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Temperature dependence of polaronic transport through single molecules and quantum dots

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Motivated by recent experiments on electric transport through single molecules and quantum dots, we investigate a model for transport that allows for significant coupling between the electrons and a boson mode isolated on the molecule or dot. We focus our attention on the temperature-dependent properties of the transport. In the Holstein picture for polaronic transport in molecular crystals the temperature dependence of the conductivity exhibits a crossover from coherent (band) to incoherent (hopping) transport. Here, the temperature dependence of the differential conductance on resonance does not show such a crossover, but is mostly determined by the lifetime of the resonant level on the molecule or dot.

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I. INTRODUCTION

In recent years there has been a growing interest in electrical transport through single molecules[1–4] and single electronic levels in quantum dots.[5–7] Some molecular devices exhibit switching behavior with large on-off ratios[1] increasing the motivation to construct molecular electronic devices.[4] In some cases it has been found that the transport is quite temperature dependent[1] and it has been suggested[8] that this is due to the presence of low energy boson modes, such as internal rotations, which couple strongly to the molecular electronic states, and can easily be excited by small temperatures.[9,10] In a similar vein, in double quantum dots it has been found that there are acoustic phonons which couple strongly to the electrons.[6,7]

Some experimental values for the phonon energy have been estimated in various papers. In Table I we give some numbers for reference. We see that the boson (usually phonon) frequency in these systems is quite small, corresponding to temperatures in the range 0.5–50 K. In addition there was a recent proposal[10] to consider transport through a quantum dot to a carbon nanotube cantilever with a resonant frequency of the order of 100 MHz, corresponding to a phonon energy of 0.4 μeV. If the electron-phonon coupling is sufficiently large polaronic transport might be important for these systems. When the electron tunnels through it can absorb or emit bosons, thus altering its energy and the current. If the temperature is much larger than the boson energy, there are many bosons available for absorption and this might heavily influence the current.

In 1959 Holstein[11] predicted that for a periodic one-dimensional molecular crystal with strong electron-phonon coupling there should be a crossover from coherent (band) to incoherent (hopping) transport with increasing temperature. When increasing the temperature the effective bandwidth becomes narrower, this gives rise to a decrease in coherent transport. In contrast, increasing temperature means that more and more phonons are activated and we are in a regime where phonon assisted intersite tunneling starts to contribute to the conductivity. This coherent-incoherent crossover is believed to have been observed for the first time quite recently in single crystals of pentacene.[12] One aim of this paper is to see whether a similar crossover should be seen in polaronic transport through molecules and quantum dots. This might be expected because of the mathematical similarity between the models for periodic systems and the resonant tunneling case. We might expect the tunneling amplitude between the leads and dot to be reduced by polaronic effects, thereby reducing the coherent part of the conductivity. When increasing the temperature the electrons can tunnel with boson assisted transport that enhances the tunneling, possibly leading to a crossover behavior. There have been many theoretical investigations of the effect of phonons on the transport through molecules[8,13–16] and quantum dots,[17–22] but none of them focuses on the temperature dependence of the current. The purpose of this paper is to clarify this aspect of the transport. Li *et al.*[21] included a Hubbard term, but did not consider multiphonon contributions. In a recent paper Emberly and Kirczenow[16] made a thorough analysis of conductance through a molecular wire. A set of self-consistent equations where set up and solved to give the distribution functions in the leads and molecule, and then transmission probabilities were calculated. However, the temperature dependence is not addressed in that paper.

In this paper we perform the analysis for the simplest possible case, where the electrons interact with a single optical boson localized on the dot or molecule. We anticipate that this is sufficient to illustrate the main physics in the more complicated case of many bosons, such as acoustic phonons. In order to obtain analytical results we have to assume that the coupling to the leads is small and the energy level in the dot or molecule is not too close to the Fermi energy in the leads.[23,24] By assuming that the coupling to the leads is small we can calculate the effects from the bosons

TABLE I. Typical values for parameters taken from experiment. $\hbar\omega_0$ is the boson energy and Γ is the line width due to coupling to the leads (defined below) of the resonant level on the molecule or dot. $I_{\max}$ is the maximal current driven through the system.

System	$\hbar\omega_0$	Γ	$I_{\max}$
2 quantum dots (Ref. 7)	40 μeV	0.2 μeV	3 pA
2 quantum dots (Refs. 6,30)	30 μeV	1 μeV	5 pA
molecule (Refs. 1,8)	3 meV		1 nA
C_{60} molecule (Ref. 2)	5 meV		0.1 nA

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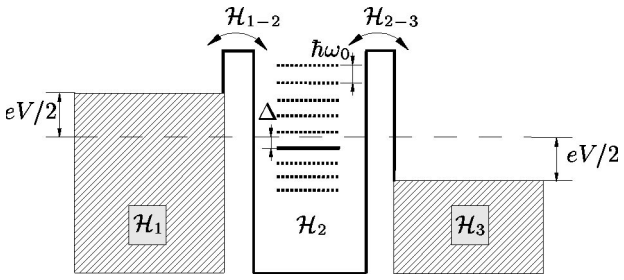

