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Effect of phonons on the ac conductance of molecular junctions

Akiko Ueda^{1*}, Ora Entin-Wohlman^{1,2}, Amnon Aharony^{1,2}

Abstract

We theoretically examine the effect of a single phonon mode on the structure of the frequency dependence of the ac conductance of molecular junctions, in the linear response regime. The conductance is enhanced (suppressed) by the electron-phonon interaction when the chemical potential is below (above) the energy of the electronic state on the molecule.

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Introduction

Molecular junctions, made of a single molecule (or a few molecules) attached to metal electrodes, seem rather well established experimentally. An interesting property that one can investigate in such systems is the interplay between the electrical and the vibrational degrees of freedom as is manifested in the I - V characteristics [1,2].

To a certain extent, this system can be modeled by a quantum dot with a single effective level ε_0 , connected to two leads. When electrons pass through the quantum dot, they are coupled to a single phonon mode of frequency ω_0 . The dc conductance of the system has been investigated theoretically before, leading to some distinct hallmarks of the electron-phonon (e-ph) interaction [3-6]. For example, the Breit-Wigner resonance of the dc linear conductance (as a function of the chemical potential μ , and at very low temperatures) is *narrowed down* by the e-ph interaction due to the renormalization of the tunnel coupling between the dot and the leads (the Frank-Condon blockade) [4,5]. On the other hand, the e-ph interaction *does not* lead to subphonon peaks in the linear response conductance when plotted as a function of the chemical potential. In the nonlinear response regime, in particular for voltages exceeding the frequency ω_0 of the vibrational mode, the opening of the inelastic channels gives rise to a sharp structure in the I - V characteristics. In this article, we consider the ac linear conductance to examine phonon-induced structures on transport properties when the ac field is present.

Model and calculation method

We consider two reservoirs (L and R), connected via a single level quantum dot. The reservoirs have different chemical potentials, $\mu_L = \mu + \text{Re}[\delta\mu_L e^{i\omega t}]$ and $\mu_R = \mu + \text{Re}[\delta\mu_R e^{i\omega t}]$. When electrons pass through the quantum dot, they are coupled to a single phonon mode of frequency ω_0 . In its simplest formulation, the Hamiltonian of the electron-phonon (e-ph) interaction can be written as $\mathcal{H}_{e-ph} = \gamma (b + b^\dagger) c_0^\dagger c_0$, where b (c_0) and b^\dagger (c_0^\dagger) are the annihilation and the creation operators of phonons (electrons in the dot), and γ is the coupling strength of the e-ph interaction. The broadening of the resonant level on the molecule is given by $\Gamma = \Gamma_L + \Gamma_R$, with $\Gamma_{L(R)} = 2\pi v t_{L(R)}^2$, where v is the density of states of the electrons in the leads and $t_{L(R)}$ is the tunneling matrix element coupling the dot to the left (right) lead.

The ac conductance of the system is derived by the Kubo formula. In the linear response regime, the current is given by $I = (I_L - I_R)/2$, where

$$I_L(\omega) = -e \text{Re} [X_{LL}^r(\omega) \delta\mu_L + X_{LR}^r(\omega) \delta\mu_R]. \quad (1)$$

Here, $X_{LL(R)}^r(\omega)$ is the Fourier transform of the two particle Green function,

$$X_{LL(R)}^r(t-t') = -i\theta(t-t') \left\langle \left[\hat{I}_L(t), \hat{N}_{L(R)}(t') \right] \right\rangle, \quad (2)$$

where $\hat{I}_L = -\partial_t \hat{N}_L$, with $\hat{N}_{L(R)} = \sum_{k(p)} c_{k(p)}^\dagger c_{k(p)}$, and $c_{k(p)}$ denoting the creation and annihilation operators

* Correspondence: akiko@bgu.ac.il

¹Department of Physics, Ben Gurion University, Beer Sheva 84105, Israel.
Full list of author information is available at the end of the article

of an electron of momentum $k(p)$ in the left (right) lead. The ac conductance is then given by

$$G = e(I_L - I_R) / (2\delta\mu), \quad \delta\mu = \delta\mu_L - \delta\mu_R. \quad (3)$$

In this article we consider the case of the symmetric tunnel coupling, $\Gamma_L = \Gamma_R$. We also assume $\delta\mu_L = -\delta\mu_R = \delta\mu/2$. The e-ph interaction is treated by the perturbation expansion, to order γ^2 . The resulting conductance includes the self-energies stemming from the Hartree and from the exchange terms of the e-ph interaction, while the vertex corrections of the e-ph interaction vanish when the tunnel coupling is symmetric. We also take into account the RPA type dressing of the phonon, resulting from its coupling with electrons in the leads [3].

