DS 33: Frontiers of Electronic Structure Theory (Joint session of DS and O, organized by DS)

Time: Wednesday 12:00–12:45 Location: H11

DS 33.1 Wed 12:00 H11

Towards Accurate Energy Level Alignment at Physisorbed Molecule-Metal Interfaces from Density Functional Theory — •David A. Egger[1], Zhenfei Liu[2], Jeffrey B. Neaton[2,3,4], and Leeor Kronik[1] — [1]Department of Materials and Interfaces, Weizmann Institute of Science, Rehovoth 76100, Israel — [2]Molecular Foundry and Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, United States — [3]Department of Physics, University of California, Berkeley, California 94720, United States — [4]Kavli Energy Nanosciences Institute at Berkeley, Berkeley, California 94720, United States

A key physical quantity for electronic transport in nanostructured molecule-metal interfaces is the energy level alignment of the molecular electronic states with respect to the Fermi level of the metal. Here, we introduce an efficient theoretical method that is based on density functional theory, but in contrast to common approximations fulfills physically motivated criteria for exchange-correlation interactions and can therefore yield quantitatively accurate energy level alignment information for physisorbed metal-molecule interfaces.[1] We validate our approach by a detailed comparison with experimental and theoretical reference data for several prototypical interfaces of this kind: benzene on graphite (0001), and 1,4-benzenediamine, Cu-phthalocyanine, and 3,4,9,10-perylene-tetracarboxylic-dianhydride on Au(111). Our results indicate that obtaining quantitatively accurate energy level alignment information from density functional theory is possible.

[1] Nano Lett. 15, 2448 (2015)

DS 33.2 Wed 12:15 H11

Fermi orbital self-interaction corrected electronic structure of molecules beyond local density approximation — •Torsten Hahn[1], Simon Liebing[1], Jens Kortus[1], and Mark Pederson[2] — [1]Institute for Theoretical Physics, TU Freiberg, 09599 Freiberg, Germany — [2]Department of Chemistry, Johns Hopkins University, Baltimore, USA

The correction of the self-interaction error that is inherent to all standard density functional theory (DFT) calculations is an object of increasing interest. We present our results on the application of the recently developed Fermi-orbital based approach [1,2] for the self-interaction correction (FO-SIC) to a set of different molecular systems [3]. Our study covers systems ranging from simple diatomic to large organic molecules. Our focus lies on the direct estimation of the ionization potential from orbital eigenvalues and on the ordering of electronic levels in metal-organic molecules. Further, we show that the Fermi orbital positions in structurally similar molecules appear to be transferable.

[1] M. R. Pederson, A. Ruzsinszky, and J. P. Perdew, J. Chem. Phys. 140, 121103 (2014). [2] M. R. Pederson, J. Chem. Phys. 142, 064112 (2015). [3] T. Hahn et al., J. Chem. Phys. (accepted: AIPID 029546JCP).

DS 33.3 Wed 12:30 H11

Charge and spin transport in two-dimensional systems using the KKR-CPA-Kubo approach — •S. Wimmer, K. Chadova, S. Bornemann, D. Ködderitzsch, and H. Ebert — Department Chemie, Ludwig-Maximilians-Universität München

We have applied the fully relativistic spin-polarized Korringa-Kohn-Rostoker method to investigate charge and spin transport properties of various two-dimensional systems such as mono- and multilayers, surfaces and thin films on surfaces. Our theoretical approach is based on the linear response Kubo formalism that allows introducing a layer-resolved conductivity σ^{IJ} [1] and that has been extended to the fully relativistic case. An implementation of the Coherent Potential Approximation for layered systems allows the treatment of disorder effects including the Vertex Corrections to the conductivity [2]. This can be used to study chemical disorder in alloys but also to include the effect of finite temperatures. For the latter the so-called Alloy-Analogy Model [3] is enployed to treat vibrations and spin fluctuations. Besides convergence tests on simple homogeneous systems the approach has been applied, e.g., to the Fe_n/GaAs(001) metal/semiconductor surface system, the topological insulator surfaces Bi_2X_3(111) (X = Se, Te, and $Se_{1-x}Te_x$), and, using a generalization of the Mott formula for thermoelectric transport [4], also to spincaloritronic transport.

[1] W.H. Butler *et al.*, *Phys. Rev. B* **52**, 13399 (1995). [2] W.H. Butler, *Phys. Rev. B* **31**, 3260 (1985); K. Palotás *et al.*, *Phys. Rev. B* **67**, 174404 (2003). [3] H. Ebert *et al.*, *Phys. Rev. B* **91**, 165132 (2015). [4] M. Jonson and G.D. Mahan, *Phys. Rev. B* **21**, 4223 (1980).