Temperature effect in the conductance of hydrogen molecule

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We present a many-body calculation for the conductance of a conducting bridge of a simple hydrogen molecule between Pt electrodes. The experimental results showed that the conductance $G = dI/dV$ has the maximum value near the quantum unit $G_0 = 2e^2/h$. The $I-V$ dependence presents a peak and dip and we consider that the electron-phonon interaction is responsible for this behavior. At $T = 0$ there is a step in this dependence for the energy of phonons $\omega$ which satisfies $eV = \omega_0$. We calculated the conductance at finite temperature and showed that $dG(T)/dV \propto 1/4T \cosh^2 (\omega_0 \omega/T)$.

I. INTRODUCTION

The recent advances in manipulation of single molecules permit to measure the transport properties of a setup formed from an individual molecule between two electrodes. In contrast to the quantum dots, based on the semiconducting islands, the molecular devices have a more complicated electronic structure.

In a remarkable experiment [1] a setup consisting from a single hydrogen molecule between Pt electrodes was measured and the conductance behavior demonstrated the influence of the vibrational degree of freedom in the transport. The effect was also observed in various organic molecules [2], carbon nanotubes [3] and fullerenes [4] but a theoretical description of these systems is more complicated because of the energy spectrum of the electrons from these molecules. The simplest model which is realistic for the hydrogen molecule setup is to consider the coupling between the vibrational mode (considered as a phonon) of frequency $\omega_0$ and electrons from the leads.

The main effect which appears in the I-V characteristic is the occurrence of a step at $V = \omega_0/e$, which corresponds to dip versus peak in $d^2I/dV^2$. Such a behavior has been studied also in [2, 3, 4]. Many models have been proposed by different authors [8, 9, 11] in order to explain the influence of vibrational modes on the transport, but only recently Egger and Gogolin [12] presented an analytical calculation explaining the current-voltage relation. Their calculation, which gives the correction to the current $\delta I$, is a perturbative calculation for the electron-phonon interaction and takes only the $g^2$ contributions similar to the approximation from Ref. [13, 14].

In this paper we present a many-body calculation of the finite temperature conductance for the hydrogen molecule between metallic leads. The procedure is similar to this from Ref. [12] but we calculate the conductance of the setup close to the characteristic frequency $\omega_0$ at finite temperature. The peak (dip) which appears at this frequency has a finite width, due to thermal effect.

The paper is structured as follows. In Sec. II we present the model and calculate the Green functions. In Sect. III we calculate the lowest order correction $\delta I$ to the current which is given by the electron-phonon processes. The concluding remarks are presented in Sec. IV.

II. MODEL AND GREEN FUNCTION

We start with the Hamiltonian which describes the interaction between electrons and the Holstein phonons used in Ref. [7] for the molecular dots and in Ref. [12] for study of the inelastic scattering influence on the tunneling current in the two-dimensional systems. The simple Hamiltonian, which is analytically tractable, has the form:

$$H = H_0 + H_i$$

where

$$H_0 = \epsilon_0 d^\dagger d + \omega_0 b^\dagger b + \sum_{k\alpha} (\epsilon_k - \mu_\alpha) c^\dagger_{k\alpha} c_{k\alpha}$$

$$H_i = \sum_{k,\alpha} (V_\alpha d^\dagger c_{k\alpha} + H.c.) + gQ d^\dagger d$$

and $Q = b + b^\dagger$.

In the Hamiltonian (1) $d$ and $d^\dagger$ are the operators for the single level $\epsilon_0$, the Holstein phonons with energy $\omega_0$ are described by the operators $b$ and $b^\dagger$, the electrons with the energy $\epsilon_{k\alpha}, (\alpha = L, R)$ and chemical potential $\mu_\alpha$, are described by the operators $c_{k\alpha}$ and $c^\dagger_{k\alpha}$. The interaction between the electrons from the leads and the impurity is $V_\alpha$ and $g$ describes the interaction between the localized electronic level $\epsilon_0$ and the phonons. This Hamiltonian has been used by Egger and Gogolin [12] in this problem at $T = 0$. Using the equation of motion method we calculate the Green function $G_0^r = d/d^\dagger > >$ as $G_0^r(\omega) = (\omega - \epsilon_0 + i\Gamma)^{-1}$ where we define $\Gamma = \Gamma_L + \Gamma_R$ and $\Gamma_\alpha = \pi N(0)|V_\alpha|^2$. The Green function describing the system is:

