rm i}\int \frac{{\rm d}\epsilon'}{2\pi} M^2 [D^{\rm R}(\epsilon-\epsilon')G^{<}_{\sigma}(\epsilon') + D^{\rm R}(\epsilon-\epsilon')G^{\rm R}_{\sigma}(\epsilon') + D^{<}(\epsilon-\epsilon')G^{\rm R}_{\sigma}(\epsilon')] \tag{5}$$

$$\Sigma^{<}_{{\rm ph},\sigma}(\epsilon) = {\rm i}\int \frac{\epsilon'}{2\pi} M^2 D^{<}(\epsilon-\epsilon')G^{<}_{\sigma}(\epsilon').$$

In order to find out the generated phonon energy current, our derivation follows the standard approach for the electron particle current. The phonon energy current operator is related as the time change rate of the total energy of the phonon modes in the heat bath: $\hat{J}_{\rm ph} = \frac{\rm d}{{\rm d}t}\sum_q \hbar\omega_q b^{\dagger}_q b_q$. Within the nonequilibrium Green's function technique and along the line of the derivation of the electron current expression by Meir and Wingreen [30], the expectation value of phonon energy current can be derived via the full dynamics of the molecular phonon part as

$$J_{\rm ph} = -\frac{1}{2}\int_{-\infty}^{\infty}\frac{{\rm d}\epsilon}{2\pi\hbar}\epsilon[\Pi^{<}_{\rm el}D^{>} - \Pi^{>}_{\rm el}D^{<}], \tag{6}$$

where $D^{>}$ is the full greater phonon Green's function. Obviously, one can see from equation (6) that, for $\Pi^{<}_{\rm el} = \Pi^{>}_{\rm el} = 0$, $J_{\rm ph} = 0$, i.e. no energy exchange between the electron and phonon subsystems, for vanishing electron–phonon interaction.

In the above analysis, the phonon energy current flows from the localized phonon mode to the heat bath. It originates from the energy loss due to electron–phonon interaction when the electron tunnels through the dot. Therefore, this device can be deemed as a fictitious two terminal device for energy transport, where the heat bath and the electron reservoirs are, respectively, the two terminals which supply or absorb energy. Due to the energy conservation, instead of considering the energy absorbed by the phonon bath as given above, the heat flux can also be measured from another 'terminal', i.e. the energy emitted by the electron reservoirs. Indeed, Frederiksen *et al* [10, 12] have derived an expression for the energy flux from the electron dynamics perspective as

$$P_{\rm ph} = \frac{1}{\hbar}\sum_{\sigma}\int_{-\infty}^{\infty}\frac{{\rm d}\epsilon}{2\pi}\epsilon{\rm Tr}[-\Sigma^{<}_{{\rm ph},\sigma}(\epsilon)G^{>}_{\sigma}(\epsilon)+\Sigma^{>}_{{\rm ph},\sigma}(\epsilon)G^{<}_{\sigma}(\epsilon)], \tag{7}$$

where the energy current is expressed by the Green's functions and self-energies of the electron part. In their derivation of equation (7), they used the *free* phonon Green's function and neglected the dynamics of the phonons. In the SCBA, identical expressions for the energy loss by tunneling electrons through the dot when the full dynamics of phonon are taken into account can be derived. By inserting the full self-energies and Green's functions, equations (7) and (6) can be proved to be identical after some straightforward calculations. This identity thus fulfils the implicit requirement of energy conservation during the energy exchange process. Therefore, the energy conservation law is obeyed by the SCBA for the electron–phonon interaction.

Analogous to the electron noise, the phonon energy current is also fluctuating in time. This fluctuation can be characterized by its power spectral density, i.e. the Fourier transform of the phonon energy current correlation function, as

$$S_{\rm ph}(\omega) = \int_{-\infty}^{\infty}{\rm d}t\,{\rm e}^{{\rm i}\omega t}\langle\{\Delta\hat{J}_{\rm ph}(t),\Delta\hat{J}_{\rm ph}(0)\}\rangle \tag{8}$$

where $\Delta\hat{J}_{\rm ph}(t) = \hat{J}_{\rm ph}(t) - J_{\rm ph}(t)$. With the help of the Wick's theorem and the Langreth rules for analytic continuation, the power spectral density of the phonon energy current fluctuation in the zero-frequency limit can be evaluated as

