O 3: Focus Session: Morphology Prediction at Interfaces

Time: Monday 10:30–13:00

Location: S051

Topical Talk   O 3.1   Mon 10:30   S051
On growth and interaction phenomena of heteromolecular adsorbates on metal surfaces — •Christian Kumpf — Peter Grünberg Institut (PGI-3), Forschungszentrum Jülich GmbH, Jülich, Germany, and Jülich-Aachen Research Alliance (JARA) - Fundamentals of Future Information Technology

The properties of functional materials and electronic devices are often dominated by their surfaces and interfaces. This is particularly true for organic thin films and adsorbate systems as they are frequently used in the field of organic electronics. Molecular blends (i.e., heteromolecular layers containing at least two different types of molecules, usually charge donors und acceptors) are of particular interest in this context. We have investigated a number of heteromolecular model systems containing either PTCDA or NTCDA as acceptor molecules, and CuPc or SnPc as donors.

In this talk I will concentrate on monolayer films consisting of PTCDA and CuPc, which we have studied using a variety of experimental methods like LEEM, LEED, STM, NIXSW, ARPES, orbital tomography and STS. A comprehensive LEEM study enabled us to understand the complex binary phase diagram containing three different ordered heteromolecular structures with different stoichiometry, several regimes with coexisting phases and a 2D-lattice gas. We identified the density of the gas phase to be the crucial parameter determining the growth behavior of the ordered structures on the surface. Furthermore, I will discuss the interaction of the two types of molecules with the surface in terms both its geometric and electronic fingerprints.

Topical Talk   O 3.2   Mon 11:00   S051
Surface Morphology from First-Principles: Thermodynamics and Kinetics — •Karsten Reuter — Technische Universität München

The structure of functionalized surfaces or surfaces exposed to realistic environments depends sensitively on the deposition conditions or gas-phase conditions. Detailed atomic-scale knowledge of this structure is often a prerequisite for further analysis and understanding. With experiment struggling to provide sufficient resolution, predictive-quality first-principles computational approaches assume a critical role in gaining this knowledge. Their task is then an efficient exploration of the vast configuration spaces, identifying (meta-)stable states and critical barriers on the high-dimensional potential energy surface.

While this task is thus simply summarized in one sentence, the words "efficient", "vast" and "high-dimensional" contained in it equally highlight the challenges faced in actually accomplishing this endeavour for anything but simple model systems. Knobs to turn are the way the algorithms navigate through configuration space, reduced-cost energy functions used for large-scale pre-screenings, or the use of coarse-grained lattice models. In this talk I will review our recent activities in this field, focusing on examples that are governed by both thermodynamic and kinetic factors.

Topical Talk   O 3.3   Mon 11:30   S051
Simulations of Surfaces and Interfaces Using High-Dimensional Neural Network Potentials — •Jörg Behler — Lehrstuhl für Theoretische Chemie, Ruhr-Universität Bochum, 44780 Bochum, Germany

In recent years a lot of progress has been made in the development of interatomic potentials based on machine learning techniques. These potentials are very efficient and thus allow to extend the length and time scales of molecular dynamics simulations significantly. Further, they are close to the accuracy of the underlying reference electronic structure methods and equally applicable to all types of bonding making them particularly useful for complex systems like interfaces. After a short overview about the available approaches, specifically the class of high-dimensional neural network potentials will be discussed. Several applications including water and solid-liquid interfaces will be presented to illustrate the advantages and current limitations of this method.

Topical Talk   O 3.4   Mon 12:00   S051
Morphology and growth of organic molecules at structured surfaces — •Sabine H.L. Klapp and Nicola Kleppmann — Institut für Theoretische Physik, Sekr. EW 7-1, Technische Universität Berlin, Hardenbergstrasse 36, 10623 Berlin

We report about recent progress in modelling organic molecules at structured inorganic, particularly semiconductor surfaces on a coarse-grained level of description. We start by discussing multilayer growth of the organic molecule C60. Based on kinetic MC simulations we quantify this time-dependent process and extract a self-consistent set of energy parameters, which lead to excellent agreement with parallel, real-time x-ray measurements. We then proceed to the structure formation and dynamics of the strongly anisotropic molecule para-sexiphenyl (6P) at ZnO surfaces. To this end we propose a new, coarse-grained model which aims at representing heuristically the different electrostatic and steric interactions occuring at the charge-patterned surface ZnO(10-10). Corresponding MC simulations of the equilibrium behavior reveal indeed a complex interplay of the different contributions of this coarse-grained Hamiltonian. Finally, based on kMC simulations and a rate equation approach we explore the non-equilibrium surface growth of 6P at finite adsorption rates, revealing re-entrant effects, a critical adsorption rate and observables that are non monotonous with the adsorption rate. Our results form a basis for understanding and predicting collective orientational ordering during growth in hybrid material systems.

Topical Talk   O 3.5   Mon 12:30   S051
Interaction of alcohols and water with carbonate surfaces — •Angelika Kühnle[1], Christoph Marutschke[1], Felix Kling[1], Dirk Lautner[1], Ralf Bechstein[1], Peter Spijker[2], John Tracey[2], Ville Loukonen[2], and Adam Foster[2] — [1]Universität Mainz — [2]Aalto University

Carbonate minerals are abundant in the earth's crust and constitute the inorganic phase in many bio-minerals created by nature. Understanding the formation and reactivity of carbonates in their natural environment requires elucidating the interaction of organic molecules with the mineral surface in the presence of water. We have studied the solid-liquid interface at the natural cleavage plane of the most common carbonates, i.e., calcite, magnesite and dolomite using high-resolution atomic-force microscopy (AFM) imaging and three-dimensional solvation layer mapping. Representing the simplest organic molecules, we have studied methanol, ethanol and their competition with water on the carbonate surfaces. In combination with molecular dynamics simulations we explain the observed solvation structures and provide evidence for stronger ethanol-calcite interaction as compared to water-calcite interaction.

We complement our liquid environment studies by an investigation in ultra-high vacuum (UHV). Thermal desorption spectroscopy obtained at sub-monolayer coverage confirms that ethanol is more strongly bound to calcite as compared to water. In combination with theoretical simulations we disclose atomic-scale details of the water-carbonate as well as the alcohol-carbonate interaction.