$$G^r(\omega) = G_0^r(\omega) + G^r(\omega)\Sigma^r(\omega)G_0^r(\omega)$$

where the self energy $\Sigma^r(\omega)$ is taken in the lowest order and has the form [14]:

$$\Delta \Sigma^r(\omega) = \epsilon_0 \Theta(\omega_0 - \omega)$$

where $\Theta$ is a step function.

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\[ \Sigma^r(\omega) = \frac{g^2}{2} \int d\omega' d\omega'' \left[ ImG_0(\omega', \epsilon_0) ImD_0(\omega'', \omega_0) \right] A(T, \omega', \omega'') \frac{\omega' + \omega'' - \omega}{\omega'} \]  

(5)

where \(A(T, \omega', \omega'') = 1 - f_F(\omega') + n_B(\omega'')\), \(f_F(\omega)\) is the Fermi function and \(n_B(\omega)\) is the Bose function, and the phononic Green function \(D_0(\omega, \omega_0)\) is:

\[ D_0^R(\omega, \omega_0) = \frac{1}{2\omega_0} \left[ \frac{1}{\omega - \omega_0 + i\delta} + \frac{1}{\omega + \omega_0 + i\delta} \right] \]

(6)

From Eqs.(5-6) we obtain:

\[ Im\Sigma^r(\omega) = -g^2 \sum_{\alpha, s=\pm 1} \frac{\Gamma_\alpha f_F(\omega_0 - s(\mu_\alpha - \omega))}{(\omega + s\omega_0) + \Gamma^2} \]

(7)

where \(\mu = (\mu_R + \mu_L)/2 - \epsilon_0\) and \(\mu_{\alpha=L/R=\pm 1} = \mu \pm eV/2\). This contribution is dominant in the correction in current for small \(\omega_0/\Gamma\) [12], and we will calculate only this contribution at finite temperature.

### III. CURRENT AND CONDUCTANCE

The electrical current through the dot can be calculated from the Green function \(G^r(\omega)\) as:

\[ I(V) = -\frac{4e}{h} \Gamma_L \Gamma_R \int d\omega [f_F(\omega) - f_R(\omega)] ImG^r(\omega) \]

(8)

and for the case of \(g = 0\) we get the current \(I_0\) as:

\[ I_0(V) = \frac{e}{h} \frac{4\Gamma_L \Gamma_R}{\Gamma} \left[ \text{arctan}(\bar{\mu}_L/\Gamma) - \text{arctan}(\bar{\mu}_R/\Gamma) \right] \]

(9)

For \(V \rightarrow 0\) the transparency of the junction, \(\Upsilon = (h/e^2)dI/dV\) is:

\[ \Upsilon = \frac{4\Gamma_L \Gamma_R}{\Gamma} \left[ 1 - \frac{1}{1 + (\bar{\mu}/\Gamma)^2} \right] \leq 1. \]

(10)

Using Eqs.(4-8) we calculate the correction to the current given by the inelastic electron-phonon scattering [12] as:

\[ \delta I_{inel} = \frac{e}{h} \frac{\Gamma_L \Gamma_R}{\Gamma} \int_{\bar{\mu}_R}^{\bar{\mu}_L} d\omega F(\omega, V) \]

(11)

where \(F(\omega, V)\) is given by:

\[ F(\omega, V) = \sum_{\alpha, s} \frac{g^2 \Gamma_\alpha f_B[\omega_0 - s(\mu_\alpha - \omega)]}{(\omega + s\omega_0) + \Gamma^2}. \]

(12)

In order to calculate the conductance \(G = dI/dV\) and its derivative \(dG/dI\) we will use the relation:

\[ \frac{d}{dV} \int_{\bar{\mu}_R}^{\bar{\mu}_L} F(\omega, V) = \frac{e}{2} \left[ F(\bar{\mu}_L, V) + F(\bar{\mu}_L, V) \right] + \int_{\bar{\mu}_R}^{\bar{\mu}_L} d\omega \frac{dF}{dV}. \]

