MM 44: Developement of Calculation Methods II

Time: Wednesday 15:45–18:00 Location: C 264

MM 44.1 Wed 15:45 C 264

FAIR Data Quality Metrics in NOMAD — •Nathan Daelman[1], Joseph F. Rudzinski[1], José M. Pizarro[1], Luca M. Ghiringhelli[2], and Silvana Botti[3] — [1]Institut für Physik und IRIS-Adlershof, Humboldt-Universität zu Berlin, Berlin — [2]Department of Materials Science and Engineering, Friedrich-Alexander-Universität, Erlangen-Nürnberg — [3]RC-FEMS and Faculty of Physics, Ruhr University Bochum, Bochum

The FAIR principles (Findable, Accessible, Interoperable, Reusable) serve as a reference for assessing the quality of data storage and publication [1]. NOMAD [nomad-lab.eu][2, 3] is an open-source data infrastructure for materials science data that is built upon these principles.

In this presentation, I will demonstrate the interplay between high-quality data and knowledge using the functionalities provided by NOMAD and with DFT as an example case. In particular, I will showcase the dynamic and flexible metadata framework, designed for a clearer, more customizable navigation of the zoo of density functionals. I will then show how precision and accuracy metrics are represented within this framework, and how they can be linked to benchmark datasets. Finally, I will present a brief outlook on the future of NOMAD as a platform that fosters an interconnected research community and engaged scientific discourse.

[1] Wilkinson, M. D. *et al.*, Sci. Data **3**, 160018 (2016).
[2] Scheffler, M. *et al.*, Nature **604**, 635-642 (2022).
[3] Scheidgen, M. *et al.*, JOSS **8**, 5388 (2023).

MM 44.2 Wed 16:00 C 264

Machine-learning interatomic potentials with beyond-DFT accuracy: application to covalent-organic frameworks — •Yuji Ikeda, Axel Forslund, and Blazej Grabowski — University of Stuttgart, Stuttgart, Germany

Covalent-organic frameworks (COFs) are nanoporous crystalline materials formed by strong covalent bonds of organic secondary building units composed mostly of light elements like C, N, O, H, etc. Most COFs are quasi-two-dimensional materials with layers interacting with van der Waals (vdW) forces. It is fascinating to investigate COFs using machine-learning interatomic potentials (MLIPs) because of their capability to access, e.g., long time- and length-scales in molecular-dynamics (MD) simulations. To simulate vdW materials, MLIPs should be trained by data including the vdW interaction. Such data are typically prepared with vdW-DFT functionals. These vdW-DFT functionals are however essentially semi-empirical in the sense that their parameters are fitted to show agreement with experiments, and hence they imply concerns about transferability. Our solution is to consider training data obtained from post-Hartree-Fock (HF) methods such as the coupled-cluster (CC) methods, which are non-empirical and have beyond-DFT accuracy. Using MLIPs trained on the beyond-DFT data, we demonstrate the calculation of structural properties of COFs.

MM 44.3 Wed 16:15 C 264

Kinetic Modeling of Stripes, Surfaces, and Solids Using the kmos3 Framework — •Martin Deimel[1], Aditya Savara[2], Karsten Reuter[1], and Sebastian Matera[1] — [1]Fritz-Haber-Institut der MPG, Berlin — [2]Oak Ridge National Laboratory, Tennessee, United States

Kinetic Monte Carlo (KMC) simulations have attracted growing attention over the years for the modeling of diverse surface or bulk processes, e.g., crystal growth, catalytic reactions, or diffusion in solids. We introduce the open-source Python package kmos3 for lattice KMC, the third revision of the original kmos package.[1] Being based on the rejection-free variable step size algorithm, kmos3 acts as a flexible code generator framework, creating model-specific efficient Fortran code which is subsequently compiled to a Python callable library. Using different prototypical models, we demonstrate the usage of kmos3. The different available backends, which target different classes of models, will be analyzed in terms of their performance with respect to model complexity and system size. Finally, we will discuss how to speed up KMC simulations of problems with time-scale separation using the implemented acceleration algorithm.[2]

[1] M. Hoffmann, S. Matera, and K. Reuter, Comput. Phys. Commun. **185**, 2138 (2014).
[2] M. Andersen, C. Plaisance, and K. Reuter, J. Chem. Phys. **147**, 152705 (2017).