$$\begin{aligned} S_{\rm ph}(0) = \frac{1}{h}\int {\rm d}\epsilon\,\epsilon^2\{&[D^{\rm R}\Pi^{<}_{\rm ph} + D^{<}\Pi^{\rm A}_{\rm ph}] \\ &\times [D^{\rm R}\Pi^{>}_{\rm ph} + D^{>}\Pi^{\rm A}_{\rm ph}] - D^{<}\Pi^{>}_{\rm ph} \\ &- D^{<}(\Pi^{>}_{\rm ph}D^{\rm A}\Pi^{\rm A}_{\rm ph} + \Pi^{\rm R}_{\rm ph}D^{>}\Pi^{\rm A}_{\rm ph} + \Pi^{\rm R}_{\rm ph}D^{\rm R}\Pi^{>}_{\rm ph}) + {\rm h.c.}\}, \end{aligned} \tag{9}$$

where $D^{>}$ and $\Pi^{>}_{\rm ph}$ are respectively, the greater Green's function of the phonon and the greater phonon self-energy due to coupling to the bath. The derivation of equation (9) is briefly outlined in the appendix. Equation (9) expresses the phonon energy current noise through a molecular dot in terms of the full dynamics of the phonon. It contains not only fluctuations in thermal equilibrium, but also nonequilibrium noise due to the phonon generation by electron–phonon interaction.

Apart from the present case where energy flux flows between the energy reservoirs and the phonon bath, equation (9) is also valid for the study of phonon energy current fluctuations where the phonon flux is driven by the temperature difference of two heat reservoirs. Scattering effects other than the electron–phonon interaction can also be studied by equation (9) with an appropriate form of the Green's functions and self-energies.

In the above derivation, we have only one energy level and vibrational mode in the presence of electron–phonon interaction. However, equations (6) and (9) are able to describe more complicated situations such as the spin-related processes by means of the full Green's functions. A generalization to include cases of the multiple modes and energy levels can be achieved by writing quantities such as the Green's functions in the matrix form analogous to multi-level electron transport problems.

2.3. Approximations to the Coulomb interaction

An exact analysis of the interplay of electron–phonon interaction and Coulomb interaction has long been nontrivial in theoretical studies. One has to rely on some approximations. We note that the SCBA, which has the advantage to take into

account the dynamics of both electrons and phonons on an equal footing, can easily be generalized to include the Coulomb interactions in the mean-field level. In this study, we will focus on two limits where appropriate mean-field approximations can be applied: (i) the weak U limit in the Hartree–Fock mean-field approximation and (ii) the infinite U Kondo regime with the slave-boson mean-field approximations. For situations where the mean-field methods are no longer valid, one has to take into account the correlation between electron–phonon and Coulomb interactions with more efforts. Other advanced techniques such as the equation of motion or the canonical transformation methods are needed. This is beyond the scope of the present paper.

For weak Coulomb interaction, it is safe to neglect the correlation effect and the Coulomb interaction term can be approximated as $Ud_\uparrow^\dagger d_\uparrow d_\downarrow^\dagger d_\downarrow \to U(\langle n_\uparrow\rangle d_\downarrow^\dagger d_\downarrow + \langle n_\downarrow\rangle d_\uparrow^\dagger d_\uparrow)$, where $\langle n_\sigma\rangle$ is the occupation number of electrons in the quantum dot with spin σ, which will be self-consistently determined from

$$\langle n_\sigma\rangle = \frac{1}{2\pi}\int \mathrm{d}\epsilon\ \mathrm{Im}\, G_\sigma^<(\epsilon). \tag{10}$$

The phonon energy current and its fluctuation can be determined after self-consistently solving the coupled Hartree–Fock mean-field approximation for the Coulomb effect and the Born approximation for the vibration effect.

For strong Coulomb interaction, the correlation effect plays an important role and the Hartree–Fock approximation is no longer valid. The low temperature of the quantum dots with strong Coulomb interaction is governed by the Kondo effect. At low bias energy, the transport of the Kondo dot in the infinite U limit can be well described in the slave-boson language [19, 22, 23]. Since the infinite U prevents any double occupation in the dot, the slave-boson technique introduces the transformation $d_\sigma = b^\dagger f_\sigma$, where the boson operator $b^\dagger$ creates one empty state in the dot and the pseudo fermion operator f_σ annihilates one occupied state in the dot. In order to fulfil the constraint of nonexistence of double occupation in the Kondo dot, an additional term with the Lagrange multiplier λ is introduced. Therefore, the Hamiltonian equation (1) in the slave-boson language reads