FIG. 1. Tunneling through a system with one level. The dashed lines indicate the bosonic satellites (see text). The Fermi energy in the leads is chosen to be zero. The electrons has to tunnel through the barriers, and can absorb or emit bosons in the process, corresponding to the lines below and above the central resonance, respectively. The Hamiltonian given in Eq. (1) contains terms describing the different parts of the system.

locally on the molecule/dot and then assume that the effect on the leads from the bosons is negligible. This enables us to use well-known results from mesoscopic transport theory. The bosons are possibly most often phonons, but since the theory will look identical (assuming linear couping) for different types of bosons (phonons, magnons, charge oscillations) we will simply refer to ''bosons.'' Even in photon assisted tunneling through quantum dots side bands have been observed when tuning the photon energy.[25] In Sec. II we will define the model we use and in Sec. III, we discuss the approximations we have to make. Different limits for the current are derived in Sec. IV, and in Sec. V we discuss the differential conductivity.

II. CURRENT THROUGH A LEVEL COUPLED TO A LOCAL BOSON MODE

We consider the simplest possible model Hamiltonian and neglect the spin degree of freedom and any effects of electron-electron interactions. The system we study consists of the individual entities (left lead, molecule or quantum dot, and right lead) coupled via tunneling. We assume that we are dealing with a resonant tunneling situation, but the states in the dot (or molecule or any single level system) couples to some boson mode with characteristic frequency ω_0, as shown in Fig. 1. The Hamiltonian is given by

$$\mathcal{H}=\mathcal{H}_1+\mathcal{H}_2+\mathcal{H}_3+\mathcal{H}_{1-2}+\mathcal{H}_{2-3}, \tag{1}$$

where

$$\mathcal{H}_1+\mathcal{H}_3=\sum_{\mathbf{k}_1}\epsilon_{\mathbf{k}_1}c^\dagger_{\mathbf{k}_1}c_{\mathbf{k}_1}+\sum_{\mathbf{k}_3}\epsilon_{\mathbf{k}_3}c^\dagger_{\mathbf{k}_3}c_{\mathbf{k}_3},$$

$$\mathcal{H}_2=\epsilon_0 c_2^\dagger c_2+\hbar\omega_0 a^\dagger a+Mc_2^\dagger c_2(a+a^\dagger),$$

$$\mathcal{H}_{1-2}=\sum_{\mathbf{k}_1}t(c^\dagger_{\mathbf{k}_1}c_2+\text{H.c.}),$$

$$\mathcal{H}_{2-3}=\sum_{\mathbf{k}_3}t(c^\dagger_{\mathbf{k}_3}c_2+\text{H.c.}).$$

Here ϵ_0 is the energy of the level in the dot/molecule and t is the energy associated with hopping onto/off the dot. The electronic dispersion in the leads are given by $\epsilon_{\mathbf{k}_1}$ and $\epsilon_{\mathbf{k}_3}$. M is the coupling to the local boson mode with energy $\hbar\omega_0$. We disregard the spin dependence for simplicity.

First we make a unitary transformation to diagonalize the Hamiltonian $\mathcal{H}_2$. The price we pay for this is that extra operators attach to the tunneling term in the Hamiltonian. The transformation is $\overline{\mathcal{H}_2}=e^S\mathcal{H}_2e^{-S}$, where $S=c^\dagger c(M/\hbar\omega_0)\times(a^\dagger-a)$. This gives us

$$\overline{\mathcal{H}_2}=\hbar\omega_0 a^\dagger a-\Delta c_2^\dagger c_2, \tag{2}$$

where

$$\Delta=\frac{M^2}{\hbar\omega_0}-\epsilon_0. \tag{3}$$

When the central system is a quantum dot ϵ_0 (and thus Δ) can be adjusted by applying a gate voltage. After the transformation the tunneling part of the Hamiltonian becomes

$$\overline{\mathcal{H}}_{1-2}=\sum_{\mathbf{k}_1}t(c^\dagger_{\mathbf{k}_1}c_2X+\text{H.c.}),$$

$$\overline{\mathcal{H}}_{2-3}=\sum_{\mathbf{k}_3}t(c^\dagger_{\mathbf{k}_3}c_2X+\text{H.c.}), \tag{4}$$

where

$$X=\exp\left[\frac{M}{\hbar\omega_0}(a-a^\dagger)\right]. \tag{5}$$

The X factors can be absorbed into a renormalized electron creation/annihilation operator in region 2, so that we are left with the usual resonant tunneling Hamiltonian except that the Greens function for the electrons on the molecule/dot has an additional complication. $\langle T_\tau c(\tau)c^\dagger(0)\rangle\rightarrow\langle T_\tau\overline{c}(\tau)\overline{c}^\dagger(0)\rangle=\langle T_\tau c(\tau)c^\dagger(0)\rangle\langle T_\tau X(\tau)X^\dagger(0)\rangle$. A formula for the current can be derived using a Landauer-Büttiker approach.[26,27] First we calculate the current from the left lead onto the dot from the rate of change of particles in the left lead. A similar expression for the current from the dot to the right lead is derived and the total current through the system is obtained by combining these two formulas. The derivation is presented in detail in Refs. 26 and 27. The result is that the current is given by

$$I(V)=-\frac{2e}{h}\int d\epsilon[f_1(\epsilon)-f_3(\epsilon)]\text{Im}\{\text{tr}[\Gamma G_2(\epsilon)]\}. \tag{6}$$

