First-Principles Based Multiscale Modeling of Dynamical Processes in Hybrid Materials Systems

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Detailed insight into electronic and molecular processes in hybrid material systems is a key driver for advances in application areas like nanotechnology, molecular electronics or drug delivery. While predictive-quality computational modeling assumes an increasing role in providing this insight, current methodology still battles with the strong variations in entropic contributions and electron localization that are often characteristic at solid-gas or solid-liquid interfaces. First-principles techniques are challenged in the description of molecular levels and concomitant electronically excited states in particular at metal surfaces. Many relevant molecular processes are rare events and correspondingly necessitate extended time-scale simulations that capture the complex and rough free energy barriers especially at solid-liquid interfaces. The need for methodological advances extends thereby over both accurate reference techniques and efficient approaches for a computational screening. I will review our recent activities in this context, focusing on functional organic molecules at surfaces, as well as on electron transfer in and dissolution from molecular crystals.

Multi-Scale Modeling of Step-Edge Barrier Crossing in HIOS

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We study the effects of a molecular step-edge barrier on the surface diffusion of a single para-sexiphenyl (p-6P) molecule physisorbed on the inorganic ZnO (10-10) surface by employing a multi-scale modeling approach, where we combine DFT, all-atom molecular dynamics ('force field') simulations, and diffusion theory. We calculate temperature- and charge-dependent (free) energy landscapes, position-dependent diffusion coefficients and ultimately the mean first passage time for crossing the step-edges. We find two completely different step-edge crossing pathways, the occurrence and rates of which simultaneously depend on both the electrostatic coupling to the surface atoms and the temperature of the system. Implications of the findings for the modeling and understanding of nucleation and growth in HIOS are briefly discussed.