$$\begin{aligned} H_{\mathrm{SB}} = {} & \sum_\sigma \epsilon_0 f_\sigma^\dagger f_\sigma + \lambda\Big(\sum_\sigma f_\sigma^\dagger f_\sigma + b^\dagger b - 1\Big) \\ & + \sum_{k\alpha\sigma} \epsilon_{k\alpha} c_{\sigma k\alpha}^\dagger c_{\sigma k\alpha} + \sum_{\sigma k\alpha}(V_{k\alpha} c_{\sigma k\alpha}^\dagger f_\sigma + \mathrm{h.c.}) \\ & + \hbar\omega_0 a^\dagger a + \sum_q \hbar\omega_q b_q^\dagger b_q + \sum_q U_q(b_q^\dagger + b_q)(a^\dagger + a) \\ & + \sum_\sigma M(a^\dagger + a) f_\sigma^\dagger f_\sigma. \end{aligned} \tag{11}$$

Following the standard approaches [19, 22, 23] by neglecting the fluctuation of the pseudo-boson operator, one can approximate b by its mean-field value $\langle b\rangle$ as a c-number. This mean-field approximation has been widely used to investigate the Kondo physics in the infinite U limit at zero temperature and low bias voltage. The mean-field value $\langle b\rangle$ and λ can be found from the equation of motion of the boson operator and the constraint as

$$\begin{aligned} \lambda b^2 + \sum_{k\eta\sigma} V_{k\eta\sigma}\langle c_{k\eta\sigma}^\dagger f_\sigma\rangle = 0 \\ \sum_\sigma \langle f_\sigma^\dagger f_\sigma\rangle + b^2 = 1. \end{aligned} \tag{12}$$

After some algebra and inserting the mean-field approximation, the above equations can be enclosed by the nonequilibrium Green function of the quantum dot as

$$\begin{aligned} \lambda\langle b\rangle^2 = \mathrm{i}\sum_\sigma \int \frac{\mathrm{d}\epsilon}{2\pi} G_{\hat f}^<(\epsilon)[\epsilon - (\epsilon_0 + \lambda)] \\ \langle b\rangle^2 - 2\mathrm{i}\int \frac{\mathrm{d}\epsilon}{2\pi} G_{\hat f}^<(\epsilon) = 1, \end{aligned} \tag{13}$$

where we have omitted the spin index of the dot Green's function due to the spin degeneracy. Both the mean-field value $\langle b\rangle$ and λ will be determined by solving the above equations in a self-consistent manner.

3. Numerical results and discussions

In this section, we present our numerical results for the phonon dynamics of the molecular quantum dot. Without losing generality, we will take $\hbar\omega_0 = 1$, $e = 1$ and $\hbar = 1$, where e is the electron charge unit. In the numerical investigations, we use the following set of parameters. The electron level is fixed at $\epsilon_0 = -2$. The coupling constants are $\Gamma = 1$, $\gamma_{\mathrm{ph}} = 0.05$ and $M = 0.075$. The system is at zero temperature. For more realistic systems or comparing with experiments, these parameters may be replaced by either experimental fitting data or first principle calculations. The combination of the nonequilibrium Green's function formalism with *ab initio* techniques to simulate either the electron transport [32] or the electron–phonon coupled transport [12] through molecular devices has achieved much success in recent years. It is therefore feasible to extend the present nonequilibrium Green's function formalism by extracting parameters of molecular devices via first principle calculations.

First we discuss the heat generation as a function of the bias voltage due to the electron–phonon interaction. The heat generated in the dot characterizes the energy transferred from the electrons to the phonon bath. The numerical results are obtained from equation (6) and displayed in figure 2 for different Coulomb interaction strengths. Due to the validity of the slave-boson mean-field method, we restrict ourselves to the low bias voltage regimes. All these calculations show that no heat is generated when bias is zero. Without Coulomb interaction ($U = 0$), as we apply voltage bias to the dot, finite heat is generated. For very low voltage, this heat generation is not obvious (note the log scale of heat generation in our figure), as few electrons can tunnel through the dot. As we increase the voltage bias, more heat will be generated as more electrons can transport through the dot and participate in the electron–phonon coupling. This behavior of heat generation for a vibrating dot without Coulomb interaction has been thoroughly discussed in the literature [14, 16]. In the