The applied voltage across the system is V and it enters the two Fermi functions (the equilibrium Fermi level of the leads is chosen to be zero) $f_1(\epsilon)=f(\epsilon-eV/2)$ and $f_3(\epsilon)=f(\epsilon+eV/2)$. Further, $G_2(\epsilon)$ is the Green function for the quantum dot including all effects from the boson system *and* the tunneling to the leads. The parameter Γ is

$$\Gamma\equiv\frac{\Gamma_1\Gamma_3}{\Gamma_1+\Gamma_3}, \tag{7}$$

where $\Gamma_{1(3)}=2\pi t^2 D_{1(3)}(\epsilon)$, $D_{1(3)}$ is the density of states (DOS) in the left (right) lead. $\Gamma_{1(3)}$ is the width of the central resonance due to the tunneling to the left (Γ_1) and right (Γ_3) lead. The total width of the local resonance, Γ_2, is the sum of the two, $\Gamma_2=\Gamma_1+\Gamma_3$

For convenience we introduce the dimensionless parameters

$$g_1 \equiv \left(\frac{M}{\hbar\omega_0}\right)^2,$$

$$g_2 \equiv \left(\frac{\Gamma}{\hbar\omega_0}\right)^2.$$

We emphasize that there are many different energy scales associated with the system: k_BT, eV, $\hbar\omega_0$, Γ, M, and ϵ_0. The relative sizes of these energy scales have a significant effect on the current through the system and what approximations can be made in evaluating it.

The electrons will deposit/absorb energy from the bosonic system that has to be carried away/supplied. Therefore a question arises about how to define the temperature, particularly of the molecule or dot. We assume that the molecule/dot is in equilibrium with a bath and that the tunneling rate is small so that the system relaxes to the initial state after each tunneling event. In a quantum dot the bath can be the substrate that the quantum dot is manufactured on. For a molecule a surrounding cooling liquid[1] can play the role of the bath. Otherwise, we have to assume that the deposited or absorbed energy is transferred to/from the molecule via the leads. As far as we are aware, this assumption is also (implicitly) made in all other theoretical work on this subject.

III. APPROXIMATE EVALUATION OF THE GREENS FUNCTION $G_2(\epsilon)$

To be able to use Eq. (6) we have to calculate the local Green function $G_2(\epsilon)$. Due to the coupling to the leads finding $G_2(\epsilon)$ is a highly nontrivial problem in many-body theory.[22–24] It is comparable in difficulty to the Kondo problem because of the possibility of nonperturbative effects. This is true even in equilibrium (i.e., in the absence of a bias, $V=0$). A recent study was made of a similar Hamiltonian (with spin) using the numerical renormalization group.[24] We are interested in the nonequilibrium case where there is a bias. In order to simplify the analysis we have to rely on approximations, and the result will depend on how the X operators from Eq. (5) are decoupled. One alternative is to assume that the coupling to the leads is small, $\Gamma_1+\Gamma_3 \ll \Delta$, this is the approach taken here. This approximation is justified for small currents, as is the case in the systems considered here. If we were to include the effect on the leads from the bosons on the molecule/dot there would be a narrowing effect on Γ. Hewson and Newns used variational and perturbation methods[23] to show that this narrowing only takes place if the following conditions apply:

$$\frac{M^2}{\hbar\omega_0} > \Gamma, \quad g_1 > 1, \quad \hbar\omega_0 > \Gamma e^{-g_1},$$

$$\hbar\omega_0 > |\Delta|.$$

The conditions on the first line means that the electron-boson coupling has to be large enough to form a polaron. The last requirement on the first line means that individual boson satellites can be distinguished from each other. The second line tells us that if the level and boson satellites are too far from the Fermi level it is energetically unlikely to have virtual boson excitations, thus the leads are unaffected by the bosons. The narrowing is approximately given by

$$t \to t e^{-g_1(1/2+n_B)}, \tag{8}$$

where n_B is the Bose function

$$n_B = \frac{1}{e^{\beta\hbar\omega_0}-1} \tag{9}$$

and $\beta=1/k_BT$.

The above considerations apply to equilibrium ($V=0$) whereas we are interested in the nonequilibrium situation of a finite bias, and particularly the resonant tunneling case where one of the leads' Fermi level is close to the dot/molecule level ($eV=\pm\Delta/2$). In that case the narrowing of the level width due to that lead (but not due to the second lead) may occur, e.g.,

$$\Gamma_2=\Gamma_1+\Gamma_3 \to \Gamma_1+\Gamma_3 e^{-g_1(1+2n_B)}. \tag{10}$$

If $\Gamma_1 \sim \Gamma_3$ this will lead to some quantitative but no significant qualitative changes in the current-voltage characteristics and so we will not consider them further.

