Transport properties of a single-molecular transistor at finite temperature

Manasa Kalla[1], Swathi. T.S [1], Narasimha Raju Chebrolu[1] and Ashok Chatterjee[1]*

[1]Schhol of Physics, University of Hyderabad, Hyderabad-500046

Email: 15phph22@uohyd.ac.in

Abstract

Quantum transport in a single molecular transistor device is studied at finite temperature in the presence of electron-electron and electron-phonon interactions and dissipation using the Anderson-Holstein-Caldeira-Leggett model. The dissipation due to substrate is treated exactly by a canonical transformation and the electron-phonon interaction is eliminated by the Lang-Firsov transformation. Finally the effective Hamiltonian is studied using the Keldysh non-equilibrium Green function technique and the tunnelling current through the single molecular transistor is obtained at finite temperature.

Keywords: *Quantum Dot, Keldysh Green function, Tunnelling current, Anderson-Holstein model and Caldeira-Leggett model.*

1. Introduction

Interest in the molecular-electronic devices like single molecular transistor (SMT) has been growing very rapidly because of the advances in the fabrication technology of nano-structures. An SMT is a voltage controlled electronic device that contains a central molecule or a Quantum dot (QD) characterized by discrete energy levels, connected to a source and drain (metallic leads) by tunnelling contacts. The first molecular electronics device was proposed theoretically in 1974 using a single organic molecule[1] connected to a source and a drain. Many research groups have studied the transport properties of an SMT device using different theoretical and numerical methods like kinetic equation method, rate equation approach, slave-boson mean-field method, modified perturbation method, numerical renormalization methodand non-equilibrium Green's function approaches (see Ref. 2 for references). In a recent work Raju and Chatterjee[2] have studied the quantum dissipation effects on the transport properties of an SMT using the Keldysh non-equilibrium Green function technique at zero temperature. They have used the Anderson-Holstein model together with the Caldeira-Leggett (AHCL) term to describe the damping effect due to the substrate and they have shown that the electron-phonon (*el-ph*) interaction reduces the current density, but the damping rate enhances the current density.In the present work we study the quantum transport properties of an SMT at finite temperature using the Keldysh non-equilibrium Green function technique.

2. Model

We consider the AHCL model Hamiltonian to describe the SMT device which consists of a central QD coupled with two metallic leads acting as a source (S) and a drain (D). The QD is assumed to have a single energy level (ε_d), an on-site electron-electron (*el-el*) interaction with strength(U), and also an on-site interaction of the electrons with a single vibrational mode of frequency ω_0 with the coupling strengthλ. We also consider a hybridization term which gives hybridization of the local QD state with the continuum energy levels of two metallic leads (S and D). The system is embedded on an insulating substrate which acts as a heat-bath. Thus the phonons of the substrate interact with the phonon of the QD through a linear coupling which leads to a damping effect to the SMT device.

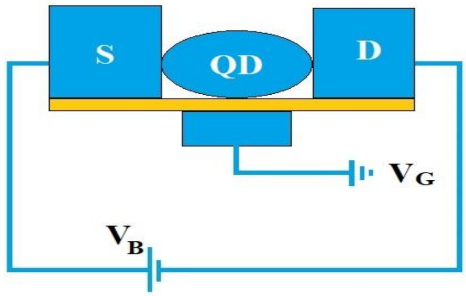


FIG.1 Schematic diagram of an SMT device.

The model Hamiltonian of the system is given by

$$H = \sum_{k\sigma \in S,D} \varepsilon_k n_{k\sigma} + \sum_{\sigma}(\varepsilon_d - eV_g)n_{d\sigma} + Un_{d\uparrow}n_{d\downarrow} + \hbar\omega_0 b^\dagger b + \lambda\hbar\omega_0(b^\dagger + b)\sum_{\sigma} n_{d\sigma}$$
$$+ \sum_{k\sigma\epsilon S,D} (V_k c_{k\sigma}^\dagger c_{d\sigma} + h.c) + \sum_{j=1}^{N}\left(\frac{p_j{}^2}{2m_j} + \frac{1}{2}m_j\omega_j^2 x_j^2\right) + \sum_{j=1}^{N}\beta_j x_j x_0 . \quad (1)$$

where, the notations have their usual meaning.The linear oscillator–bath interaction is eliminated by performing a unitary transformation. As a result, the local phonon frequency gets renormalized to$\widetilde{\omega}_0 = \sqrt{(\omega_0^2 - \Delta\omega^2)}$, where $\Delta\omega^2 = 2\pi\gamma\omega_c$, ω_c being the cut-off frequency and γ the damping rate. The total transformed Hamiltonian reads as,

$$\overline{H} = \sum_{k\sigma\epsilon S,D} \varepsilon_k n_{k\sigma} + \sum_{\sigma}(\varepsilon_d - eV_g)n_{d\sigma} + Un_{d\uparrow}n_{d\downarrow} + \hbar\widetilde{\omega}_0 b^\dagger b + \lambda\hbar\widetilde{\omega}_0(b^\dagger + b)\sum_{\sigma} n_{d\sigma}. \quad (2)$$

