O 43: Frontiers of Electronic Structure Theory: Focus on Artificial Intelligence Applied to Real Materials 1

Time: Wednesday 10:30–13:00   Location: S054

O 43.1   Wed 10:30   S054
Structure of Amorphous Phosphorus from Machine Learning-Driven Simulations — •Yuxing Zhou, William Kirkpatrick, and Volker L. Deringer — Department of Chemistry, Inorganic Chemistry Laboratory, University of Oxford Oxford OX1 3QR, UK

Amorphous phosphorus (a-P) has long attracted interest because of its complex atomic structure, and more recently as an anode material for batteries. However, accurately describing and understanding a-P at the atomistic level remains a challenge. In this talk, we show that a general-purpose Gaussian approximation potential (GAP) model for phosphorus can be created by machine learning (ML) from a suitably chosen ensemble of quantum-mechanical results. Its accuracy in describing the amorphous phase is demonstrated via large-scale molecular-dynamics simulations on the atomic structure of a-P: the calculated structure factors yield good agreement with earlier experimental evidence. Abundant five-membered rings are found in the structural model, which are the building block of more complex clusters. We provide new insights into the cluster fragments under pressure: an analysis of cluster fragments, large rings, and voids suggests that moderate pressure (up to about 5 GPa) does not break the connectivity of clusters, but higher pressure does. Changes in the simulated first sharp diffraction peak during compression and decompression indicate a hysteresis in the recovery of medium-range order. Our work provides a starting point for further computational studies of a-P, and more generally it exemplifies how ML-driven modeling can accelerate the understanding of disordered functional materials.

O 43.2   Wed 10:45   S054
Realistic Structural Properties of Amorphous SiNx from Machine-Learning-Driven Molecular Dynamics — •Ganesh Kumar Nayak[1], Prashanth Srinivasan[2], Juraj Todt[3], Rostislav Daniel[1], and David Holec[1] — [1]Department of Materials Science, Montanuniversität Leoben, Leoben, Austria — [2]Franz-Josef-Strasse 18 — [3]Erich Schmid Institute of Materials Science of the Austrian Academy of Sciences, Jahnstrasse 12, Leoben, Austria

Machine-learning(ML)-based interatomic potentials can enable simulations of extended systems with an accuracy that is largely comparable to DFT, but with a computational cost, that is orders of magnitude lower. Molecular dynamics simulations further exhibit favorable linear (order N) scaling behavior.

Amorphous silicon nitride (a-SiNx) is a widely studied noncrystalline material, and yet the subtle details of its atomistic structure and mechanical properties are still unclear. Due to the small sizes of representative models, DFT cannot reliably predict its structural properties and hence left an anisotropic order parameter. Here, we show that accurate structural models of a-SiNx can be obtained using an ML-based inter-atomic potential. Our predictions of structural properties are validated by experimental values of mass density by X-ray reflectivity measurements and by radial distribution function measured by synchrotron X- ray diffraction.

Our study demonstrates the broader impact of ML potentials for elucidating structures and properties of technologically important amorphous materials.

O 43.3   Wed 11:00   S054
Combined experimental-computational directed sampling approach to modelling amorphous alumina — •Angela Harper[1], Steffen Emge[2], Pieter Magusin[2,3], Clare Grey[2], and Andrew Morris[4] — [1]Cavendish Laboratory, University of Cambridg — [2]Department of Chemistry, University of Cambridge — [3]Institute for Life Sciences & Chemistry, Hogeschool Utrecht — [4]School of Metallurgy and Materials, University of Birmingham

Understanding the electronic and atomic level structure of materials is imperative for discovering the next generation of solid state electronic devices. Yet for amorphous materials, it is non-trivial to determine the exact local ordering. In this talk, I outline a method for modelling disordered materials, using experimentally directed sampling of static configurations from *ab initio* molecular dynamics[1]. We calculate experimentally relevant spectra and properties including X-ray absorption edges, nuclear magnetic resonance chemical shieldings, and the electronic density of states, with first principles accuracy. This model

is validated on amorphous alumina, a widely used coating material in electronic devices, and identify two distinct five-fold coordinated geometries of AlO_5, as well as localised states at the conduction band minimum. By leveraging both experimental and computational data in our approach we highlight the need for experimentally informed calculations which lead to a more detailed understanding of complex materials, and develop an approach that is widely applicable to the modelling community.
[1]Harper, AF et al., Under review (2021)
doi.org/10.33774/chemrxiv-2021-qjzbj

