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Heat Dissipation and Non-Equilibrium Phonon Distributions in Molecular Devices

A. Pecchia*, A. Gagliardi†, Th. Frauenheim†, A. Di Carlo*
*Dep. of Electronics Engineering, University of Rome 'Tor Vergata', Via del Politecnico 1, 00133, Roma, Italy
†Theoretishe Physik, Universitat Paderborn, D-33098 Paderborn, Germany
e-mail: pecchia@ing.uniroma2.it

ABSTRACT

Using a density functional approach we compute vibrations of molecules adsorbed on metal and semiconducting substrates and the electron-phonon coupling of these modes. A non-equilibrium Green's function approach is used to compute the partially coherent transmission in molecular junctions due to electron-vibration scattering [1], [2]. The electronic power dissipated into molecular vibrations allows to set a rate equation for the phonon population in the vibrational degrees of freedom of the molecule. The rate equation includes the phonon emission rate and phonon decay due to absorption, electron-hole pair production and dissipation into the contacting leads, which are assumed to behave as reservoirs. The rate of phonon decay is computed using a microscopic approach which includes a first-principle calculation of the coupling of the molecular modes with the vibrations of the contacts. In turn, the calculated phonon lifetime is used to correct the phonon propagator. As the power dissipated in the molecular junction depends non-trivially on the phonon populations, the equilibrium distribution under bias condition is a complex issue.

A self consitent loop allows to compute the steady state non-equilibrium phonon population of the molecular junction under bias condition. We find that the resulting average population is far from the equilibrium thermal distribution, frequently assumed in such calculations, which allows to obtain a mean temperature of the junction. As expected the deviations increase with applied bias.

As molecular electronics is moving to semiconducting substrates, it is relevant to explore and understand thermal issues. Metallic substrates have in fact a very efficient channel of phonon damping into electron-hole pairs, while vibrational coupling to the substrate may not be very efficient due to ionic mass mismatch and a weaker bond. On the other hand the the electron-hole generation is prevented on semiconducting substrates, where instead vibrational coupling is stronger. We compare thermal behavior of organic molecules adsorbed on Si with molecules adsorbed on metallic substrates (Au or Cu) and determine the molecular temperature as a function of applied bias and for different temperatures of the termostats.

PRELIMINARY RESULTS

While the electrons cross the system, they interact with the molecular ionic vibrations from which they can be inelastically scattered. The electron-phonon scattering within the leads is not considered. The electronic system is described via a single-particle tight-binding Hamiltonian derived from Density functional theory (DFTB) [3], [4]. The method has been extended to the non-equilibrium Green's function (NEGF) approach [**?**]. In order to study the electron-phonon coupling we consider the first order diagram in the expansion of the Green's functions [6], [7] as

$$\Sigma_{ph}^{<,>}(\omega) = i\sum_q \gamma_q^2 \int \frac{d\omega'}{2\pi} G^{<,>}(\omega-\omega') D_{0,q}^{<,>}(\omega'), \tag{1}$$

where the $D_{0,q}^{<,>}$ are the correlation functions related to the vibrational modes. The power dissipation is obtained using the virtual contact concept [1].

In this illustrative example we consider a di-thio-phenyl molecule bridging two metal contacts. The analysis is restricted to those vibrational modes which give non-negligible incoherent

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electron-phonon scattering [8]. These have frequencies of w_q =756 cm^{-1}, w_q =1147 cm^{-1}, w_q =1182 cm^{-1} and w_q =1754 cm^{-1}, respectively. The molecule and its vibrational modes are represented in Figure 1.

The coupling of the vibrational modes with the reservoirs gives a phonon decay rate of the order of 10^{-13} sec. The self-consistent solution of the steady-state phonon population of the different modes gives the results shown in Figures 2 and 3.

Figure 2 shows the power dissipated and the non-equilibrium equivalent temperature of each mode as a function of applied bias for a contact temperature of 300 K. Similarly, Figure 3 is computed for a contact temperature of 100 K. It is possible to see that the highest energy modes heat up considerably, reaching a temperature of almost 600 K, while low energy modes are less sensitive. This is related to the larger power emitted in such modesand the fact that the power emitted depends on the population itself. Since the lowest modes have an equilibrium population larger than N=1 the absorption probability becomes similar to the emission, decreasing the net emission rate.

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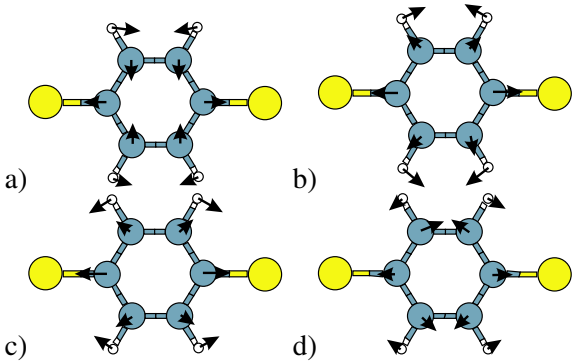

Fig. 1. Vibrational mode representation of the most important modes for incoherent electron-phonon scattering. a) 756 cm^{-1}, b) 1147 cm^{-1},c) 1182 cm^{-1},d) 1754 cm^{-1}.

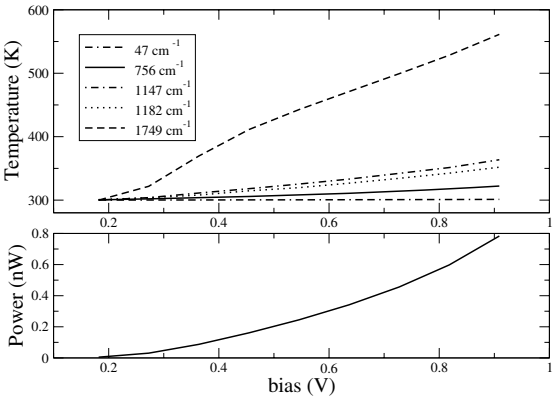

Fig. 2. Effective mode temperature and total power dissipated into the molecule as a function of bias for a reservoir temperature of 300 K.

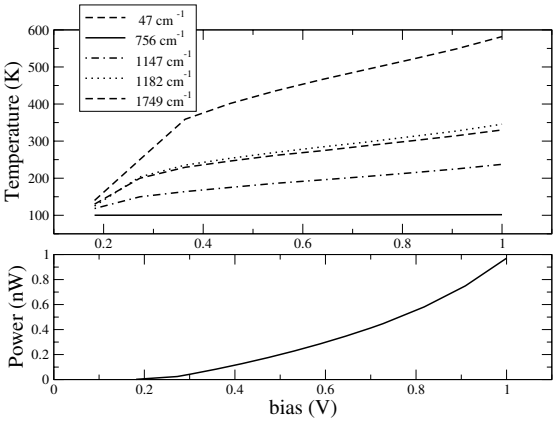

Fig. 3. Like figure 2 but for a reservoir temperature of 100 K