Increasing the Scope of Computational Nanochemistry by Combining Density-Functional Theory with Machine-Learned Interatomic Potentials

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Computational chemistry has proven to be a valuable tool in understanding low-dimensional materials with atomic resolution. While the direct application of high-level ab initio methods to periodic systems is still very challenging and out of reach for most cases, density-functional theory (DFT) provides an ideal compromise of accuracy and computational speed and makes the routine treatment of hundreds of atoms possible [1]. In recent years, machine-learned interatomic potentials (MLIPs) emerged as an increasingly powerful tool to simulate much larger systems of thousands to tens of thousands of atoms with DFT accuracy, providing that they were parametrized adequately [2].

In this talk, I will give an overview of efficient applications of DFT and MLIPs to surface and interface systems, with the focus on the adsorption of complex molecules on metal surfaces and the formation and properties of two-dimensional nanomaterials. The close collaboration with experimentalists provides initial structural guesses by STM or LEED patterns, which are then relaxed with a suitable DFT functional. Calculated STM images can then be used to back-verify the experimental starting point [3]. Besides geometries, the explicit treatment of electrons by DFT enables a systematic explanation of binding and reactive processes on a quantum-mechanical level.

I will show two possible applications of MLIPs in the realm of surface chemistry. Firstly, large scale structures like moire patterns or herringbone-reconstructions can now be simulated as a whole, enabling the verification of, e.g., site-selective adsorption on them. Secondly, time-dependent phenomena like surface diffusions or modifications due to interactions with gas phase molecules can now be studied in real time via molecular dynamics (MD), opening an opportunity for studying the influence of minor changes in temperature or partial pressures on the resulting material.

During the talk, I will lay out how DFT and MLIPs can be used together as closely connected tools that open new possibilities in the computational treatment of low-dimensional materials. The outline will be combined with a couple of concrete examples, like the adsorption of porphin derivatives on a Cu(111) surface [4, 5], the adsorption of Br on graphene and h-BN layers a on Rh(111) surface [6], the low-temperature diffusion of hydrogen on metal surfaces and in a two-dimensional confinement between Ni(111) and an adsorbed graphene layer [7], and finally how to simulate huge structures like herringbone patterns on Au(111) efficiently.

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