O 42: Poster Session III: Poster to Mini-Symposium: Machine learning applications in surface science I

Time: Tuesday 10:30–12:30 Location: P

O 42.1 Tue 10:30 P

Learning electron densities in condensed phase space — •Alan Lewis[1], Andrea Grisafi[2], Michele Cerrioti[2], and Mariana Rossi[1] — [1]MPI for Structure and Dynamics of Materials, Hamburg, Germany — [2]École Polytéchnique Fédèrale de Lausanne, Lausanne, Switzerland

The electron density is a fundamental quantity for modelling and understanding physical phenomena in materials. Not only is it central to theories like density-functional theory, but it also allows the calculation of a wide range of observables that are either directly or indirectly connected to it, like total energies, dipole moments, the electrostatic potential, work functions, and others. In this work, we present a model that is able to learn and predict the electronic density of diverse materials, ranging from liquids to solid semiconductors and metals. This is achieved by extending the framework presented by Grisafi et al [1] to work with periodic boundary conditions and when using a resolution of the identity on a numeric atom-centered orbital basis [2] to obtain coefficients for the expansion of the periodic density. This density is learned through a Gaussian process regression model that makes use of local symmetry-adapted representations of the atomic structure, which makes our method both data-efficient and highly transferable. We discuss the applicability of this model for large-scale periodic systems and its transferability across the periodic table.

[1] Grisafi et al, ACS Cent. Sci. 5, 57-64, 2019
[2] Blum et al, Comput. Phys. Commun. 180, 2175-2196, 2009

O 42.2 Tue 10:30 P

A fourth-generation high-dimensional neural network potential — •tsz wai ko[1], jonas a. finkler[2], stefan goedecker[2], and jörg behler[1] — [1]Theoretische Chemie, Institut für Physikalische Chemie, Georg-August-Universität Göttingen, Tammannstr. 6, 37077 Göttingen, Germany — [2]Department of Physics, Universität Basel, Klingelbergstrasse 82, 4056 Basel, Switzerland

Machine learning potentials (MLPs) have become an important tool for performing reliable atomistic simulations in surface science due to their nearly ab-initio accuracy and efficiency comparable to empirical force field. The majority of MLPs relies on the representation of energies and sometimes charges as a function of the local atomic environments. They are thus unable to describe non-local changes in the electronic structure due to long-range charge transfer or different global charges of a system.

Here we proposed a fourth-generation high-dimensional neural network potential (4G-HDNNP) for capturing the global charge distributions and corresponding non-local effects. We demonstrate the performance of 4G-HDNNPs for different benchmark systems showing that 4G-HDNNPs are in excellent agreement with electronic structure calculations.

O 42.3 Tue 10:30 P

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 42.4 Tue 10:30 P

Analysis of grazing-incidence wide-angle X-ray scattering by neural networks — •Vladimir Starostin, Alessandro Greco, Alina Pleli, Alexander Hinderhofer, Alexander Gerlach, and Frank Schreiber — Institute of Applied Physics, University of Tübingen, Germany

Grazing-incidence wide-angle x-ray scattering (GIWAXS) is an indispensable tool for studying nanostructure surfaces and thin films. It is widely used in real-time studies of thin film growth. However, high acquisition rates of real-time experiments lead to enormous amounts of data to be analysed. For instance, a modern 2D X-ray detector has around 4.5 million of pixels and produces up to 6 Gb of data per second at the maximum frame rate of 750 Hz. In the future, these numbers will only increase and it may become unfeasible to analyze or even save unprocessed data. To address these problems, some automated tools need to be developed [1].

In this work, we present a machine learning approach that provides feature detection of GIWAXS images in an automated fashion. This simplifies the experimental data analysis and might enable on-the-fly preprocessing of GIWAXS data.

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

O 42.5 Tue 10:30 P

Symmetry-Equivariant Representations of Ab Initio Hamiltonians for Machine Learning Purposes — •Michael Luya[1] and Reinhard Maurer[2] — [1]Department of Mathematics, University of Warwick — [2]Department of Chemistry, University of Warwick

High-dimensional machine learning is consistently improving the quality of condensed matter simulations. These simulations provide us with the Hamiltonian matrix, useful for calculating a wide variety of material properties. Predicting these matrices requires a deep understanding of covariance properties and directional coordinate dependence.

In an attempt to capture rotation invariance we introduce methods involving crystal field theory and bijective minimal basis transformations, in order to extract symmetry invariant parameters that can describe high-order contributions to the Hamiltonian, with application towards modelling a variety of molecules, metallic clusters, and eventually metal-organic interfaces. We also investigate the physical significance of these parameters and present a scheme as to how they should be best extracted from data, for the purposes of providing significant training data for neural network models.