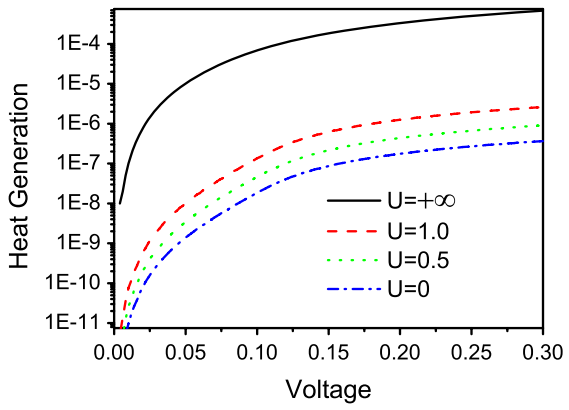

Figure 2. The heat generation in the quantum dot as a function of the applied voltage bias. The curves are obtained by setting different Coulomb interaction strengths U as displayed in the figure. More heat is generated when we increase the Coulomb interaction strength. For finite U, the Coulomb interaction is approximated by the self-consistent Hartree–Fock approximation. For infinite U, the dot operates in the Kondo regime; the transport properties are obtained by the slave-boson mean-field approximation.

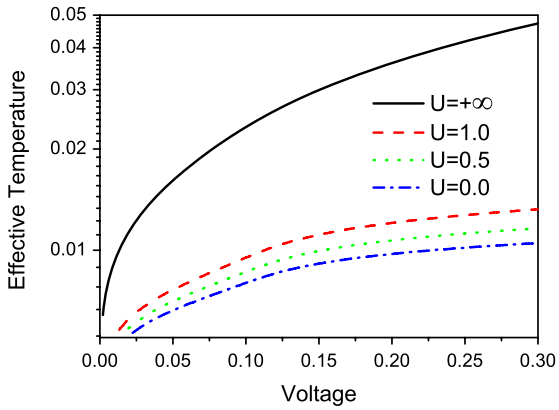

Figure 3. The effective temperature of the dot as a function of the bias voltage for different Coulomb strengths U. The parameters are the same as figure 2.

following, the interplay of Coulomb interaction and electron–phonon coupling is studied for different Coulomb strength parameters with appropriate approximations as discussed above. We have chosen the Coulomb strength to be $U = 0, 0.5, 1.0, \infty$ to make the dot operate from the noninteracting limit to the strongly correlated Kondo regime. The results for finite U are obtained in the self-consistent Hartree–Fock approximation. This approximation is valid at low interaction strength and the correlation effects are not included. With the increase of Coulomb interaction strength, the Hartree–Fock approximation is less reliable and misses important correlation effects such as the multiple transition between the lead and the dot and therefore cannot give a correct description in the Kondo regime. We must therefore go beyond the Hartree–Fock approximation to investigate the phonon dynamics in the large U regime. In this study, we adopt the infinite U slave-boson mean-field theory to study the phonon dynamics in the Kondo regime. We emphasize that, due to the neglecting of correlation effects in Hartree–Fock approximations, the numerical results can only be understood in a qualitative manner, especially for relatively large Coulomb interaction strength U. Our results show that, for fixed voltage bias, more heat is generated if the Coulomb interaction strength becomes greater. This can be qualitatively understood as the following. From the mean-field approximation, we can see that the main contribution from the Coulomb interaction is the renormalization of the energy level of the dot. As we increase the Coulomb strength from zero to infinity, the energy level of the dot will be pushed from the bare energy state of the noninteracting dot, which is far below the Fermi energy, to the Kondo resonance, where a peak of density of states appears at the Fermi energy. This is most significant for the infinite U case, where the dot operates in the Kondo regime. For electron transport at low temperatures, the transport properties are mainly determined by the electrons near the Fermi level. As the electron's dwell time in the dot is proportional to the density of states, the electrons near the Fermi energy can stay a longer time in the dot if the Coulomb strength increases. These electrons can have higher probability to interact with the phonon degree of freedom and thus prefer to transfer energy to the phonons. As a consequence, more heat is generated when the Coulomb interaction pushes the effective energy level of the dot to the resonance.

