Multi-configuration electronic Scattering matrix calculations for electron tunneling through a metal-molecule-metal junction

Mathilde Portais
GNS-CEMES-CNRS
29 rue Jeanne Marvig, BP 94347
31055 Toulouse Cedex 4, France

Lately, the inclusion of Coulomb and exchange interactions for tunnel current intensity calculations has become an important challenge. For example, some experimental results disagree with the common mono-electronic interpretation that an STM image of a molecule at a given electronic tunneling resonance is representative of the spatial electronic density of one (or a few) molecular orbitals of this molecule [1,2]. So, it is now important to describe the corresponding metal-molecule-metal junction as a many-body electronic system using a full Slater determinant basis set instead of mono-electronic states of this junction.

Here we present a method to calculate the scattering matrix and the corresponding electronic transmission spectrum of a metal-molecule-metal tunnel junction described using an electronic multi-configuration basis set. This new elastic scattering calculation (ESQC) like method is applicable to nano-scale systems where the interconnection electrodes can be considered in a ballistic regime of transport and where the N-electrons electronic structure of the central metal-molecule-metal nano-junction is taken into account. Simple applications demonstrate that the resonances of the electronic transmission spectrum of such a junction can be interpreted as instantaneous multi-electronic states fluctuations. They are created by the multiple possible virtual excitations of the metal-molecule-metal junction electronic structure induced by the tunneling electrons transferred through this junction.

References:

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[2] Soe W.H, Wong H.S, Manzano C, Grisolia M, Hliwa M, Feng X, Mullen K and Joachim C, ACS Nano, 6, 3230 (2012).