We treat the leads as unaffected by the bosons, i.e., no narrowing of the bands in the leads. This means that we ignore the averages of the X operators that appear in the tunneling part of the Hamiltonian, Eq. (4), the justification for this is given above. Below we will also assume that the leads give rise to a flat, energy independent, density of states. This is sometimes called the wide band limit.[19] Otherwise Γ would be energy dependent. The quantum dot Green function calculated using these approximations is

$$G_2(t) = -i\Theta(t) e^{(i\Delta-\Gamma_2/2)t/\hbar} e^{-\Phi(t)}. \tag{11}$$

The factor $e^{-\Phi(t)}$ is due to the coupling to the boson and can be written[28]

$$e^{-\Phi(t)} = e^{-g_1(1+2n_B)} \sum_{l=-\infty}^{\infty} I_l[2g_1\sqrt{n_B(1+n_B)}] e^{il\omega_0(t+i\beta/2)}, \tag{12}$$

where I_l denotes a modified Bessel function.

We Fourier transform the Green function and get an expression for the total current

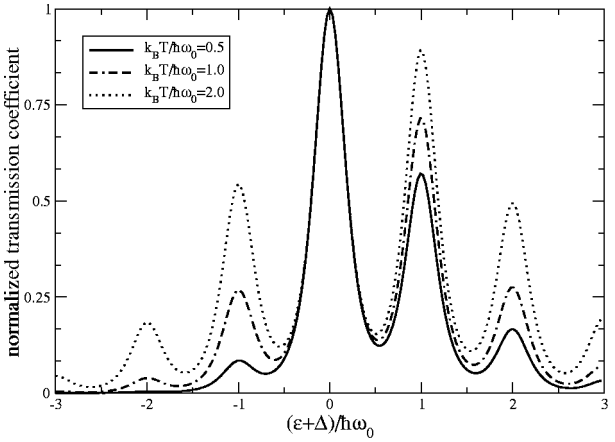

FIG. 2. Transmission coefficient $\mathrm{Im}[\mathrm{tr}(\Gamma G_2)]$, as a function of the energy, for three different temperatures. The satellites are due to the boson modes. $g_1=(M/\hbar\omega_0)^2=0.5$ and $g_2=(\Gamma/\hbar\omega_0)^2=0.09$. The vertical axis is normalized to the highest peak in the plot.

$$I(V)=-\frac{e\Gamma}{h}\int_{-\infty}^{\infty}d\epsilon[f_1(\epsilon)-f_3(\epsilon)]e^{-g_1(1+2n_B)}$$
$$\times\sum_{l=-\infty}^{\infty}I_l[2g_1\sqrt{n_B(1+n_B)}]e^{-l\hbar\omega_0\beta/2}$$
$$\times\frac{\Gamma_1+\Gamma_3}{(\epsilon+\Delta+l\hbar\omega_0)^2+\dfrac{(\Gamma_1+\Gamma_3)^2}{4}}. \qquad (13)$$

We can interpret $\mathrm{Im}[\mathrm{tr}(\Gamma G_2)]$, in Eq. (6), as the transmission coefficient for the tunneling. We plot this in Fig. 2 for a certain choice of parameters. The resonances to the left of $(\epsilon+\Delta)/\hbar\omega_0$ corresponds to absorption of bosons, and the ones to the right to emission of bosons. The middle line can be identified as the so called zero-boson transition. The width of each satellite depends on Γ_2 directly. When increasing the temperature the satellites increase in amplitude, indicating that it is easier to emit/absorb bosons. The asymmetry between negative and positive energies is due to the factor $e^{-l\hbar\omega_0\beta/2}$. This is a due to the fact that at low temperatures there are no available bosons to absorb.

In Fig. 3 we plot the current as a function of voltage using Eq. (13) for a set of parameters. It show steps indicating that more and more satellites participate in conducting electrons. Note that the steps in Fig. 3 occur every second $\hbar\omega_0$. This is simply because the satellites are positioned equidistant on each side of the central resonance, we have to increase the voltage by $2\hbar\omega_0$ in order to cover the satellite. The first satellite starts to contribute to the current when $eV=2\Delta$. A decrease of Γ_2 (g_2 decrease) results in sharper steps, and a decrease in the amplitude of the current. When $\Gamma\gg\hbar\omega_0$ (large g_2) the step structure disappears. Increasing the temperature results in the step structure being washed out to a smooth curve.

When increasing M, the amplitude of the current drops due a decrease of the factor $e^{-(M/\hbar\omega_0)^2(1+2n_B)}$ in Eq. (13). Increasing the temperature has the same effect. Without any coupling to the boson $(M,g_1=0)$ we get a single resonant level without any satellites. This can be seen in Fig. 4 where we plot the current as a function of ϵ_0, the location of the energy level in the dot or molecule. The application of a gate voltage in a quantum dot would be equivalent to changing the level ϵ_0 (or Δ).[6,7] We see a shoulder developing corresponding to the first boson satellite. A similar effect has been seen in a double quantum dot system.[6] The absence of a boson absorption peak in Fig. 4 is due to the low temperature, this comes from the factor $e^{l\hbar\omega_0\beta/2}$. If we increased the temperature, or the electron-boson coupling, enough there would be more side bands visible.