A direct result of heat generation is the rising of local temperature. Recently, experimental measurement of the current-induced local temperature was reported [33]. It is instructive to introduce an effective temperature to characterize such a heating effect. This effective temperature is related to local population of phonons determined by the Bose distribution function: $n_{\rm ph} = n_{\rm B}(T_{\rm eff})$. To find the phonon population, we start from the balance equation of the phonon number as

$$\frac{{\rm d}n_{\rm ph}}{{\rm d}t} = \frac{j_{\rm ph}}{\hbar\omega_0} - \gamma_{\rm ph}[n_{\rm ph} - n_{\rm B}(\hbar\omega_0, T)] = 0, \qquad (14)$$

where T is the environment temperature at the equilibrium phonon bath. The numerical result for the effective temperature at the dot is displayed in figure 3. The parameters are the same as those of figure 2. One can see that at zero bias the local temperature of the dot increases as we apply the bias voltage. Our results show that the temperature increases more drastically when the Coulomb interaction becomes stronger in accordance with previous observations.

Another question of interest is how much energy is lost due to this electron–phonon coupling. The heat efficiency is defined as the ratio of the generated heat $j_{\rm ph}$ and the total energy supplied by the electric source $P_{\rm e} = I_{\rm e}V$, where $I_{\rm e}$ is the total electric current. Figure 4 presents the numerical results of the heat efficiency for the model device with different Coulomb parameters as discussed above. From the numerical results, we can see that even for the very strong Coulomb interaction and relatively large electron–phonon coupling parameters used here, the efficiency is around 1%.

In figure 5, we show the numerical examples of the fluctuation of the phonon energy current using equation (9),

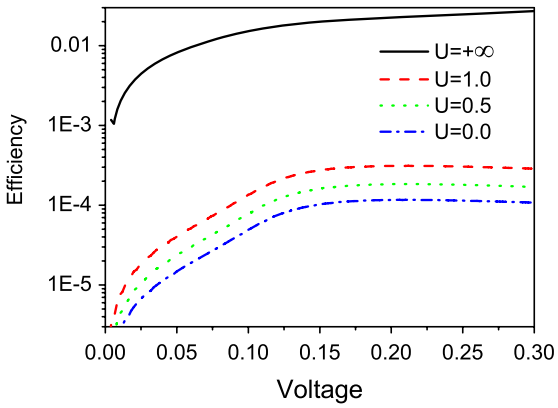

Figure 4. The efficiency of the energy exchange between the electrons and phonons for different Coulomb strengths U. The parameters are the same as figure 2.

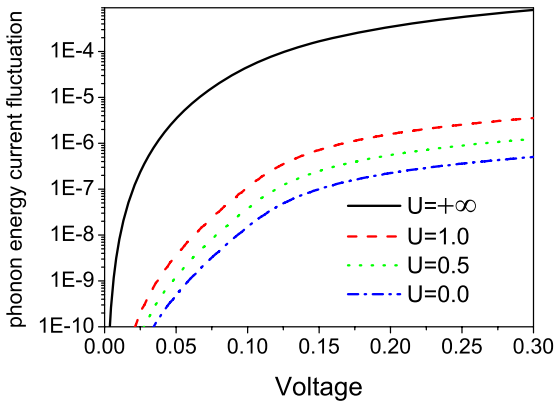

Figure 5. Fluctuation of the phonon energy current as a function of the bias voltage for different Coulomb strengths U.

where the phonon energy current originates from electron–phonon coupling. The parameters of the calculation are identical with the previous results in figures 2–4. We can see that the power spectral density of the phonon energy current fluctuation can be enhanced by the Coulomb interaction. Previous studies on the fluctuation of phonon energy current are based on the generalization of the Landauer scattering formalism and are valid in the elastic thermal transport. We note that our formula is based on the nonequilibrium Green's function technique and is able to deal with the interplay of electron–phonon coupling or Coulomb interaction in the phonon transport as shown in the model calculations presented here. The phonon energy current fluctuation presented here is not only interesting as a phononic analogue of mesoscopic electron phenomenon, but might also be used to provide new insights on fundamental questions, such as the entropy generation [34, 35]. We hope more interesting phenomena of the influence of interaction effects on the phonon current fluctuations can be found in the future. Before the application of the present formula to more general physical systems of phonon dynamics, it is meaningful to discuss some differences between the phonon energy current fluctuation and the electron current fluctuation. The electron current fluctuation has been widely studied to reveal useful information, such as the elementary charge transferred, the statistics of the particles and their correlation effect, in electron transport. However, if one considers the phonon dynamics, some peculiar merits of the phonon must be take into account. (i) The phonon number is not a conserved quantity, in contrast to the total charge in electron transport. The phonons which obey the boson distribution can be generated or destroyed in their transport processes. (ii) The energy quanta of phonons are different for different modes in contrast to the electron charge quanta for electrons or holes. All these peculiar merits make it inappropriate to have a naive analogy of electron transport with the phonon transport. More efforts in the future are needed to characterize the phonon dynamics by its phonon energy current fluctuation.

