FIRST PRINCIPLES DESCRIPTION OF INELASTIC SCATTERING IN MOLECULAR CONDUCTORS

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The intense investigations of molecular devices, as an alternative to standard electronics components, underlines the need to model the conduction mechanisms of nano-scale conductors. In addition to measurements of the elastic conductance through point-contacts, atomic-wires and molecules, recent experiments have shown inelastic tunneling currents through, for example, a hydrogen molecule attached to platinum contacts [1], atomic gold wires [2] and alkanedithiol molecules [3]. In this work we calculate the inelastic conductance of these three systems, using an ab-initio (DFT) method and/or simpler models to describe the systems. The conductance is calculated by solving the Non Equilibrium Greens Function (NEGF) equations. We have also developed an approximate treatment that both simplifies the calculations dramatically and allows for transparent interpretations of the results.

The inelastic conductance is calculated using the Self Consistent Born Approximation (SCBA) in conjunction with the NEGF method. Since the emission/absorption of phonons couple the density of states (DOS) at energies separated by a phonon energy, the solution of the NEGF equations becomes computationally difficult and the results non-transparent. Assuming a weak electron-phonon interaction and constant DOS close to the Fermi-energy, we have overcome these difficulties by using Lowest Order Perturbation Theory (LOPT). The LOPT gives analytical formulas for the inelastic current that simplifies interpretation and lowers the computational burden by several orders of magnitude.

To model the conductance measurements of a hydrogen molecule connected to platinum contacts, we have derived analytical formulas for a one level model. These formulas show an almost perfect fit to the measured conductance (see Fig. 1) with a conductance drop at the phonon energy (50 meV). The linear conductance drop at higher bias is caused by absorption / stimulated emission of phonons as the molecule is heated by the current through the device. Noteworthy is that the only way to fit the experimental results is to assume that the damping of the phonons by the contacts (γ_d), i.e., escape of the phonons into the contacts, is much smaller than the damping by the electrons ($\gamma_{e\text{-}h}$) which is reasonable because of the large difference in the mass between hydrogen and platinum. A similar analytical model of the current through a gold-wire shows, in contrast to the hydrogen experiment, that the escape rate of phonons to the contacts is not negligible.

Using the Siesta DFT code [4] we have developed methods for calculating phonon energies, electron phonon interaction as well as describing semi-infinite contacts. These methods have been used to model the inelastic current through atomic gold wires with good results [5]. However, calculations on the alkanedithiol molecule reveal several discrepancies between theory and the experimentally measured conductance. We briefly discuss our calculated results and possible causes of the discrepancy.

References:

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Figures:

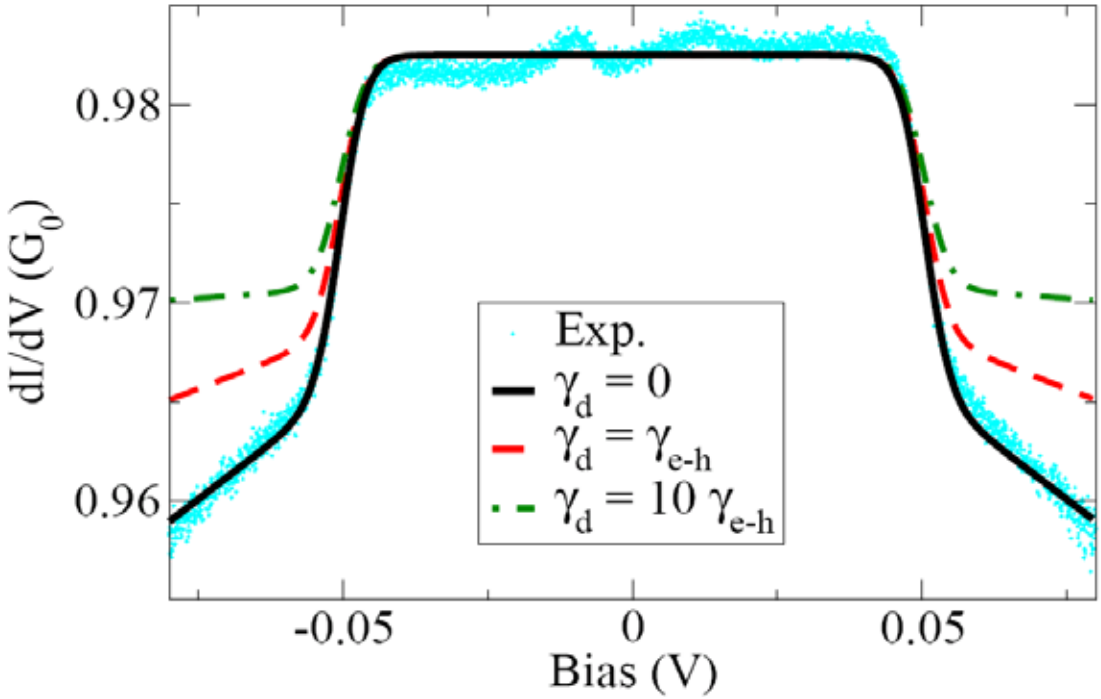

Figure 1: Comparison between theory and the experimentally measured conductance [6] through a deuterium molecule connected to platinum electrodes. The conductance shows a clear conductance step at the phonon energy and a linear decrease caused by heating at higher bias. The theory reveals that the escape-rate of the phonons from the molecule into the contacts (γ_d) has to be small compared to the damping caused by electron-hole pair creating ($\gamma_{e\text{-}h}$).