O 43.4   Wed 11:15   S054
Structural phases and thermodynamics of $BaTiO_3$ from an integrated machine learning model — •Lorenzo Gigli[1], Max Veit[1], Michele Kotiuga[2], Giovanni Pizzi[2], Nicola Marzari[2], and Michele Ceriotti[1] — [1]Laboratory of Computational Science and Modeling (COSMO), Institute of Materials, École Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland — [2]Theory and Simulation of Materials (THEOS) and National Centre for Computational Design and Discovery of Novel Materials (MARVEL), École Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland

Modeling the ferroelectric transition of any given material requires three key ingredients: (1) a model of the potential energy surface, that describes the energetic response to a structural distortion; (2) the free energy surface sampled at the relevant, finite-temperature conditions; and; (3) the polarization of individual configurations that determines the observed polarization and the phase transitions. To this aim, we make use of an integrated machine-learning framework, based on a combination of an interatomic potential and a microscopic polarization model, which we use to run Molecular Dynamics simulations of ferroelectrics with the same accuracy of the underlying DFT method, on time and length scales that are not accessible to direct ab-initio modeling. This allows us to uncover the microscopic nature of the ferroelectric transition in barium titanate ($BaTiO_3$) and to identify the presence of an order-disorder transition as the main driver of ferroelectricity. The framework also allows us to reconstruct the temperature-dependent $BaTiO_3$ phase diagram, with first-of-its-kind accuracy.

O 43.5   Wed 11:30   S054
Dielectric properties of $BaTiO_3$ from an integrated machine-learning model — •Max Veit[1], Lorenzo Gigli[1], Michele Kotiuga[2], Giovanni Pizzi[2], Nicola Marzari[2], and Michele Ceirotti[1] — [1]Laboratory for Computational Science and Modeling (COSMO), Ecole Polytechnique Fédérale de Lausanne, Lausanne, CH — [2]Laboratory for Theory and Simulation of Materials (THEOS), Ecole Polytechnique Fédérale de Lausanne, Lausanne, CH

Modeling the finite-temperature behavior of ferroelectric materials from first principles has always been challenging due to the large supercells and long simulation times required for adequate sampling. Here we demonstrate the use of an integrated machine learning (ML) model of the potential energy and polarization surfaces of barium titanate ($BaTiO_3$) to overcome these difficulties and run long MD simulations with DFT accuracy. We use these simulations to compute the frequency-dependent dielectric response function, finding a spectrum qualitatively similar that obtained with previous effective-Hamiltonian simulations as well as to experimentally measured profiles, with some remaining discrepancies that we trace back to the underlying DFT model. Finally, we discuss possible extensions of the model to explicitly include long-range interactions, previously included only in an implicit, emergent manner. We expect this integrated, generally applicable modeling technique to become a valuable tool for elucidating the ferroelectric behavior of a wide variety of materials.

O 43.6   Wed 11:45   S054
The first-principles phase diagram of monolayer nanoconfined water — •Venkat Kapil[1], Christoph Schran[1], Andrea Zen[2], Ji Chen[3], Chris Pickard[1], and Angelos Michaelides[1] — [1]University of Cambridge, UK — [2]Universitá di Napoli Federico II, Italy — [3]Peking University, Beijing, China

Water in nanoscale cavities is ubiquitous and of central importance to everyday phenomena in geology and biology, and at the heart of cur-

rent and future technologies in nano-science. A molecular-level picture of the structure and dynamics of nano-confined water is a prerequisite to understanding and controlling the behavior of water under confinement. Here we explore a monolayer of water confined within a graphene-like channel using a framework that combines developments in high-level electronic structure theory, machine learning, and statistical sampling. This approach enables a treatment of nano-confined water at unprecedented accuracy. We find that monolayer water exhibits surprisingly rich and diverse phase behavior that is highly sensitive to temperature and the van der Waals pressure acting within the nano-channel. Monolayer water exhibits numerous molecular ice phases with melting temperatures that vary by over 400 degrees in a non-monotonic manner with pressure. In addition, we predict two unexpected phases: a *hexatic-like* phase, which is an intermediate between a solid and a liquid, and a superionic phase with a high electrical conductivity exceeding that of battery materials. Our work suggests that nano-confinement could be a promising route towards superionic behaviour at easily accessible conditions.

