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Understanding the Conductance of Single-Molecule Junctions from First Principles[1]

SU YING QUEK, Molecular Foundry, Lawrence Berkeley National Laboratory

Discovering the anatomy of single-molecule junctions, in order to exploit their transport behavior, poses fundamental challenges to nanoscience. First-principles calculations based on density-functional theory (DFT) can, together with experiment, provide detailed atomic-scale insights into the transport properties, and their relation to junction structure and electronic properties. Here, a DFT scattering state approach [1] is used to explore the single-molecule conductance of two prototypical junctions as a function of junction geometry, in the context of recent experiments. First, the computed conductance of 15 distinct benzene-diamine-Au junctions is compared to a large robust experimental data set [2]. The amine-gold bonding is shown to be highly selective, but flexible, resulting in a conductance that is insensitive to other details of the junction structure. The range of computed conductance corresponds well to the narrow distribution in experiment, although the average calculated conductance is approximately 7 times larger. This discrepancy is attributed to the absence of many-electron corrections in the DFT molecular orbital energies; a simple physically-motivated estimate for the self-energy corrections results in a conductance that is much closer to experiment [3]. Second, similar first-principles techniques are applied to a range of bipyridine-Au junctions. The extent to which Au-pyridine link bonding is affected by the constraints of forming bipyridine-Au junctions is investigated. In some contrast to the amine case, the computed conductance shows a strong sensitivity to the tilt of the bipyridine rings relative to the Au surfaces. Experiments probing the conductance of bipyridine-Au junctions are discussed in the context of these findings. [1] H. J. Choi et al, Phys Rev B, 76, 155420 (2007) [2] L. Venkataraman et al, Nano Lett 6, 458 (2006) [3] S. Y. Quek et al, Nano Lett. 7, 3477 (2007)

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