Long-time electron dynamics of open molecular junctions

Gianluca Stefanucci, Dept. of. Phys., Tor Vergata

Abstract: We use non-equilibrium Green's function theory to study the asymptotic time-evolution of a noninteracting electrode/molecule/electrode system for different histories of the applied bias and different initial embeddings. We show that in the absence of bound states history effects and initial embedding correlations are washed out in the long-time limit and that the current can be expressed in terms of a Landauer-like formula. This is no longer guaranteed if the biased Hamiltonian has bound-state eigensolutions. In this case the system does not evolve towards a steady regime and both current and density oscillate with frequencies given by bound-bound transition energies. The amplitude of the oscillations depend on the history of the bias as well as on the initial conditions. The consequences of the above result are discussed in the framework of Time-Dependent Density Functional Theory which has the merit of including electron-electron interactions in a one-particle picture. For steady-state situations we show how to generalize the Landauer formula to include exchange-correlation effects in the total current.