MM 34: Data Driven Materials Science: Interatomic Potentials / Reduced Dimensions

Time: Thursday 15:45–18:30 Location: H45

MM 34.1 Thu 15:45 H45

Constructing Training Sets for Transferable Moment Tensor Potentials: Application to Defects in Bulk Mg — •Marvin Poul, Liam Huber, Erik Bitzek, and Joerg Neugebauer — Max-Planck-Institut fuer Eisenforschung

Machine learned interatomic potentials promise to bring quantum mechanical accuracy to system sizes that are inaccessible with traditional QM approaches. Here, we present a set of unary Mg Moment Tensor Potentials[1] with different speeds and accuracies in the range of 100–5 meV/atom. We focus on understanding the role of the training data in the fitting process. We discuss several ways in which the structural complexity of the training structures and a physical understanding of them helps to design an efficient training set construction. The resulting potentials are verified on out-of-fold structures, like vacancies, surfaces, and high-symmetry grain boundaries. This work is implemented as a pyiron[2] workflow and we identify challenges and opportunities of a fully automated setup to fit machine-learned potentials.

[1]: https://doi.org/10.1088/2632-2153/abc9fe
[2]: https://doi.org/10.1016/j.commatsci.2018.07.043

MM 34.2 Thu 16:00 H45

Active learning and uncertainty quantification for atomic cluster expansion models — •Yury Lysogorskiy, Anton Bochkarev, and Ralf Drautz — Atomistic Modelling and Simulation, ICAMS, Ruhr-University Bochum, D-44801 Bochum, Germany

Interatomic potentials (IP) are widely used in computational materials science, in particular for simulations that are too computationally expensive for density functional theory (DFT). Recently the atomic cluster expansion (ACE) was proposed as a new class of data-driven IP with basis set completeness. Development of any IP requires numerous iterations and careful selection of training data. Thus automation of both construction of training dataset as well as IP validation would significantly speed up the development process. In this work we apply the Maxvol algorithm for training dataset selection and study the extrapolation grade metric (Podryabinkin and Shapeev, 2017) in the context of ACE and compare it to the query-by-committee approach for uncertainty estimation. These methods allow us to introduce extrapolation control in ACE models and to design different exploration automated protocols for accurate interatomic potentials development.

MM 34.3 Thu 16:15 H45

Take Two: Δ-Machine Learning for Molecular Co-Crystals — •Simon Wengert[1,2], Gábor Csányi[3], Karsten Reuter[1], and Johannes T. Margraf[1] — [1]Fritz Haber Institut der MPG, Berlin, Germany — [2]TU Munich, Germany — [3]University of Cambridge, UK

Co-crystals are a highly interesting material class, as varying their components and stoichiometry in principle allows tuning supramolecular assemblies towards desired physical properties. The *in silico* prediction of co-crystal structures represents a daunting task, however, as they span a vast search space and usually feature large unit-cells. This requires theoretical models that are accurate and fast to evaluate, a combination that can in principle be accomplished by modern machine-learned (ML) potentials trained on first-principles data. Crucially, these ML potentials need to account for the description of long-range interactions, which are essential for the stability and structure of molecular crystals. In this contribution, we present a strategy for developing Δ-ML potentials for co-crystals, which use a physical baseline model to describe long-range interactions. The applicability of this approach is demonstrated for co-crystals of variable composition consisting of an active pharmaceutical ingredient and various co-formers. We find that the Δ-ML approach offers a strong and consistent improvement over the density-functional tight binding baseline. Importantly, this even holds true when extrapolating beyond the scope of the training set, for instance in molecular dynamics simulations at ambient conditions.

MM 34.4 Thu 16:30 H45

Magnetic Atomic Cluster Expansion and application to Iron — •Matteo Rinaldi, Matous Mrovec, and Ralf Drautz — Interdisciplinary Centre for Advanced Materials Simulation (ICAMS)

The atomic cluster expansion (ACE)[1,2,3] has proven to be a valuable tool to parametrize complex energy landscapes of pure elements and alloys. However, its application to potential energy surfaces determined also by additional degrees of freedom, such as magnetic moments, has been still lacking. In particular, ferromagnetic materials cannot be tackled with the original ACE formalism, where the single-site energies depend parametrically only on interatomic distances and chemical species, since these descriptors cannot distinguish between atoms with different magnetic moments. The solution of this issue was given theoretically by Drautz[4], where the ACE formalism was extended to take into account additional labels of the atomic sites of scalar, vectorial and tensorial nature by including them in the definition of the atomic neighbor density. We have employed this formalism to parametrize a magnetic ACE for the prototypical ferromagnetic element Fe using a dataset of both collinear and non-collinear magnetic structures calculated with spin density functional theory. We will show that the new ACE model is able to describe correctly not only various magnetic phases of Fe at 0 K but also their finite temperature properties in good agreement with the reference *ab-initio* and experimental values.