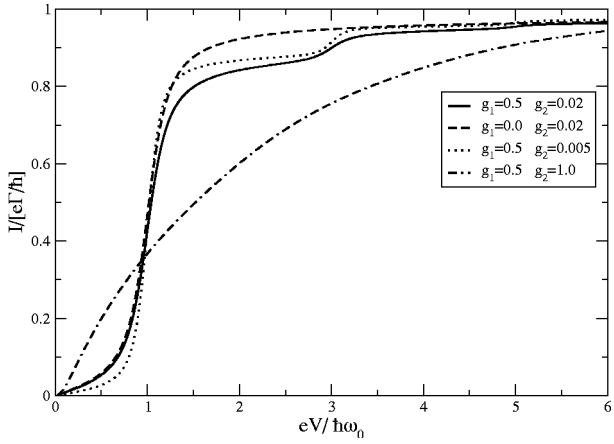

FIG. 3. Current as a function of the applied voltage for different choice of coupling strengths. We set $k_BT=0.1\hbar\omega_0$ and $\Delta=\hbar\omega_0/2$.

IV. LIMITING BEHAVIOR FOR THE CURRENT

In order to better understand the influence from the bosons on the current. Let us now have a look at the current in some limits.

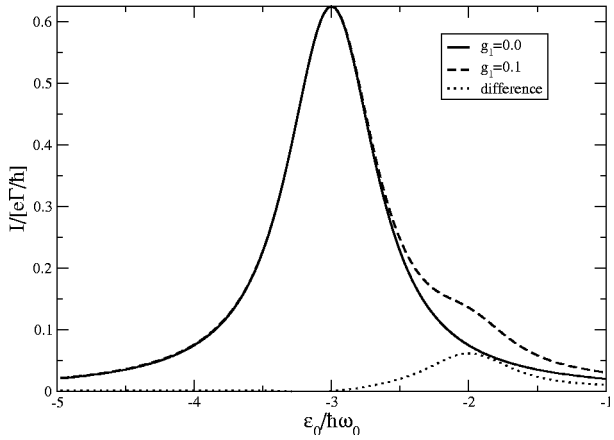

FIG. 4. Current as a function of the location of the energy level in a quantum dot when bosons are present $(g_1=0.1)$ and absent $(g_1=0)$. $k_BT=0.03\hbar\omega_0$ and we put $g_2=0.5$. eV is set to $0.2\hbar\omega_0$ so that we only scan a small region around ϵ_0. Parameters are taken from Ref. 7. We only see the boson emission satellite due to the low temperature and the small electron-boson coupling.

A. $M=0$

If we put the coupling between the boson and the electrons to zero, we get

$$I(V)=-\frac{e}{h}\int_{-\infty}^{\infty}d\epsilon[f_1(\epsilon)-f_3(\epsilon)]\frac{\Gamma_1\Gamma_3}{(\epsilon-\epsilon_0)^2+\frac{(\Gamma_1+\Gamma_3)^2}{4}}. \tag{14}$$

This would correspond to resonant tunneling without any bosons.

1. $k_BT\gg\Gamma,eV$

In this limit Eq. (14) reduces to the linear response expression

$$\frac{I}{V}=\frac{4e^2}{h}\frac{\pi\Gamma}{k_BT\cosh^2\left(\frac{\epsilon_0}{k_BT}\right)}. \tag{15}$$

A similar form was used by Qin *et al.*[7] to fit their experimental data.

2. $T=0$

If the temperature goes to zero we can approximate the Fermi functions with step functions. Then, the integral over ϵ can be performed and the result is

$$\lim_{T\to 0}I(V)=\frac{2e\Gamma}{h}\left[\tan^{-1}\left(\frac{eV-2\epsilon_0}{\Gamma_1+\Gamma_3}\right)+\tan^{-1}\left(\frac{eV+2\epsilon_0}{\Gamma_1+\Gamma_3}\right)\right]. \tag{16}$$

Further, if eV and $2\epsilon_0$ is small compared to $\Gamma_1+\Gamma_3$ we can use the property that $\tan^{-1}(x)\sim x$, and we get

$$\lim_{\substack{T\to 0\\(eV,2\epsilon_0)/(\Gamma_1+\Gamma_3)\ll 1}} I=\frac{4e^2\Gamma}{h(\Gamma_1+\Gamma_3)}V, \tag{17}$$

i.e., a *linear* regime at low voltages. If, on the other hand, we take $V\to\infty$ in Eq. (16) we get

$$\lim_{\substack{T\to 0\\V\to\infty}} I=\frac{e\Gamma}{\hbar}. \tag{18}$$

This means that the whole resonant level contributes maximal to the current.