4. Conclusion

In summary, we have studied the mesoscopic phonon dynamics in molecular electronics mediated by Coulomb interaction. The electron–phonon interaction is taken into account within the self-consistent Born approximation. With the help of mean-field methods, the Coulomb interaction is investigated from the lower interaction strength to the strongly correlated Kondo regime. It is found that the Coulomb interaction contributes energy level shift and enhances the nonequilibrium phonon generation. A general formula has been presented to describe the fluctuations of the phonon energy current generated by electron–phonon coupling in molecular devices with the help of the nonequilibrium Green's function method.

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Appendix

In this appendix, we outline the derivation of the power spectral density of the phonon energy current fluctuation formula in equation (9). The phonon energy current operator can be found from the change of the Hamiltonian (equation (1)) of the bath region as

$$J_{\rm ph} = \frac{{\rm d}\sum_q \epsilon_q b_q^\dagger b_q}{{\rm d}t} = \sum_q \frac{\rm i}{\hbar}\epsilon_q [H, N_q]$$
$$= -\sum_q \epsilon_q V_q \frac{\rm i}{\hbar}(b_q^\dagger - b_q)(a^\dagger + a), \qquad (15)$$

where we used the notation $\epsilon_q = \hbar\omega_q$.

This phonon energy current operator can be rewritten as

$$J_{\rm ph} = -\sum_q \frac{\epsilon_q V_q}{\hbar}\hat{P}_q\hat{Q}_a \tag{16}$$

where the displacement and momentum operators are defined by

$$\hat{Q}_a = a^\dagger + a \qquad \hat{P}_q = {\rm i}(b_q^\dagger - b_q). \tag{17}$$

The phonon energy current fluctuation is defined analogous to the electron noise [24, 36] as

$$S_{\rm ph}(t) = \tfrac{1}{2}\langle\{\Delta J_{\rm ph}(t), \Delta J_{\rm ph}(0)\}\rangle \tag{18}$$

where $\Delta J_{\rm ph}(t) = J_{\rm ph}(t) - \langle J_{\rm ph}(t)\rangle$, and the power spectral density is defined as

$$\begin{aligned} S_{\rm ph}(\omega) &= 2\int_{-\infty}^{\infty} {\rm d}t\, {\rm e}^{{\rm i}\omega t} S_{\rm ph}(t) \\ &= \int_{-\infty}^{\infty} {\rm d}t\, {\rm e}^{{\rm i}\omega t}[\langle\{J_{\rm ph}(t), J_{\rm ph}(0)\}\rangle - 2\langle J_{\rm ph}\rangle^2]. \end{aligned} \tag{19}$$

We are interested in the power spectral density at zero frequency, i.e.

$$S_{\rm ph}(\omega = 0) = \int_{-\infty}^{\infty} {\rm d}t[\langle\{J_{\rm ph}(t), J_{\rm ph}(0)\}\rangle - 2\langle J_{\rm ph}\rangle^2]. \tag{20}$$

Inserting the explicit form of the phonon energy current operator, the phonon energy current correlation is given by

$$\begin{aligned} \langle\{J_{\rm ph}(t), J_{\rm ph}(0)\}\rangle &= \left\langle \sum_{qq'} \frac{\epsilon_q\epsilon_{q'}V_qV_{q'}}{\hbar^2}[Q_a(t)P_q(t)Q_a(0)P_{q'}(0) \right. \\ &\quad \left. + Q_a(0)P_{q'}(0)Q_a(t)P_q(t)]\right\rangle \\ &= M^>(t,0) + M^<(t,0). \end{aligned} \tag{21}$$

Here, we have defined the operator time-ordered operator $M^t(t,0)$ as

$$M^t(t,0) = \left\langle T\left\{\sum_{qq'} \frac{\epsilon_q\epsilon_{q'}V_qV_{q'}}{\hbar^2} Q_a(t)P_q(t)Q_a(0)P_{q'}(0)\right\}\right\rangle, \tag{22}$$

where T is the time-ordering operator. $M^<$ and $M^>$ are the lesser and greater analytic operator of M^t, respectively.

