Modelling Injection Rates at Metal/Organic Interfaces

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Abstract

We present a model for charge injection at the interface between a metal and organic molecules. We focus on the interface ITO and the hole transporter DPBIC (Tris[(3-phenyl-1H-benzimidazol-1-yl-2(3H)-ylidene)-1,2-phenylene]Ir), relevant for phosphorescent OLEDs. Models for the amorphous In2-xSnxO3 are obtained using an optimized Buckingham force field and melting/quench cycles. Models of the hybrid interface are obtained either via a molecule-by-molecule simulation of deposition or via an annealing of the organic overlayer starting from bulk configurations [1]. The two simulations provide the limits of kinetically-driven and thermodynamically-driven depositions. Statistics of the orientations and distances of the molecules on the surface are extracted from the MD simulations and used for the calculation of the injection rates. These are obtained by calculating from first principles the ingredients of a Marcus transfer rate formula. The Quantum Espresso package is used for the calculation of the transfer integrals, based on the Hamiltonian partitioning and projection technique [2], here generalized to periodic systems and k-sampling. The transfer integrals are found to depend on distance but also are sensitive to the molecular orientation. However, a key parameter for injection is the energy level alignment between metal Fermi level and molecular HOMO level. DFT calculations are notoriously rather poor in accurately describing this alignment and perturbation methods such as GW are still challenging for the complex system at hand, comprising about 400 atoms. I will discuss progresses to obtain the alignment using constrained DFT as implemented in CP2K [3, 4]. The method is validated on DPBIC-DPBIC transfer rates [5] and results obtained for simpler model interfaces such as NPD/Au will be discussed.

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