

Abstract Submitted
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Self-energy-corrected electronic energy level alignment in molecular junctions and at interfaces with hybrid functionals¹ MICHELE KOTIUGA, Physics Department, UC Berkeley & Molecular Foundry, LBNL, DAVID EGGER, LEEOR KRONIK, Department of Materials and Interfaces, Weizmann Institute of Science, JEFFREY B. NEATON, Physics Department, UC Berkeley & Molecular Foundry, LBNL & Kavli Energy NanoSciences Institute at Berkeley, Berkeley — Accurate calculations of energy level alignment at complex interfaces are imperative for understanding a variety of transport and spectroscopy measurements, as well as for elucidating new interfacial electronic structure phenomena. However, standard approaches to such calculations, based on density functional theory (DFT), are well known to be deficient. In prior work on molecular junctions and physisorbed molecules on surfaces, an approximate GW approach, DFT+ Σ , has been successful in describing the conductance and level alignment of amine and pyridine terminated molecules on gold surfaces and in junctions. Here, via the use of hybrid functionals, we perform quantitative studies of the level alignment of thiol- and carbon-terminated phenyls on gold, where the formation of a strong chemical bond and presence of gateway states limit the validity of the DFT+ Σ approximation as currently formulated. We contrast these systems to prior work on weakly-coupled molecules, including bipyridine or phenyl-diamines. Additionally, we compute transmission functions using both DFT-PBE and DFT-HSE starting points and predict conductance and thermopower with these methods, comparing to experiments where possible.

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