B. $M\neq 0$

1. $eV\gg k_BT,\hbar\omega_0$

In this case we get the same limit as in Eq. (18) even if $M\neq 0$ from Eq. (13). This can be seen in Fig. 3 where all curves tend to the same value at large V. If we have that $eV\gg\Delta,k_BT$ we can replace $f_1(\epsilon)-f_3(\epsilon)$ by a factor 1, and the integral would extend between $-eV/2$ and $eV/2$. But since eV is greater than all other energies we extend the integral from $-\infty$ to ∞. The integral gives a contribution π. All parts coming from the boson gives 1 and we again have the limit

$$I(eV\gg\Delta,k_BT)\simeq\frac{e\Gamma}{\hbar}. \tag{19}$$

This limit can be seen in Fig. 3 where all curves tend to the same limit at high applied voltage.

2. $k_BT\ll\hbar\omega_0$

Let us now investigate the limit $k_BT\ll\hbar\omega_0$. In this limit (corresponding to low temperatures) we can approximate the Bose function as $n_B\simeq e^{-\hbar\omega_0/k_BT}\ll 1$. All terms corresponding to positive l vanishes. This is a result of the physical fact that positive l corresponds to boson absorption but at $T=0$ there are no bosons. The Bessel function can be approximated as $I_l(z)\sim\frac{1}{l!}(z/2)^l$ when $z\to 0$. Then we get that the current becomes

$$I_{k_BT\ll\hbar\omega_0}=-\frac{2e\Gamma}{h}e^{-g_1}\int_{-\infty}^{\infty}d\epsilon[f_1(\epsilon)-f_3(\epsilon)]\times\sum_{l=-\infty}^{0}\frac{g_1^{|l|}}{|l|!}\frac{\frac{\Gamma_1+\Gamma_3}{2}}{(\epsilon+\Delta+l\hbar\omega_0)^2+\frac{(\Gamma_1+\Gamma_3)^2}{4}}. \tag{20}$$

3. $k_BT\gg\hbar\omega_0$

For high temperatures we approximate the Bose function as $n_B\simeq k_BT/\hbar\omega_0$. The argument in the Bessel function is large and we can use the property

$$I_l(z)\simeq\frac{e^z}{\sqrt{2\pi z}},\quad z\gg 1. \tag{21}$$

Using this, the current becomes

$$I_{k_BT\gg\hbar\omega_0}=-\frac{2e\Gamma}{h}\frac{e^{-g_1\frac{\hbar\omega_0}{4k_BT}}}{\sqrt{4\pi g_1k_BT/\hbar\omega_0}}\int_{-\infty}^{\infty}d\epsilon[f_1(\epsilon)-f_3(\epsilon)]\times\sum_{l=-\infty}^{\infty}\frac{\frac{\Gamma_1+\Gamma_3}{2}}{(\epsilon+\Delta+l\hbar\omega_0)^2+\frac{(\Gamma_1+\Gamma_3)^2}{4}}. \tag{22}$$

C. Saddle point approximation

If $g_2\gg 1$ (i.e., $\Gamma\gg\hbar\omega_0$) we can evaluate the current using a saddle-point approximation similar to that used previously in Refs. 29 and 10. The exponential factor, $e^{-\Phi(t)}\equiv\langle X(t)X^\dagger(0)\rangle$, in Eq. (12) can be written as

$$e^{-g_1[(n_B+1)(1-e^{-i\omega_0t})+n_B(1-e^{i\omega_0t})]}. \tag{23}$$

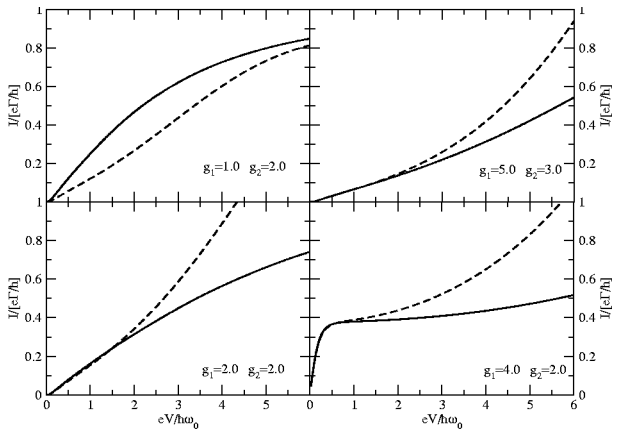

FIG. 5. Failure of the saddle point approximation. Current calculated in two different ways: the full lines were obtained using the exact result [Eq. (13)] and the dashed lines using the saddle point approximation [Eq. (25)] in the expression for the current. We see that the saddle approximation does not reproduce the full expression for the current. Here we set $k_BT=0.1\hbar\omega_0$ and $\Delta=\hbar\omega_0/2$.

