MAR08-2007-005337

Abstract for an Invited Paper
for the MAR08 Meeting of
the American Physical Society

First-principles studies of electrical transport in nanoscale molecular junctions[1]

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Understanding the conductance of individual molecular junctions is a forefront topic in theoretical nanoscience. The development of a general, efficient atomistic approach for treating an open system out of equilibrium with good accuracy, and then using it to inform experiment, is a significant open challenge in the field. Here I will describe studies where first-principles techniques, based on density functional theory (DFT) and beyond, are used to investigate some of the fundamental issues associated with single-molecule transport measurements. After a brief summary of previous work, a DFT-based scattering-state approach is presented and applied to H_2 and amine-Au linked molecular junctions [1], two systems for which there exist reliable data [2]. Similar to most ab initio studies, we rely on a Landauer approach within DFT for junction conductance. Using this framework, which has proven relatively accurate for metallic point contacts, good agreement with experiment is obtained for the H_2 conductance. For amine-Au linked junctions, however, the computed conductance is significantly larger than that measured,although structural trends are reproduced by the calculations. To explore this further, we draw on GW calculations of a prototypical metal-molecule contact, benzene on graphite, where interfacial polarization effects are found to drastically modify frontier orbital energies [3]. A physically motivated model self-energy correction is developed from our GW calculations,applied to the amine case, and shown to quantitatively explain the discrepancy with experiment. The importance of many-electron corrections beyond DFT for accurately computing molecular conductance and understanding experiments is thoroughly discussed. [1] S. Y. Quek *et al.*, Nano Lett **7**, 3482 (2007); K. H. Khoo *et al.*, submitted (2007). [2] R. Smit *et al.*, Nature **419**, 906 (2002); L. Venkataraman *et al.*, Nature **442** ,904 (2006). [3] J. B. Neaton *et al.*, Phys. Rev. Lett. **97**, 216405 (2006).

[1]This work was supported by the U.S. Department of Energy under Contract No. DE-AC02-05CH11231.