First-Principles Approaches to Charge Transport and Energy Conversion with Self-Energy Corrections

J. B. Neaton
Molecular Foundry
Lawrence Berkeley National Laboratory

A fundamental challenge in nanoscience is to understand and control the flow of charge at the molecular scale. Recent reliable transport measurements of single-molecule junctions have provided a unique opportunity to improve upon standard first-principles approaches for computing observables related to charge dynamics, while quantitatively exploring foundational mechanisms of molecular-scale transport. In this talk, I will describe the development and application of a new, parameter-free computational approach to charge transport at the nanoscale, DFT+Σ, which has led to good agreement with recent measurements of conductance [1-3] and thermopower of single-molecule junctions, and photoemission spectroscopy on related systems [4]. Based on first-principles density functional theory (DFT), DFT+Σ [5] improves Kohn-Sham junction level alignment with a physically-motivated approximate self-energy correction, Σ, based on first-principles GW calculations of metal-molecule interfaces. In addition to describing recent results, advantages and limitations of this method will be discussed in detail.

[1] S. Y. Quek, L. Venkataraman, H. J. Choi, S. G. Louie, M. S. Hybertsen, and J. B. Neaton, Nano Lett. **7**, 3477 (2007)
[2] S. Y. Quek, M. Kamenetska, M. L. Steigerwald, H. J. Choi, S. G. Louie, M. S. Hybertsen, J. B. Neaton, and L. Venkataraman, Nature Nano. **4**, 230 (2009)
[3] S. Y. Quek, H. J. Choi, S. G. Louie, and J. B. Neaton, Nano. Lett. **9**, 3949 (2009)
[4] M. Dell'Angela, G. Kladnik, A. Cossaro, A. Verdini, M. Kamenetska, I. Tamblyn, S.Y. Quek, J. B. Neaton, D. Cvetko, A. Morgante, L. Venkataraman, submitted (2010)
[5] S. Y. Quek, D. Strubbe, H. J. Choi, S. G. Louie, and J. B. Neaton, to be published (2010)