We approximate the exponential function in the exponent, $e^z\sim 1+z+z^2/2$, and we get

$$G_2(t)\simeq -ie^{i(\Delta+g_1\hbar\omega_0)t/\hbar-\Gamma_2 t/2\hbar-g_1/2(1+2n_B)(\omega_0 t)^2}. \quad (24)$$

Let us assume that we can neglect the term linear in t in the exponent compared to the quadratic one, i.e., $g_1\gg g_2$. We Fourier transform the resulting Green function and get that the relevant factor entering Eq. (6) becomes

$$\mathrm{Im}[G_2(\epsilon)]\simeq\frac{\exp\left[-\dfrac{(g_1\hbar\omega_0+\Delta+\epsilon)^2}{2g_1(\hbar\omega_0)^2(1+2n_B)}\right]}{\omega_0\sqrt{g_1(1+2n_B)}}. \quad (25)$$

This approximation gives a broad Gaussian line shape covering all the boson satellites. This is in contrast to the individual boson satellites shown in Fig. 2. Using the saddle point approximation would give a Gaussian line shape in $I(\epsilon_0)$, whereas a Lorentzian line shape occurs in the regime, $k_BT\ll\Gamma\ll\hbar\omega_0$, illustrated in Fig. 4.

The current using Eq. (25) is plotted in Fig. 5, and compared to the full expression (13). In this figure we can clearly see that the saddle point approximation cannot reproduce the actual current. Only for a small range of bias voltages, for low temperature and large coupling is there an agreement.

V. DIFFERENTIAL CONDUCTANCE

The differential conductance, defined by

$$C=\frac{dI}{dV}, \quad (26)$$

more clearly reveals the effect of the bosons. In general this is given by

$$C=\frac{e^2\Gamma\beta}{h}\int_{-\infty}^{\infty}d\epsilon\{f_1(\epsilon)[1-f_1(\epsilon)]+f_3(\epsilon)[1-f_3(\epsilon)]\}$$
$$\times e^{-g_1(1+2n_B)}\sum_{l=-\infty}^{\infty}I_l[2g_1\sqrt{n_B(1+n_B)}]$$
$$\times e^{-l\hbar\omega_0\beta/2}\frac{\dfrac{\Gamma_1+\Gamma_3}{2}}{(\epsilon+\Delta+l\hbar\omega_0)^2+\dfrac{(\Gamma_1+\Gamma_3)^2}{4}}. \quad (27)$$

Later we will set $eV=2\Delta$, which corresponds to resonant transport through the zero phonon feature. If we let the temperature go to zero in this expression we can approximate the Fermi functions together with the temperature as a delta function, $\beta n_f(\epsilon)[1-n_f(\epsilon)]\sim\delta(\epsilon)$, and again only negative l contributes, corresponding to emission of bosons, and we get

$$[C(V)]_{T\to 0}=\frac{e^2\Gamma_1\Gamma_3}{2h}e^{-g_1}\sum_{l=-\infty}^{0}\frac{g_1^{|l|}}{|l|!}$$
$$\times\left[\frac{1}{(\Delta+l\hbar\omega_0+eV/2)^2+\dfrac{(\Gamma_1+\Gamma_3)^2}{4}}\right.$$
$$\left.+\frac{1}{(\Delta+l\hbar\omega_0-eV/2)^2+\dfrac{(\Gamma_1+\Gamma_3)^2}{4}}\right]. \quad (28)$$

We define

$$(C_{\mathrm{res}})^0\equiv[C(eV=2\Delta)]_{T\to 0}. \quad (29)$$

For the particular case $\Delta\gg\hbar\omega_0, g_1\hbar\omega_0$, this simplifies to $(C_{\mathrm{res}})^0\simeq(2e^2/h)[\Gamma/(\Gamma_1+\Gamma_3)]e^{-g_1}$, showing how polaronic effects reduce the differential conductance.

In Fig. 6 we plot the differential conductance as a function of the applied voltage for different values of temperature and coupling parameters. The peak at $eV=2\Delta$ correspond to the zero-boson peak, and in the consecutive peaks one, two, three, . . . , and so on, bosons are emitted or absorbed. As seen in this figure increasing the temperature, or the level widths, drastically affects the shape of the differential conductance.

In Fig. 7 we plot the differential conductance on resonance with the zero phonon line as a function of temperature for a range of parameters. In this figure we see that the differential conductance generally decreases with increasing temperature, in contrast to the nonmonotonic dependence found by Holstein[11] for periodic molecular crystals. The corresponding crossover behavior *does not occur* for transport through molecules/quantum dots, since this would be indicated by an upturn in Fig. 7 when increasing the temperature. The absence of a crossover can also be seen by looking at Fig. 3 from that the slope at $eV=2\Delta$ (i.e., the differential conductance) is almost constant when changing g_1 from 0 to