O 42.6 Tue 10:30 P

Active Discovery of Organic Semiconductors — •Christian Kunkel[1,2], Johannes T. Margraf[1], Ke Chen[1], Harald Oberhofer[1], and Karsten Reuter[1,2] — [1]Chair for Theoretical Chemistry and Catalysis Research Center — [2]Fritz-Haber Institut der Max-Planck-Gesellschaft

Improving charge-transport of organic semiconductors (OSCs) for electronic applications is usually tackled by empirical structural tuning of promising compounds. Howver, the versatility of organic molecules generates a rich design space whose vastness dictates efficient search strategies. We thus here present an active machine learning (AML) approach that explores this virtually unlimited design space iteratively. Judging suitability of OSC candidates by charge injection and mobility-related descriptors, the AML approach iteratively queries first-principle evaluation on well-selected molecules. We first optimize the approach in a fully characterized, but truncated molecular test space, gaining deep methodological insight about its exploratory behavior. Outperforming a conventional computational funnel, the devised algorithm can thereby successfully leverage its gradually improving knowledge and focus on promising regions of the design space. When subsequently lifting the artificial truncation, high-performance candidates are constantly found while the algorithm meanders ever more deeply through the endless OSC design space. The demonstrated high efficiency in the detection of candidate compounds with superior charge conduction properties highlights the usefulness of autonomously operating systems for a targeted OSC design.

O 42.7 Tue 10:30 P

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 42.8 Tue 10:30 P

IrO_2 surface complexions identified through machine-learned interatomic potentials — •Jakob Timmermann[1,2], Yonghyuk Lee[1,2], Carsten Staacke[1,2], Christoph Scheurer[1,2], and Karsten Reuter[1,2] — [1]Fritz-Haber-Institut der MPG — [2]Technische Universität München

IrO_2 is currently the preferred catalyst for the electrochemical oxygen evolution reaction in proton exchange membrane electrolyzers. Full *ab initio* molecular dynamics (MD) simulations of the reactive processes at the surface would be highly desirable for mechanistic catalyst improvement, but are computationally not tractable for a foreseeable time. To overcome the limitations regarding system size and propagation time, MDs based on machine-learned interatomic potentials are an appealing alternative. Here, we present a Gaussian Approximation Potential (GAP) approach for IrO_2 combining two-body and smooth overlap of atomic positions (SOAP) descriptors to capture the atomic environments. For maximum data efficiency, we pursue an iterative parametrization protocol, in which preliminary GAP potentials based on limited first-principles data are used to generate most meaningful additional structures for retraining. The final GAP potential enables a global geometry optimization of low-index rutile IrO_2 facets through simulated annealing. Consecutive *ab initio* thermodynamics and detailed surface science investigations fully confirm the identified novel (101) and (111) (1x1) terminations as competitive with the most studied (110) facet in reducing environments [1]. [1] J. Timmermann *et al.*, Phys. Rev. Lett. **125**, 206101 (2020).

O 42.9 Tue 10:30 P

The data-driven search of new catalysts for an OCM reaction based on the properties of surface carbonates — •Aliaksei Mazheika[1], Frank Rosowski[1,2], and Ralph Kraehnert[1] — [1]BasCat, Technische Universitaet Berlin, Berlin, DE — [2]BASF SE, Ludwigshafen, DE

The interest in oxidative coupling of methane (OCM) reaction is caused by the fact that this is a relatively simple way for conversion of methane to C2 products (ethane, ethylene). Despite quite many years spent for the search of an efficient catalyst, still a catalyst which would be commercially viable has not been found. Recently Wang et al. have experimentally observed the volcano-like dependence of OCM performance of oxide catalysts on decomposition of their carbonates [1]. In this study we develop a way for calculations of carbonates formation energies based on adsorption of CO_2 on the surfaces of corresponding oxides. This allows us to reformulate experimentally observed volcano-like dependence in terms of theoretically calculated quantities. Based on this, we develop the strategy for high-throughput screening using artificial intelligence methodology - subgroup discovery [2] and SISSO [3]. With that we have done the screening of more than 800k materials, and obtained new materials promising for OCM reaction.

[1] H. Wang, PhD thesis, TU Berlin (2018).
[2] M. Boley *et al.*, Data Min. Knowl. Disc. **31**, 1391 (2017).
[3] R. Ouyang *et al.*, Phys. Rev. M **2**, 083802 (2018).