Applying the Wick's theorem, we have

$$\begin{aligned} &\langle T\{Q_a(t)P_q(t)Q_a(0)P_{q'}(0)\}\rangle \\ &= \langle T\{Q_a(t)P_q(t)\}\rangle\langle T\{Q_a(0)P_{q'}(0)\}\rangle \\ &\quad + \langle T\{Q_a(t)P_{q'}(0)\}\rangle\langle T\{Q_a(0)P_q(t)\}\rangle \\ &\quad + \langle T\{Q_a(t)Q_a(0)\}\rangle\langle T\{P_{q'}(0)P_q(t)\}\rangle. \end{aligned} \tag{23}$$

For the sake of simplicity, we can define the following Green's function:

$$D_{AB}(t,t') = -{\rm i}\langle T\{A(t)B(t')\}\rangle, \tag{24}$$

where A and B can be either the displacement or the momentum operator. Equation (23) can then be written as

$$\begin{aligned} \langle T\{Q_a(t)P_q(t)Q_a(0)P_{q'}(0)\}\rangle &= -D_{Q_aP_q}(t,t)D_{Q_aP_{q'}}(0,0) \\ &\quad - D_{Q_aP_{q'}}(t,0)D_{Q_aP_q}(0,t) - D_{Q_aQ_a}(t,0)D_{P_qP_{q'}}(0,t). \end{aligned} \tag{25}$$

If A and B are both displacement operators, D_{AB} is nothing but the phonon Green's functions used in our calculation. Now the task is to evaluate the Green's functions such as $D_{Q_aP_{q'}}$ and $D_{P_qP_{q'}}$. For this purpose, we can make use of the equation of motion method [37]. After some straightforward calculations, we arrive at

$$\begin{aligned} D_{P_qP_{q'}}(t,t') &= D^0_{Q_qQ_{q'}}(t,t')\delta qq' \\ &\quad - \frac{V_qV_{q'}}{\omega_q\omega_{q'}}\int\int {\rm d}t_1\,{\rm d}t_2 D^0_{Q_qQ_q}(t,t_1) \\ &\quad \times\left[\frac{\partial^2}{\partial t_1^2}D_{Q_aQ_a}(t_1,t_2)\right]D^0_{Q_{q'}Q_{q'}}(t_2,t') \end{aligned} \tag{26}$$

and

$$D_{Q_aP_q}(t,t') = \frac{V_q}{\omega_q}\int\left[\frac{\partial}{\partial t_1}D_{Q_aQ_a}(t,t_1)\right]D^0_{Q_qQ_q}(t_1,t'){\rm d}t_1 \tag{27}$$

where

$$D^0_{Q_qQ_q}(t,t') = \left[\frac{\partial^2}{\partial t^2} + \omega_q^2\right]^{-1}(-2\omega_q)\delta(t-t') \tag{28}$$

is the noninteracting phonon Green's function of mode q.

Now, inserting these expressions in the noise expression, making use of the Langreth rules for analytic continuation [29], and after the Fourier transform, we can arrive at the expression for the phonon energy current fluctuation equation (9).

$$\begin{aligned} S_{\rm ph}(0) &= \frac{1}{h}\int {\rm d}\epsilon\,\epsilon^2\{[D^{\rm R}\Pi^<_{\rm ph} + D^<\Pi^{\rm A}_{\rm ph}] \\ &\quad \times[D^{\rm R}\Pi^>_{\rm ph} + D^>\Pi^{\rm A}_{\rm ph}] - D^<\Pi^>_{\rm ph} \\ &\quad - D^<(\Pi^>_{\rm ph}D^{\rm A}\Pi^{\rm A}_{\rm ph} + \Pi^{\rm R}_{\rm ph}D^>\Pi^{\rm A}_{\rm ph} + \Pi^{\rm R}_{\rm ph}D^{\rm R}\Pi^>_{\rm ph}) + {\rm h.c.}\}, \end{aligned} \tag{29}$$

where we have introduced the self-energy due to the coupling of the phonon mode in the molecule with the phonon bath, which is given by $\Pi_{\rm ph}(\omega) = \sum_q V_qD^0_{Q_qQ_q}(\omega)V_q$.

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