O 86: Mini-Symposium: Machine learning applications in surface science II

Time: Thursday 10:30–12:30 Location: R1

Invited Talk O 86.1 Thu 10:30 R1

Machine learning for robotic nanofabrication with molecules — •Christian Wagner — Peter Grünberg Institut (PGI-3), Forschungszentrum Jülich, Germany — JARA Fundamentals of Future Information Technology, Jülich, Germany

The ability to handle single molecules as effectively as macroscopic building-blocks would enable the construction of complex supramolecular structures that are not accessible by self-assembly. A central difficulty on the way towards this technology is the uncontrolled variability and poor observability of atomic-scale conformations, especially during the manipulation process. We present a generic strategy to overcome both obstacles, and demonstrate autonomous robotic nanofabrication with single molecules using reinforcement learning (RL). Quite generally, RL is able to learn strategies even in the face of large uncertainty and with sparse feedback. Indeed, RL based prediction models recently exceeded human performance in several games. However, to be useful for nanorobotics, standard RL algorithms must be adapted to also cope with the limited training opportunities that are available there. We demonstrate our correspondingly enhanced RL approach by applying it to an exemplary task of subtractive manufacturing with a scanning probe microscope (SPM). Complementary to that we outline how machine learning and control theory methods in combination with molecular simulations can be utilized to recover atomic-scale conformations from the sparse experimental SPM data available during manipulation.

Invited Talk O 86.2 Thu 11:00 R1

Chemisorbed or Physisorbed? Resolving surface adsorption with Bayesian inference and atomic force microscopy — •Milica Todorović — Department of Applied Physics, Aalto University, P.O. Box 11100, Aalto 00076, Finland

The knowledge on structure, bonding and properties of organic adsorbates to inorganic substrates underpins key technologies from catalysis through coatings to optoelectronics. Atomic force microscopy (AFM) and ab initio simulations are powerful tools for characterising molecular adsorption, but both struggle with complex bulky adsorbates where lack of chemical intuition and inconclusive imaging render the structure identification problematic. We address this challenge with Bayesian Optimization Structure Search (BOSS), a computational tool for global configurational structure search at surfaces and interfaces.

We employed BOSS to study the adsorption of (1S)-camphor on the Cu(111) surface, where AFM experiments recorded an unusual variety of images. From a single configurational search we retrieved 8 unique stable adsorbates. We discovered that camphor undergoes both covalent and dispersive binding to this substrate. Matching our findings to experimental data allowed us to categorise the AFM images into those associated with chemisorbed and physisorbed molecules. By simulating AFM images of the chemisorbed model structures, we identified three distinct adsorbates in the experimental images, further clarifying AFM image interpretation. This study illustrates how machine learning applications advance understanding in surface science by complementing both computation and experiment.

O 86.3 Thu 11:30 R1

Ab initio structure search of flexible molecules at interfaces — •Dmitrii Maksimov[1,2] and Mariana Rossi[1,2] — [1]Fritz Haber Institute of the Max Planck Society, Berlin, Germany — [2]Max Planck Institute for the Structure and Dynamics of Matter, Hamburg, Germany

We investigate how the accessible conformational space of two flexible amino acids, Arg and Arg-H^+, changes upon adsorption, by building and analyzing a database of thousands of structures optimized at Cu(111), Ag(111) and Au(111) surfaces with the PBE functional including screened pairwise (vdW) interactions. We employ an unsupervised dimensionality reduction procedure that enables us to understand the alteration of the high-dimensional conformational space [1]. The creation of this database, which is paramount to train further diverse machine-learning models, suffers from well-known bottleneck related to the efficiency of the geometry optimizer. We introduce a flexible way of preconditioning approximate Hessian matrices in the BFGS algorithm that is tailored to accelerate the relaxation of vdW bonded structures that can handle large structural changes. An automated sampling of these systems is implemented within a random structure search package [2] that can take explicitly into account the flexibility of molecules, their position and orientation with respect to fixed surroundings and interfaces.

