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Probing Transport through Single Molecule Junctions by Electrolytic Gating¹ BRIAN CAPOZZI, Department of Applied Physics and Applied Mathematics, Columbia University, New York, Q. CHEN, Columbia University, P. DARANCET, Molecular Foundry, Lawrence Berkeley National Labs, M. BUZZEO, Barnard College, J.B. NEATON, Molecular Foundry, Lawrence Berkeley National Labs, C. NUCKOLLS, L. VENKATARAMAN, Columbia University — Organic field effect transistors made using ionic liquids or electrolyte solutions as gate dielectrics have received significant attention due to their ability to generate huge interfacial capacitances at the nano-scale. We apply this technique to single-molecule junctions created using the scanning tunneling microscope-based break-junction technique. We demonstrate that we can tune the transport characteristics of single-molecule junctions, modulating the conductance of junctions with molecules that are electrochemically inactive, within the gate bias range probed. For molecules that conduct through the highest occupied molecular orbital (HOMO), we see a decreasing conductance while applying a positive electrochemical gate potential while those that conduct though the lowest unoccupied molecular orbital (LUMO) show the opposite trend. Furthermore, we are able to fit the experimental gating data with a Lorentzian transmission function, and find the fitting parameters to be in quantitative agreement with self-energy corrected density functional theory calculations. This work shows that electrolyte gating can directly modulate the alignment of the conducting orbital relative to the metal Fermi energy, thereby changing the junction transmission characteristics.

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Brian Capozzi Department of Applied Physics and Mathematics, Columbia University, New York

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