

Abstract Submitted
for the MAR13 Meeting of
The American Physical Society

Theory of Solvent-Mediated Environmental Effects on Transport in Molecular Junctions MICHELE KOTIUGA, Department of Physics, UC Berkeley & Molecular Foundry, LBNL, PIERRE DARANCET, Molecular Foundry, LBNL & Department of Applied Physics and Applied Mathematics, Columbia, LATHA VENKATARAMAN, Department of Applied Physics and Applied Mathematics, Columbia, JEFFREY NEATON, Molecular Foundry, LBNL — Single-molecule junctions, formed with well-defined and robust metal-molecule contacts, can provide an ideal model system to study mechanisms of charge transport at the molecular scale. However, the presence of solvent is often unavoidable, and recent experiments have shown that the junction conductance can be altered by a factor of two depending on the solvent present. Here, we use a first-principles scattering-state approach, based on self-energy corrected density functional theory (DFT), to explore how solvent and coverage impacts the transmission and conductance of bipyridine-Au molecular junctions. We find the conductance can shift by more than a factor of 5 by varying the bipyridine coverage, which is an effect associated with work function shifts that can be understood with a 2D polarizable dipole model fit with DFT values. A generalization of this electrostatic model to include solvent molecules allows us to estimate the work function shift for a mixed molecular coverage based both on the experimental parameters and system thermodynamics. By combining the results of our transmission calculations and the electrostatic model, we can accurately describe the conductance shifts observed experimentally. We acknowledge DOE for support, and NERSC for computational resources.

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Date submitted: 14 Nov 2012

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