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

Alessandro Pecchia[1]*, Alessio Gagliardi[1,2], Th. Niehaus[2], Th. Frauenheim[2], Aldo Di Carlo[1], Paolo Lugli[3]

[1] *Dip. di Ingegneria Elettronica, Universita` di Roma Tor Vergata, 00133 Roma (Italy)*
[2] *Dept. of Theoretical Physics, University of Paderborn, D-33098, Paderborn (Germany)*
[3] *Institute for Nanoelectronics, Technical University of Munich, D-80333 München (Germany)*
*e-mail: pecchia@ing.uniroma2.it

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 sandwitched 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, behond DFT.

[1] A. Di Carlo, et al. , Phisica 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.

A full journal publication of this work will be published in the Journal of Computational Electronics.
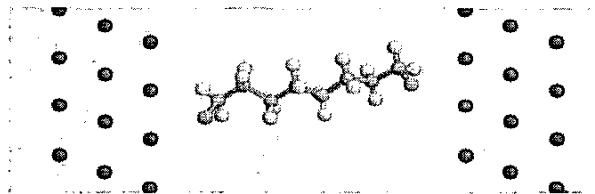
Figure 1. Molecular system comprising a *octane-dithiol* bridging two *Au* tips. The geometry and vibrational modes are calculated with DFTB.
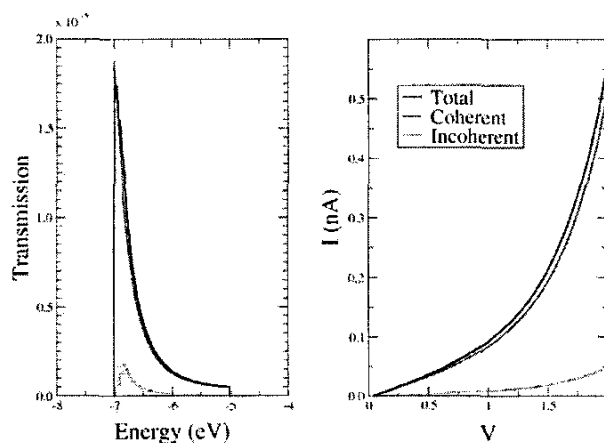

Figure 2. Left. Transmission through the system shown in Figure 1 with electron-phonon coupling at the bias voltage of 2.0 V and T=0 K. Right. I-V characteristics. Color curves show the contributions of the coherent and incoherent current.
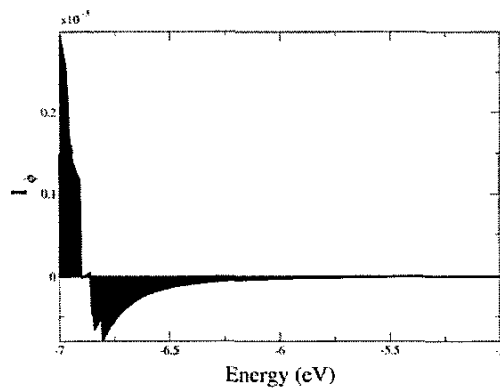

Figure 3. Current density at the virtual reservoir, calculated for a bias of 2.0 V and T=0 K. Negative (positive) sign corresponds to a net incoming (outcoming) current. The corresponding total power dissipated is ~4 pW.