[1] R. Drautz, Phys. Rev. B 99, 014104. [2] Y. Lysogorskiy et al., npj Comput Mater 7, 97 (2021). [3] A. Bochkarev et al., Phys. Rev. Materials 6, 013804. [4] R. Drautz, Phys. Rev. B 102, 024104.

MM 34.5 Thu 16:45 H45

Kernel Charge Equilibration: Learning Charge Distributions in Materials and Molecules — •Martin Vondrak, Nikhil Bapat, Hendrik H. Heenen, Johannes T. Margraf, and Karsten Reuter — Fritz-Haber-Institut, Berlin, Germany

Machine learning (ML) techniques have recently been shown to bridge the gap between accurate first-principles methods and computationally cheap empirical potentials. This is achieved by learning a systematic relationship between the structure of molecules and their physical properties. However, the modern ML models typically represent chemical systems in terms of local atomic environments. This inevitably leads to the neglect of long-range interactions (most prominently electrostatics) and non-local phenomena (e.g. charge transfer), which can lead to significant errors in the description of polar molecules and materials (particularly in non-isotropic environments). To overcome these issues, we recently proposed a ML framework for predicting charge distributions in molecules termed Kernel Charge Equilibration (kQEq). Here, atomic charges are derived from a physical model using environment-dependent atomic electronegativities. These models can be trained to reproduce electrostatic properties (e.g. dipole moments) of reference systems, computed from first principles. The impact of different fitting targets on predicted charge distributions is compared. Furthermore, strategies for fitting to energies are discussed, including combination of Gaussian Approximation Potential (GAP) with kQEq.

15 min. break

MM 34.6 Thu 17:15 H45

Machine Learning of ab-initio grain boundary Segregation Energies — •Christoph Dösinger[1], Daniel Scheiber[2], Oleg Peil[2], Vsevolod Razumovskiy[2], Alexander Reichmann[1], and Lorenz Romaner[1] — [1]Montanuniversität Leoben, Department of Materials Science, Leoben, Austria — [2]Materials Center Leoben Forschung GmbH, Leoben, Austria

Grain-boundary (GB) segregation is an important phenomenon in alloys, where the resulting GB excess can strongly influence their properties, for example induce intergranular fracture or lead to phase transformations. A fundamental quantity that uniquely describes the propensity of a solute towards GB segregation is the segregation energy. It determines the tendency of a solute atom to enrich or deplete at the GB. This quantity can be directly calculated from first principles. However, such calculations are computationally expensive and can become computationally unfeasible as the complexity of the GB crystal structure increases. The aim of this work is to reduce the computational cost of GB segregation energies by applying machine learning methods trained at series of representative DFT calculations and expanding them to more complex GB structures. The atomic structure, together with the segregation energies are used to train a model, which then is employed to predict the segregation energy for arbitrary segregation sites and GB types. In our work we apply this method to tungsten alloys. The results show, that this approach in-

deed gives reliable results for the segregation energies and can be used to get a complete description of segregation profiles.

MM 34.7 Thu 17:30 H45

Stability of binary precipitates in Cu-based alloys investigated through active learning and quantum computing — •Angel Diaz Carral[1], Xiang Xu[2], Azade Yazdan Yar[1], Siegfried Schmauder[2], and Maria Fyta[1] — [1]Institute for Computational Physics (ICP), Universität Stuttgart, Allmandring 3, 70569, Stuttgart, Germany — [2]Institut für Materialprüfung, Werkstoffkunde und Festigkeitslehre (IMWF), Pfaffenwaldring 32 70569, Stuttgart, Germany

Understanding the structure of thermodynamically stable precipitates is of great interest in material science as they can affect the electrical conductivity and mechanical properties of the matrix to a great degree. In this work, we use a relaxation-on-the-fly active learning algorithm in order to scan all possible binary candidates, for different types and concentrations of alloy elements (mainly Cu, Si, and Ni). Quantum-mechanical calculations are performed on a small number of candidates to train and improve the machine-learned potential. The model is then used to predict the enthalpy of formation of all candidates. The stability of binary precipitates, based on predicting the convex hull, is further assessed by the phonon density of states analysis calculated by classic and quantum computing.