Next the Lang-Firsov transformation[3] is applied to Eq. (2) with the generator: $S = \lambda(b^\dagger - b)\sum_\sigma n_{d\sigma}$ to decouple the *el-ph* interaction term. The effective Hamiltonian becomes

$$H_{eff} = \sum_{k\sigma \in S,D} \varepsilon_{k\sigma} n_{k\sigma} + \sum_{\sigma} \tilde{\varepsilon}_{d\sigma} n_{d\sigma} + \widetilde{U} n_{d\uparrow}n_{d\downarrow} + \sum_{k\sigma \in S,D} \widetilde{V}_k\left(c_{k\sigma}^\dagger c_{d\sigma} + h.c\right), \quad (3)$$

with,

$$\tilde{\varepsilon}_{d\sigma} = \varepsilon_d - eV_G - \lambda^2\hbar\omega_0,$$

$$\widetilde{U} = U - \lambda^2\hbar\omega_0 ,$$

$$\widetilde{V}_k = V_k e^{\lambda(b^\dagger - b)}.$$

Using the Keldysh Green function technique, the tunneling current passing through the interacting region

coupled to two metallic leads can be written as

$$J = \frac{e}{2h}\int \{\mathrm{A}_{d\sigma}(\omega)(f_S\Gamma^S - f_D\Gamma^D) + (\Gamma^S - \Gamma^D)G^{<}(\omega)\}d\omega, \tag{4}$$

where, $f_{S(D)}$ is the Fermi distribution function of the source (drain), $\Gamma^{S(D)}$ is the coupling strength between the QD and source (drain). Here we have considered symmetric coupling i.e.,$\Gamma^S = \Gamma^D = \Gamma = (\Gamma^S + \Gamma^D)/2,$ so that we get

$$\Gamma^{S(D)} = 2\pi\rho_{S(D)}(\omega)|\mathrm{V}_k|^2 e^{-\lambda^2\left(f_p + \frac{1}{2}\right)}, \tag{5}$$

where, $\rho_{S(D)}(\omega)$ represents the density of states of the source (drain). The chemical potentials of the source and the drain are related to the bias and mid voltages as $eV_B = (\mu_S - \mu_D),\ and\ eV_m = (\mu_S + \mu_D)/2$ and the spectral function$\mathrm{A}(\varepsilon)$, which describes the possible excitation energy spectrum, can be written in terms of the Green functions as,

$$A_{d\sigma}(\varepsilon) = \sum_{n=-\infty}^{\infty} iL_n\left(2\lambda^2\sqrt{f_p(1+f_p)}\right)\left[\tilde{G}^{>}(\varepsilon - n\hbar\tilde{\omega}_0) \ - \ \tilde{G}^{<}(\varepsilon + n\hbar\tilde{\omega}_0)\right], \tag{6}$$

where, n is the number of phonons, $f_p = 1/\left(e^{\beta\hbar\tilde{\omega}_0} - 1\right),\ \beta = 1/k_BT$, and

$$L_n\left(2\lambda^2\sqrt{f_p(1+f_p)}\right) = e^{-\lambda^2/2(2f_p+1)}e^{n\beta\hbar\tilde{\omega}_0/2}I_n\left(2\lambda^2\sqrt{f_p(1+f_p)}\right). \tag{7}$$

