Sources, Sinks And Electron Correlations

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A description of quantum transport using ab initio electronic structure methods continues to be a challenge. Going back to the basics of electronic structure theory permits an accurate treatment of the quantum many-electron problem [1,2]. From this picture, approximations can be devised that allow for treatment of larger systems to be simulated and to reduce the computational demand to enable engineering simulations for molecular transport [3,4]. However, the action of electrodes in contact with a molecular region must be described within a many-body theory to allow for calculation of current-voltage characteristics. The conditions describing electrodes or electron reservoirs are expressed in terms of single particle pictures and need to be generalized for the many-body problem. Generalization of the reservoir boundary conditions has been achieved using the Wigner transform of the one-body reduced density matrix [1]. In addition, electrons leaving the molecular region must flow into the electrodes and equilibrate and this property needs to be incorporated into a description of the electrodes [5]. In this talk, we present a configuration interaction scheme for transport calculations with sources generated from the Wigner distributions of in-coming electrons, and electron sinks within the electrodes are generated from the use of complex absorbing potentials [5].

Explicit treatment of electron correlations allows us to investigate physics beyond the single-particle pictures of transport. Applying our approach to tunnelling currents observed in molecular junctions, we identify a criterion for defining a “best” independent particle model [3]. Maximizing the overlap of a Slater determinant composed of single electron states to a many-body current-carrying state is more important than energy minimization for defining single particle approximations in a system with open boundary conditions [6]. Generalization of our approach allows for a treatment for quantum mechanical modeling of quantum devices and systems [7].

[1] P. Delaney and J. C. Greer, Physical Review Letter, **93**, 036805 (2004)

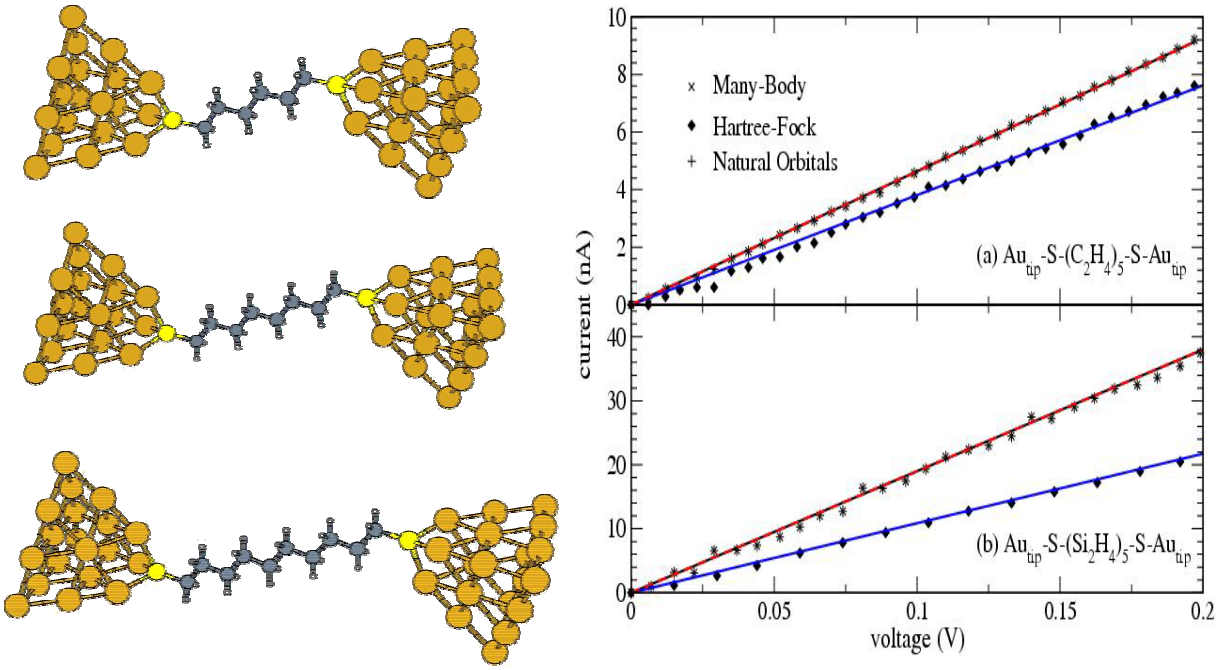
[2] P. Delaney and J. C. Greer, International Journal of Quantum Chemistry, **100**, pp. 1163-1169 (2004)

[3] J. Fransson, O. Bengone, J. A. Larsson, and J. C. Greer, IEEE Transactions on Nanotechnology, **5**, pp. 745-749 (2006)

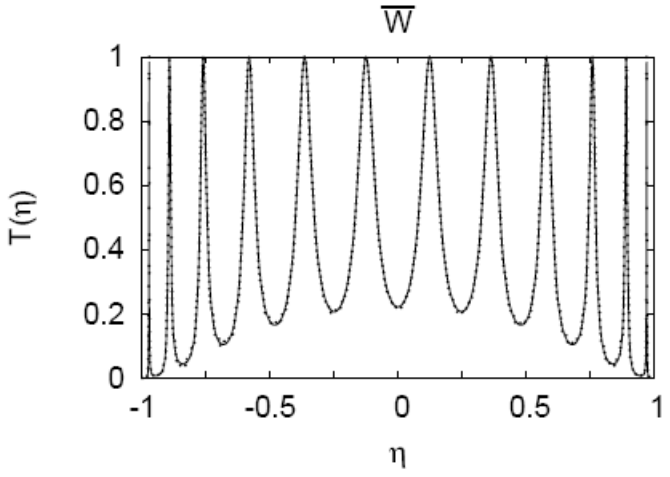
[4] T. M. Henderson, G. Fagas, E. Hyde, and J. C. Greer, Journal of Chemical Physics, accepted (2007)

[5] G. Fagas, P. Delaney, and J. C. Greer, Physical Review B **73**, 241314(R) (2006)

[6] P. Delaney and J. C. Greer, Proceedings of the Royal Society A: Mathematical, Physical and Engineering Sciences, **462**, pp. 117-135 (2006)



Comparison of different tunnelling characteristics for alkane and silane chain using different theoretical methods



State broadening for transmission resonances using a complex absorbing potential that is directly determined from the electron self-energies in the electrodes