Developing realistic models of interfaces from simulation

I will describe several recent pieces of work focussing on developing an atomistic-level understanding of interfaces. A variety of computational approaches are used, and compared to state-of-the-art experiments using the non-contact atomic force microscope (NC-AFM).

(i) Insulator – molecule – metal. In collaboration with NC-AFM experimental researchers at the University of Hamburg, we unambiguously identified the adsorption configurations of a complex molecule on ionic crystals (NaCl, NiO) and the nature of surface ions [1,2,3]. These systems can provide a sensitive test system to calibrate the accuracy of computational approaches and also act as a tape measure, providing an accurate measure of tip-sample distance, which is extremely hard to estimate independently. The identification of a permanent tip dipole inherent to metallic tips opens the way to using this mode of AFM operation for metrology of complex insulating surfaces.

(ii) Insulator – water. High-resolution imaging and force spectroscopy using AFM in solution opens a wide area of possible applications allowing real-time and real-space imaging of surfaces in solution. To obtain full benefit and provide a significant new analytical ability, it is vital to understand the underlying imaging mechanism(s) that can lead to high (atomic or molecular) resolution. Our simulations of solvated nanoparticles near surfaces show several possible mechanisms that lead to measurable force differences and image contrast over surface sites [4]. We used a variety of simulation methods to calculate the free energies of nanoparticle-surface interactions [5]. Water-mediated interactions can cause significant force differences above different surface sites, and are in most cases larger, and longer ranged, than the direct vacuum-like interactions. I will compare and contrast predicted imaging mechanisms from vacuum with solution.

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