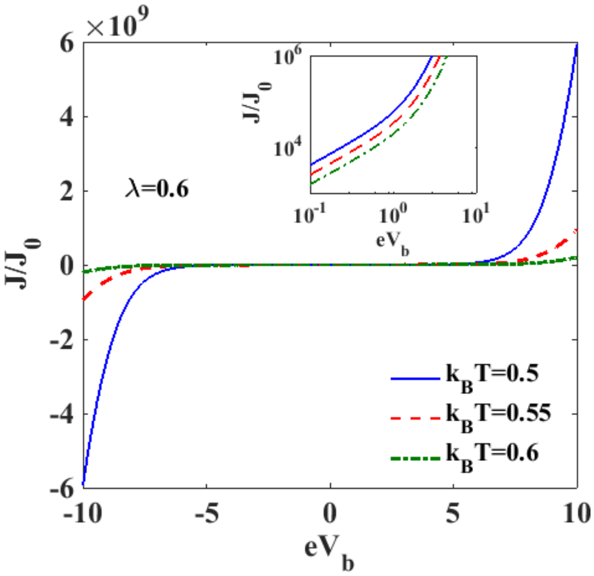
The lesser and greater Green functions can be written as,

$$\tilde{G}^{>(<)}(\varepsilon) = \tilde{G}^{r}_{dd}(\varepsilon)\Sigma^{>(<)}(\varepsilon)\tilde{G}^{a}_{dd}(\varepsilon)\ , \tag{8}$$

where,

$$\Sigma^{<}(\varepsilon) = i\,\Gamma[f_S(\varepsilon) + f_D(\varepsilon)]\,, \quad \Sigma^{>}(\varepsilon) = -i\,\Gamma[2 - (f_S(\varepsilon) + f_D(\varepsilon))]. \tag{9}$$

3. Results and Discussion



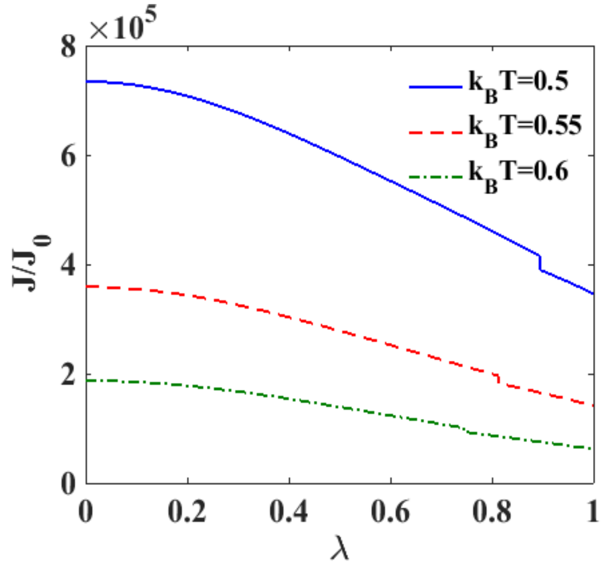


FIG. 2 J/J_0 as a function of eV_b for different values values of k_BT at $\lambda = 0.6$. (Inset: J/J_0 as a function of eV_b in logarithmic scale).

FIG. 3 J/J_0 as a function of λ for different of k_BT at $V_b = 2.5$

In our numerical calculations, we measure all the energies in units of phonon energy $\hbar\omega_0$ and have chosen the values of the other parameters as: $\Gamma = 0.2,\ eV_m = 0.5,\ U = 3, \gamma = 0.01$ and $\varepsilon_d = 0$.Also we work with constant density of states for the conduction electrons in the source and drain. Fig.2 shows the variation of the normalized tunnelling current density J/J_0 , as a function of the bias voltage for different values of temperature at $\lambda = 0.6$. As we can clearly see, J/J_0 increases non-linearly with increasing bias voltage and decreases with increasing k_BT. In Fig.3, we present J/J_0 as a function of *el-ph* interaction strength and we observe that J/J_0 decreases with increasing λ due to strong polaronic effect.

4. Conclusions

We have studied the electronic transport properties of an SMT device in the presence of *el-el* and *el-ph* interactions and phononic dissipation. We have used the Anderson-Holstein-Caldeira-Leggett model Hamiltonian to describe the device and calculated the tunnelling current density using the Keldysh Green function method. It is shown that current density decreases with $\boldsymbol{\lambda}$, due to polaronic effect. With increasing temperature, the number of phonons increases, which enhances the polaronic effect, and also due to the thermal smearing, the current density decreases with increasing temperature.

5. Acknowledgements

One of the authors (M.K.) gratefully acknowledges the financial support from CSIR, India for SRF (09/414(1100)/2015-EMR-I). CNR would like to acknowledge the financial support from UGC NRC, School of Physics, University of Hyderabad.

REFERENCES

[1] AVIRAM. A. AND RATNER. M, *CHEM. PHYS. LETT.*29, 277 (1974).

[2] NARASIMHA RAJU.CH AND ASHOK. C. *SCIENTIFIC REPORTS*. 6, 18511 (2016)

[3] LANG. I. G &FIRSOV. YU.A, *SOV.PHYS. JETP* 16, 1301 (1962).