MM 34.8 Thu 17:45 H45

How to teach my deep generative model to create new RuO_2 surface structures? — •Patricia König, Hanna Türk, Yonghyuk Lee, Chiara Panosetti, Christoph Scheurer, and Karsten Reuter — Fritz-Haber-Institut der MPG, Germany

Many widely used catalyst systems still hold complicated longstanding structural puzzles that hamper their full atomistic understanding and thus further knowledge based progress. Here, we address the well-known RuO_2 catalyst for the oxidative conversion of CO exhaust gases in combustion processes.

To explore the chemical space of RuO_2 surface structures, we trained a Generative Adversarial Network (GAN) that is capable of cheaply generating diverse structural guesses for novel surface structures. For the training set, 28,903 RuO_2 surface terminations were created with a grand-canonical basin hopping method. The atomic positions of these structures were mapped to Gaussian densities on a three-dimensional grid to generate the GAN input. We demonstrate how two-dimensional images of cuts through RuO_2 structures with inferred lattice lengths and energy conditioning can be created as a first step to realistic three-dimensional surface structures.

MM 34.9 Thu 18:00 H45

Data-Driven Design of Two-Dimensional Non-van der Waals Materials — •Rico Friedrich[1,2,3], Mahdi Ghorbani-Asl[1], Stefano Curtarolo[2], and Arkady V. Krasheninnikov[1,4] — [1]Helmholtz-Zentrum Dresden-Rossendorf, Dresden — [2]Duke University, Durham, USA — [3]TU Dresden — [4]Aalto University, Aalto, Finland

Two-dimensional (2D) materials are traditionally associated with the sheets forming bulk layered compounds bonded by weak van der Waals (vdW) forces. The weak inter-layer interaction leads to a natural structural separation of the 2D subunits in the crystals, giving rise to the possibility of mechanical and liquid-phase exfoliation as well as enabling the formulation of exfoliability descriptors.

The unexpected experimental realization of non-vdW 2D compounds, for which the previously developed descriptors are not applicable, opened up a new direction in the research on 2D systems [1]. Here, we present our recent data-driven search for representatives of this novel materials class [2]. By screening the AFLOW database according to structural prototypes, 28 potentially synthesizable candidates are outlined. The oxidation state of the surface cations is found to regulate the exfoliation energy with low oxidation numbers giving rise to weak bonding — thus providing an enabling descriptor to obtain novel 2D materials. The candidates showcase a diverse spectrum of appealing electronic, optical and magnetic features.

[1] A. Puthirath Balan *et al.*, Nat. Nanotechnol. **13**, 602 (2018).
[2] R. Friedrich *et al.*, Nano Lett. **22**, 989 (2022).

MM 34.10 Thu 18:15 H45

Robust recognition and exploratory analysis of crystal structures via Bayesian deep learning — •Andreas Leitherer, Angelo Ziletti, and Luca M. Ghiringhelli — The NOMAD Laboratory at the Fritz Haber Institute and at the Humboldt University of Berlin, Germany

Atomic-resolution studies are routinely being performed in modern materials-science experiments. Artificial-intelligence tools are promising candidates to leverage this valuable – yet underutilized – data in unprecedented, automatic fashion to discover hidden patterns and eventually novel physics. Here, we introduce ARISE (Nat. Commun. 2021, https://doi.org/10.1038/s41467-021-26511-5), a crystal-structure-identification method based on Bayesian deep learning. As a major step forward, ARISE is robust to structural noise and can treat more than 100 crystal structures, a number that can be extended on demand. While being trained on ideal structures only, ARISE correctly characterizes strongly perturbed single- and polycrystalline systems, from both synthetic and experimental sources. The probabilistic nature of the Bayesian-deep-learning model yields principled uncertainty estimates, which are found to be correlated with crystalline order of metallic nanoparticles in electron-tomography experiments. Application of unsupervised learning to the internal neural-network representations reveals grain boundaries and (unapparent) structural regions sharing interpretable geometrical properties. This work enables the hitherto hindered analysis of noisy atomic structural data from computations or experiments.