(13)

Using this formula we calculate the derivative \(d\delta I_{inel}/dV\) as:

\[ \frac{d\delta I_{inel}}{dV} = -\frac{e^2 \Gamma_L \Gamma_R}{\hbar} \frac{g^2}{\Gamma} \left[ f_F(\omega_0 - eV)S_1 + \Theta(V - \omega_0)S_2 \right] \]

(14)

where:

\[ S_1 = \sum_{\alpha = \pm 1} \frac{\Gamma_\alpha (\Gamma^2 - \mu_{\alpha}^2)}{(\Gamma^2 + \mu_{\alpha}^2)(\Gamma^2 + (\mu_{\alpha} - \alpha\omega_0)^2)} \]

and

\[ S_2 = \sum_{\alpha = \pm 1} \frac{\Gamma^2 - (\mu_{\alpha} - \alpha\omega_0)^2}{(\mu_{\alpha}^2 + \Gamma^2)(\mu_{\alpha}^2 + \alpha\omega_0)^2}. \]

(15)

Using these results we calculate the contribution of the inelastic scattering in \(\frac{dG}{dI}\) as:

\[ \frac{d^2\delta I_{inel}}{dV^2} = -\frac{e^2 \Gamma_L \Gamma_R}{\hbar} \frac{g^2}{\Gamma} \frac{S_1 + S_2}{4T \cosh^2\left(\frac{eV - \omega_0}{2T}\right)}. \]

(17)

This is the main result of this paper, which shows that at finite temperature the peak (dip) in the conductance derivative \(dG/dV\) has a finite width at \(eV = \omega_0\) at low temperature, but finite temperature. At \(T = 0\) we reobtain the \(\delta(eV - \omega_0)\) behavior predicted in Ref.[12].

The contribution of the elastic scattering present a logarithmic divergence at \(eV = \omega_0\) which create symmetric dip or peak in the differential conductance. The relative importance of the inelastic versus quasi-elastic contributions has been analyzes in [12] where it was reobtain the result from [12] in this limit. For \(\Gamma_L = \Gamma_R\) and \(\bar{\mu} = 0\) we have \(\Upsilon = 1\) for \(\omega_0 > 2\Gamma\) instead of a dip we get a peak. This is a particular case, and for \(\bar{\mu} \neq 0\) and \(\bar{\mu} > \sqrt{\Gamma^2 + \omega_0^2}/4\) we have a peak. We also have at \(\omega_0 > 2\Gamma\) a peak. We have also a peak at \(\omega_0 = 2\Gamma\). We have also a peak at \(\omega_0 > 2\Gamma\).
IV. CONCLUDING REMARKS

We analyzed, using the many body method, the transport in the simplest molecular dot consisting from a hydrogen molecule between the Pt leads. The analytical calculations of the effect given by the electron-phonon interaction on current at finite temperature have been performed. The obtained results can be regarded as complementary to the $T = 0$ similar calculations presented in Ref. [12]. However, at zero temperature the concept of phonon is not defined and in the transport we use the concept of inelastic scattering. On the other hand the experimental results showed the existence of behavior in the $dG/dI$ which cannot be described by the simple $\delta(eV - \omega_0)$ behavior, which is specific for $T = 0$. We showed that at finite temperature a dip or a peak in this quantity is described by $dG/dI = C(\bar{\mu}, \Gamma, \omega_0)1/(4T \cosh^2(\frac{eV-\omega_0}{2T}))$. This behavior is given by the inelastic scattering between electrons and phonons, the elastic contribution giving a non relevant contribution to the transport in this system, which can be considered as a molecular quantum dot. Our results at $T = 0$ are identical with the results from Ref. [12], and the signature the constant $C(\bar{\mu}, \Gamma, \omega_0)$ gives the same conditions. Our calculations completed the microscopic model presented in Ref. [12], but we consider that the transport in more complicated molecules is difficult to be treated analytically. However, the model can be a starting point for the study of transport in the more complex molecular systems.

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