O 43.7   Wed 12:00   S054
Exploring amorphous graphene with empirical and machine-learned potentials — •Zakariya El-Machachi[1], Mark Wilson[2], and Volker L. Deringer[1] — [1]Department of Chemistry, Inorganic Chemistry Laboratory, University of Oxford, Oxford OX1 3QR, UK — [2]Department of Chemistry, Physical and Theoretical Chemistry Laboratory, University of Oxford, Oxford OX1 3QZ, UK

The structure of amorphous graphene (aG) lacks long range order whilst having short and medium range order yielding a rich and complex configurational space, which is yet to be fully understood. Here we report on an atomistic modelling study of aG using a machine learning (ML) based force field. ML force fields are typically "trained" on data from highly accurate but computationally costly density functional theory (DFT) computations. Atomistic models created by such ML methods can achieve near DFT accuracy at a fraction of the computational cost. One key assumption is that the global energy can be separated into sums of local energies. The physical interpretation of ML local energies is an interesting research question. We find that local and nearest neighbour (NN) ML energies can inform the generation of aG models from crystalline graphene via a Monte–Carlo bond switching algorithm. Bond switches are introduced as Stone–Wales defects, with the local energies of the defect pair and its NNs used in the acceptance criterion. Established empirical force fields are used in the same way and the resulting structures are studied. Our results provide insight into the modelling of amorphous graphene and into the nature of ML potential-energy models.

O 43.8   Wed 12:15   S054
Machine learning for estimation of spin models in undoped cuprates — •Denys Y. Kononenko[1], Ulrich K. Rössler[1], Jeroen van den Brink[1,2], and Oleg Janson[1] — [1]Institute for Theoretical Solid State Physics, IFW Dresden, Dresden, Germany — [2]Institute for Theoretical Physics, TU Dresden, Dresden, Germany

Undoped cuprates tailor a fascinating variety of low-dimensional and frustrated spin models, which can be indirectly characterized by the transfer integrals. The estimation of transfer integrals is related to a relatively complicated computational procedure which includes besides DFT calculation also a Wannierization. We propose a data-driven approach to replace this computationally demanding procedure.

We employ the Gaussian Process Regression model, trained on the results of high-throughput DFT calculations to estimate transfer integrals in undoped cuprates. The model learns from data the dependency between the local crystal environment of copper atoms pair and the corresponding value of transfer integral. The site position function of the local crystal environment is represented as a finite-dimensional vector composed of decomposition coefficients in the truncated basis of Zernike 3D functions [1]. The vector descriptor incorporates the spatial configuration and chemical composition of the local crystal environment. The proposed approach can be utilized for a rapid assessment of the spin models of new cuprates using structural information as the only input.

[1] M. Novotni and R. Klein, Computer Aided Design 36, 1047 (2004)

O 43.9   Wed 12:30   S054
Machine-learning Based Screening of Lead-free Perovskites for Photovoltaic Applications — •Elisabetta Landini[1,3], Harald Oberhofer[2], and Karsten Reuter[1] — [1]Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, D-14195 Berlin, Germany — [2]Chair for Theoretical Physics VII, Physikalisches Institut Universität Bayreuth, 95440, Bayreuth, Germany — [3]Chair for Theoretical Chemistry, Technische Universität München, Lichtenbergstr. 4, D-85747 Garching, Germany

Lead-free halide double perovskites are promising stable and non-toxic alternatives to methylammonium lead iodide in the field of photovoltaics. In this context, the most commonly used double perovskite is $Cs_2AgBiBr_6$, due to its favorable charge transport properties [1]. However, the maximum power conversion efficiency obtained for this material does not exceed 3%, as a consequence of its wide indirect gap and its intrinsic and extrinsic defects [2]. On the other hand, the materials space that arises from the substitution of different elements in the 4 lattice sites of this structure is large and still mostly unexplored.

In this work a neural network is used to predict the band gap of double perovskites from an initial space of 7056 structures and select candidates suitable for visible light absorption. Successive hybrid DFT calculations are used to evaluate the thermodynamic stability and the power conversion efficiency of the selected compounds, and propose novel potential solar absorbers.

[1] E.T. McClure *et al.*, *Chemistry of Materials* **28**, 1348 (2016).
[2] X. Yang *et al.*, *Energy & Fuels* **34**,10513 (2020).

O 43.10   Wed 12:45   S054
Equivariant graph neural network for linear scaling electron density estimation and applications in battery materials — •Arghya Bhowmik and Peter Jorgensen — 301 Anker Engelunds vej, Kgs. Lyngby, DK-2800

We present a machine learning framework for the prediction of $\rho(r)$ based on equivariant graph message passing neural networks. The electron density is predicted at special query point vertices that are part of the message passing graph, but only receive messages. The model is tested across multiple data sets of molecules (QM9), liquid ethylene carbonate electrolyte (EC) and LixNiyMnzCo(1-y-z)O2 lithium ion battery cathodes (NMC). The model is used to explore large materials phase space for safer battery materials and uncovering new understanding how redox mediated diffusion occurs and battery materials.