Results

The total conductance is given by $G = G_0 + G_{\text{int}}$, where G_0 is the ac conductance without the e-ph interaction, while $G_{\text{int}} \equiv G_{\text{H}} + G_{\text{ex}}$ contains the Hartree contribution G_{H} and the exchange term G_{ex} . Figure 1 shows the conductance G as a function of $\varepsilon_0 - \mu$, for a fixed ac frequency $\omega = 0.5\Gamma$. The solid line indicates G_0 . The dotted line shows the full conductance G , with $\gamma = 0.3\Gamma$. The peak becomes somewhat narrower, and it is shifted to higher energy, which implies a lower (higher) conductance for $\varepsilon_0 < \mu$ ($\varepsilon_0 > \mu$). However, no additional peak structure appears.

Next, Figure 2a shows the full ac conductance G as a function of the ac frequency ω , when $\varepsilon_0 - \mu = \Gamma$. The solid line in Figure 2a indicates G_0 . Two broad peaks appear around ω of order $\pm 1.5(\varepsilon_0 - \mu)$. The broken lines show G in the presence of the e-ph interaction with $\omega_0 = 2\Gamma$, $\omega_0 = \Gamma$, or $\omega_0 = 0.5\Gamma$. The e-ph interaction increases the conductance in the region between the original peaks, shifting these peaks to lower $|\omega|$, and decreases it slightly outside this region. Figure 2b indicates the additional conductance due to the e-ph

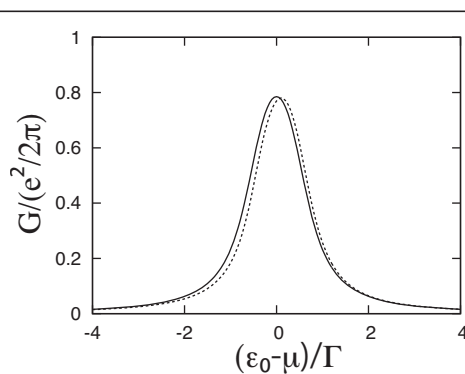


Figure 1 The ac conductance as a function of $(\varepsilon_0 - \mu)$. The ac frequency $\omega = \Gamma$, $\Gamma_L = \Gamma_R$ and $\delta\mu_L = -\delta\mu_R$. Solid line: without e-ph interaction. Dotted line: $\gamma = 0.3\Gamma$ and $\omega_0 = \Gamma$.

