

Atomistic simulation of the electronic transport in organic nanostructures: electron-phonon and electron-electron interactions.

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The field of molecular electronics has seen a tremendous expansion in recent years, thanks to the realization of ingenious experimental setups and the fundamental achievement of reproducible results and behaviours. Significant progresses have also been made from a theoretical point of view, although the agreement with experiments is still not satisfactory. The challenges for a complete understanding of transport in such systems are still considerable.

Recently, we have extended our density-functional tight-binding (DFTB) simulator for transport computations [1-3] to the non-equilibrium Green's function approach (NEGF). This scheme allows to treat contacts and molecules to an equal footing and the computation of the current flowing between the contacts in a fully self-consistent manner with the open boundary and non-equilibrium conditions encountered in such transport problems.

Particular attention is devoted to the evaluation of the electron-phonon interactions in molecular systems and the role of Joule heating and thermal dissipation. The problem is tackled within the formalism of NEGF by the calculation of appropriate electron-phonon self-energies. The electron-phonon coupling is derived from the DFTB Hamiltonian. The Power dissipated is calculated from the virtual contact current [4] originated from phonon emission and absorption processes.

As well known, DFT tends to underestimate the electronic band-gap of semiconducting and insulating materials, due to the mean-field approximation of the exchange-correlation energy functional. In particular the band-gap of conjugated organic molecules is usually underestimated by few electronvolts. However, band-gap corrections are crucial for quantitatively correct calculations of the tunneling current through organic molecules.

We show a novel implementation of the *GW* correction applied to our DFTB method and show its applications to molecular systems sandwiched in-between electrodes to obtain a first-principle correction of the *e-e* interaction energy. The resulting self-energy is used to correct the system *GF* and to obtain a correction of the tunneling current, beyond DFT.

[1] A. Di Carlo, et al. , *Physica B*, **314**, 86 (2002)

[2] A. Di Carlo, et al., *J. Comp. Elect.*, **1**, 109 (2002)

[3] A. Pecchia and A. Di Carlo, *Rep. Prog. Phys.*, in press.

[4] S. Datta, "Electronic Transport in Mesoscopic Systems", Cambridge Univ. Press, page 319.

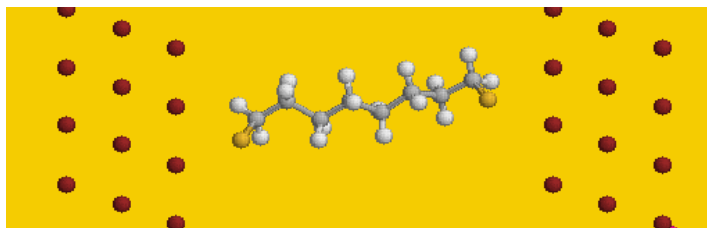


Figure 1. Molecular system comprising a *octane-dithiol* bridging two Au tips. The geometry and vibrational modes are calculated with DFTB.

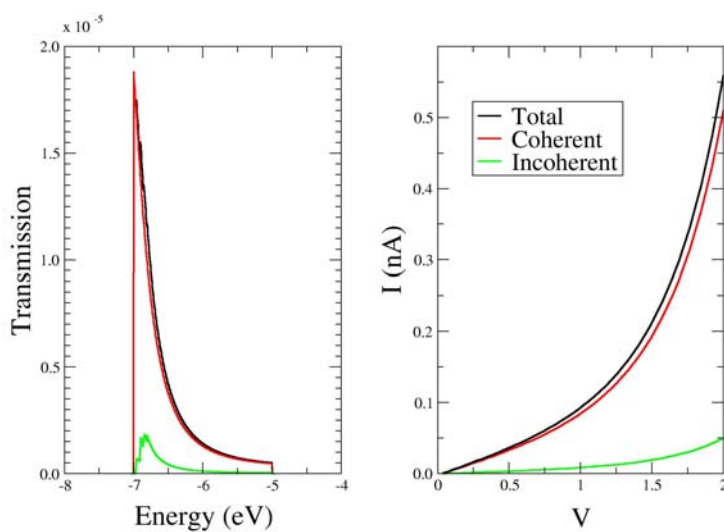


Figure 2. Left. Transmission through the system shown in Figure 1 with electron-phonon coupling at the bias voltage of 2.0 V and $T=0$ K. Right. I-V characteristics. Color curves show the contributions of the coherent and incoherent current.

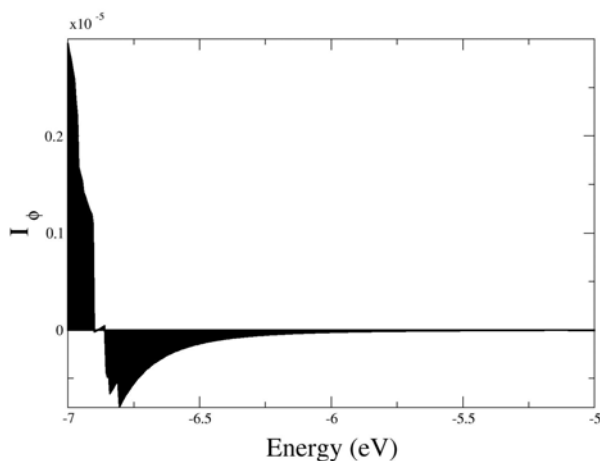


Figure 3. Current density at the virtual reservoir, calculated for a bias of 2.0 V and $T=0$ K. Negative (positive) sign corresponds to a net incoming (outcoming) current. The corresponding total power dissipated is ~ 4 pW.

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