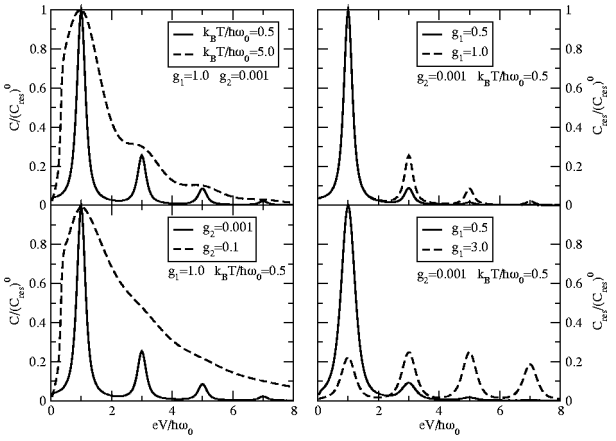

FIG. 6. Differential conductance as a function of applied voltage when changing the temperature (upper left), electron-boson coupling g_1 (upper right and lower right), and the level width g_2 (lower left). $\Delta=\hbar\omega_0/2$. At $eV=2\Delta$ it has a maxima for moderate couplings $g_1\lesssim 1$. To obtain a maximal signal it would be desirable to perform the experiments at this value.

0.5. If we were to calculate the differential conductance in the limit when $k_BT\gg\hbar\omega_0$ from Eq. (22) we see that the temperature dependence of the differential conductance is governed by the pre-factor $e^{-g_1(\hbar\omega_0/4k_BT)}/\sqrt{4\pi g_1 k_B T/\hbar\omega_0}$ and this is a strictly *decreasing* function of the temperature, for reasonably values of g_1. Thus, there will never be an upturn in the differential conductance when increasing the temperature. This general behavior is not changed when g_1 is changed. Even an increased applied voltage, meaning that more boson satellites contributes, was not able to induce a crossover. However, changing Γ does alter the amplitude of the differential conductance, as seen in Fig. 7.

As mentioned above the temperature behavior is dominated by Γ. If we put $M=0$ in Eq. (27) we can write the differential conductance as

$$[C_{\rm res}]_{M\to 0}=\frac{e^2\Gamma}{h}\frac{\tilde{\Gamma}}{k_BT}\int_{-\infty}^{\infty}dy\left[f'(y)+f'\left(y+\frac{2\epsilon_0}{k_BT}\right)\right]\times\frac{1}{y^2+(\tilde{\Gamma}/k_BT)^2},\tag{30}$$

where $\tilde{\Gamma}\equiv(\Gamma_1+\Gamma_3)/2$. If we now take $\epsilon_0=0$ or $k_BT\ll\epsilon_0$ we will have that the differential conductance is a *universal* function of $\tilde{\Gamma}/k_BT$, i.e,

$$[C_{\rm res}]_{M\to 0}=F(\tilde{\Gamma}/k_BT).\tag{31}$$

This can be seen in the lower graph in Fig. 7, where the two graphs for $g_1=0$ (but $g_2=0.02$ and 2.0, respectively) collapse on the same line when the temperature axis is rescaled.

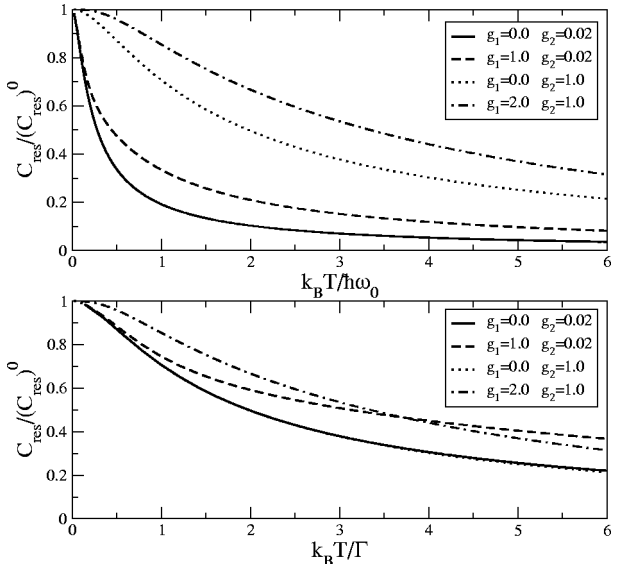

FIG. 7. Differential conductance at the resonance as a function of temperature for different parameters. The upper graph shows that, for moderate couplings $g_1\lesssim 1$, the differential conductance is almost unaltered by the presence of the bosons. The lower graph shows that the differential conductance, for moderate couplings $g_1\lesssim 1$, is determined by the parameter k_BT/Γ. The plots were made assuming a constant DOS in the leads, $\Delta=0$ and $eV=0$.

VI. CONCLUSION

In conclusion we see that the polaronic transport through a single molecule or quantum dot does not clearly exhibit the crossover from coherent to incoherent transport expected for the case of a periodic molecular crystal considered by Holstein.[11] The general behavior of the temperature dependence of the differential conductance is in large *unaffected* by the presence of the bosons. The temperature dependence is mostly determined by the linewidth (due to coupling to the leads) of the resonant energy level. The bosons produce side bands corresponding to absorption and emission of bosons. We also stressed that because of the interaction of the polaron on the dot or molecule with the leads there are potentially some very interesting problems in many-body physics[23,24] to be explored in the model system we have considered.

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