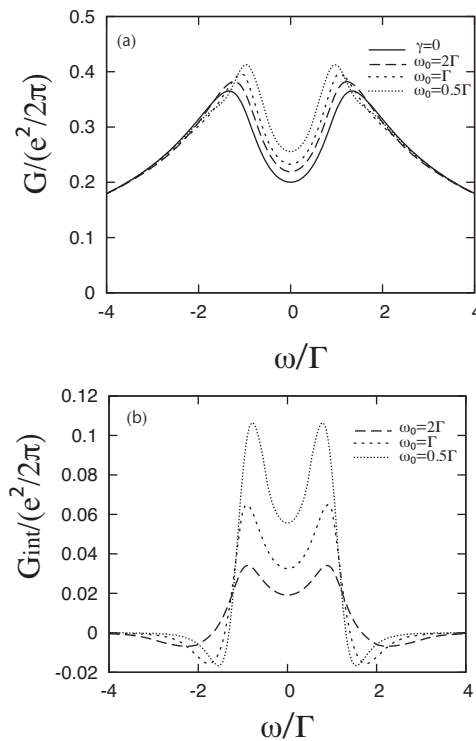
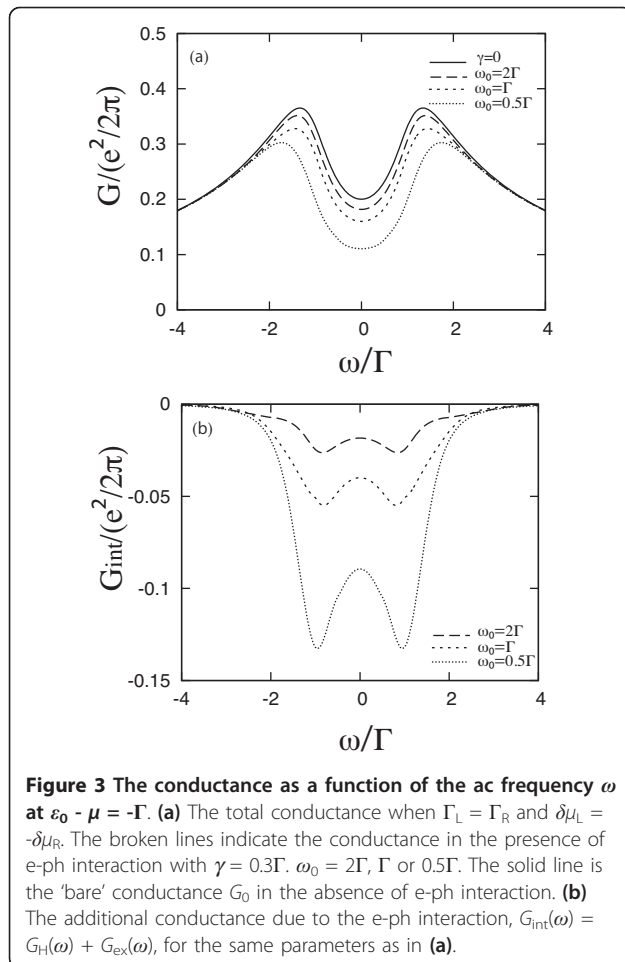


Figure 2 The ac conductance as a function of the ac frequency ω at $\varepsilon_0 - \mu = \Gamma$. **(a)** The total conductance when $\Gamma_L = \Gamma_R$ and $\delta\mu_L = -\delta\mu_R$. The broken lines indicate the conductance in the presence of e-ph interaction with $\gamma = 0.4\Gamma$, $\omega_0 = 2\Gamma$, or 0.5Γ . The solid line is the 'bare' conductance G_0 , in the absence of e-ph interaction. **(b)** The additional conductance due to the e-ph interaction, $G_{\text{int}}(\omega) = G_{\text{H}}(\omega) + G_{\text{ex}}(\omega)$, for the same parameters as in **(a)**.

interaction, G_{int} , for the same parameters. Similar results arise for all positive $\varepsilon_0 - \mu$. Both G_{H} and G_{ex} show two sharp peaks around $\omega \sim \pm (\varepsilon_0 - \mu)$ (causing the increase in G and the shift in its peaks), and both decay rather fast outside this region. In addition, G_{ex} also exhibits two negative minima, which generate small 'shoulders' in the total G . For $\varepsilon_0 > \mu$, G_{int} is dominated by G_{ex} . The exchange term virtually creates a polaron level in the molecule, which enhances the conductance. The amount of increase is more dominant for lower ω_0 . The situation reverses for $\varepsilon_0 < \mu$, as seen in Figure 3. Here, G_0 remains as before, but the ac conductance is suppressed by the e-ph interaction. Now G_{int} is always negative, and is dominated by G_{H} . The Hartree term of the e-ph interaction shifts the energy level in the molecule to lower values, resulting in the suppression of G . The amount of decrease is larger for lower ω_0 .

Conclusion

We have studied the additional effect of the e-ph interaction on the ac conductance of a localized level, representing a molecular junction. The e-ph interaction



enhances or suppresses the conductance depending on whether $\varepsilon_0 > \mu$ or $\varepsilon_0 < \mu$.

Abbreviations

e-ph: Electron-phonon.

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Author details

¹Department of Physics, Ben Gurion University, Beer Sheva 84105, Israel. ²Tel Aviv University, Tel Aviv 69978, Israel.

Authors' contributions

AU carried out the analytical and numerical calculations of the results and drafted the manuscript. OE conceived of the study. AA participated in numerical calculations. All authors discussed the results and commented and approved the manuscript.

Competing interests

The authors declare that they have no competing interests.

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