[1] Maksimov et. al., Int. J. Quantum Chem., e26369 (2020)
[2] https://github.com/sabia-group/gensec

O 86.4 Thu 11:45 R1

Configurational polaron energies using machine learning — •Viktor Birschitzky, Michele Reticcioli, and Cesare Franchini — University of Vienna, Faculty of Physics

Polarons are quasiparticles formed by the coupling of excess charge carriers with the phonon field. Polarons form preferentially at surfaces and have a wide range of effects on the chemical and physical properties of the hosting material.[1] First principles calculations of polarons conformational energies typically require large supercells and long molecular dynamics (MD) simulations, making the modeling of multipolaron system within reasonable timescales very challenging. Here, we propose a supervised machine learning scheme based on kernel-regression to solve this problem by learning single polaron energies for the prototypical oxygen-defective rutile TiO_{2-x}(110) surface, where each oxygen vacancy provides two excess electrons. To achieve accurate predictions on an ab initio MD database of polaronic energies[2] a descriptor has been developed, which embodies the interactions between polarons with defects and other localized charge carriers. Our results show that the proposed ML method is able to expand the DFT database with energetically more favorable polaron configurations – improving the convex hull construction – and that generalization at arbitrary polaron concentration and defect types is possible.

[1] C. Franchini et al., Polarons in Material, Nature Review Materials, (2021)
[2] M. Reticcioli et al., Formation and dynamics of small polarons on the rutile TiO_2 surface, Physical Review B, (2018)

O 86.5 Thu 12:00 R1

Neural Network Analysis of Neutron and X-Ray Reflectivity Data: Pathological Cases, Performance and Perspectives — •Alessandro Greco[1], Vladimir Starostin[1], Alexander Hinderhofer[1], Alexander Gerlach[1], Maximilian Skoda[2], Stefan Kowarik[3], and Frank Schreiber[1] — [1]Institute of Applied Physics, University of Tübingen, Germany — [2]Rutherford Appleton Lab, ISIS Neutron and Muon Source, UK — [3]Department of Physical Chemistry, University of Graz, Austria

Neutron and X-ray reflectometry (NR and XRR) are powerful techniques to investigate the structural, morphological and even magnetic properties of solid and liquid thin films. Having demonstrated the general applicability of neural networks to analyze XRR and NR data before [1], this work discusses challenges arising from certain pathological cases as well as performance issues and perspectives. These cases include a low signal to noise ratio, a high background signal (e.g. from incoherent scattering), as well as a potential lack of a total reflection edge (TRE). We show that noise and background intensity pose no significant problem as long as they do not affect the TRE. However, for curves without strong features the prediction accuracy is diminished. Furthermore, we discuss the effect of different scattering length density combinations on the prediction accuracy. The results are demonstrated using simulated data of a single-layer system.

[1] Greco et al., *J. Appl. Cryst.*, **52**, 1342 (2019)

O 86.6 Thu 12:15 R1

Materials genes of heterogeneous catalysis from clean experiments and AI — •Lucas Foppa[1,2], Luca M. Ghiringhelli[1,2], Frank Rosowski[3], Robert Schloegl[1,4], Annette Trunschke[1], and Matthias Scheffler[1,2] — [1]Fritz-Haber-Institut der Max-Planck-Gesellschaft — [2]Humboldt-Universität zu Berlin — [3]BASF SE — [4]Max-Planck-Institut für Chemische Energiekonversion

Heterogeneous catalysis is an example of a complex materials function, governed by an intricate interplay of several processes, e.g. the dynamic re-structuring of the catalyst material at reaction conditions and different surface chemical reactions. Modelling the full catalytic progression via first-principles statistical mechanics is impractical, if not impossible. Instead, we show here how an artificial-intelligence

approach can be applied, even to an extremely small number of materials, to model catalysis and determine the key descriptive parameters (materials genes) reflecting the processes that trigger, facilitate, or hinder the catalyst performance. We start from a consistent, unparalleled experimental set of "clean data", containing nine vanadium-based oxidation catalysts which were carefully synthesized, fully characterized, and tested according to standardized protocols.[1] By applying the symbolic-regression SISSO approach,[2,3] we identify correlations between the few most relevant materials properties and their reactivity. This approach highlights the underlying physico-chemical processes, and accelerates catalyst design. [1] A. Trunschke, et al., Top. Catal. 63, 1683 (2020). [2] R. Ouyang et al., Phys. Rev. Mater. 2, 083802 (2018). [3] R. Ouyang et al., J. Phys. Mater. 2, 024002 (2019).