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Electronic level alignment at metal-molecule interfaces from first principles[1] JEFFREY B. NEATON, The Molecular Foundry, LBNL, MARK S. HYBERTSEN, Department of Applied Physics and Applied Mathematics and Center for Transport in Molecular Nanostructures, Columbia University, STEVEN G. LOUIE, The Molecular Foundry, LBNL and Department of Physics, UC-Berkeley — Electronic transport through nanoscale molecular junctions critically depends on the energetic alignment of frontier molecular states with the contact Fermi levels. In this work, a first-principles Greens function approach is used to explore how frontier molecular energy levels are modified at metal-molecule interfaces. The electronic structure of a model interface, benzene on graphite (0001), is computed using the GW approximation for the electron self-energy operator. Upon adsorption on the surface, the benzene HOMO-LUMO gap is predicted to be 7.2 eV, substantially reduced from its calculated gas-phase value of 10.5 eV, and slightly smaller than its computed solid-phase gap of 7.5 eV. This decrease is attributed to the change in the electronic correlation energy of the frontier states in different environments. Comparison with a classical image interaction provides a quantitative measure of the contribution of the molecule-substrate coupling to the gap narrowing of the molecule.

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