MM 44.4 Wed 16:30 C 264

Extracting Gibbs free energies from local composition fluctuations in atom probe data — Jianshu Zheng[1], Rüya Duran[1], Marvin Poul[2], Guido Schmitz[1], and •Sebastian Eich[1] — [1]Institute for Materials Science, University of Stuttgart, Germany — [2]Department of Computational Alloy Design, Max-Planck-Institut für Eisenforschung GmbH, Düsseldorf, Germany

While thermodynamic fluctuation theory has been applied to liquids for decades to obtain Gibbs free energies from local composition fluctuations, the present work extends this theory to solids by considering an additional elastic work term. This theory is firstly verified via atomistic embedded-atom simulations in an exemplary Cu–Ni system using Monte Carlo techniques. Composition fluctuations, which are evaluated for various-sized subvolumes, reveal a systematic dependence on the size of the evaluation volume, but extrapolation to an infinitely large subvolume leads to perfect agreement with the prediction by the extended theory. Thus, recovering Gibbs free energies becomes indeed possible also in solids. In a second step, this method is now applied to experimental atom probe tomography (APT) data with high chemical and spatial resolution, enabling the analysis of local composition fluctuations. From these data, the Gibbs free energy is recovered and remarkable agreement is found between our approach and the latest CALPHAD representation of the miscibility gap. This methodology is therefore believed to efficiently improve the accuracy of thermodynamic information (e.g. miscibility gap, mixing/demixing tendencies, critical solution temperature) from direct APT measurements.

MM 44.5 Wed 16:45 C 264

Improving the Diversity of Transition State Searches with On-the-fly Learned Biasing Potentials — •Nils Gönnheimer[1,2], King Chun Lai[1], Karsten Reuter[1], and Johannes T. Margraf[1,2] — [1]Fritz-Haber-Institut der MPG Berlin — [2]Universität Bayreuth

Constructing reaction networks of reaction pathways is fundamental for long-timescale simulations and the theoretical analysis of complex chemical processes. A common approach involves using minimum mode following (MMF) methods to identify these reaction pathways, and especially transition states (TSs), on potential energy surfaces (PESs). However, MMF methods may miss essential reaction pathways due to their tendency of converging towards a limited set of transition states, even when simulations are repeated with different starting conditions. Herein, we address this limitation by introducing a biasing potential which modifies the PES based on on-the-fly gathered information. The bias drives the TS search algorithm away from known TSs and therefore increases the diversity of the outcome. We demonstrate the impact of the on-the-fly generated bias on the MMF method by showing the change of basins of attraction for first-order saddle points within a benchmark 2D PES. Furthermore, we will illustrate the effect of the bias on the diversity of identified TSs during a restricted number of TS searches for the self-diffusion of a heptamer on a Pt(111) surface.

15 min. break

MM 44.6 Wed 17:15 C 264

Kernel Charge Equilibration: Machine Learned Interatomic Potentials With Full Long-Range Electrostatics — •Martin Vondrak[1], Johannes T. Margraf[1,2], and Karsten Reuter[1] — [1]Fritz-Haber-Institut der MPG, Berlin — [2]University of Bayreuth

Machine learning (ML) interatomic potentials have recently been shown to bridge the gap between accurate first-principles methods and computationally cheap empirical potentials. This is achieved by learning a mapping between a system's structure and its physical properties. To this end, state-of-the-art models typically represent chemical structures 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), resulting in significant errors in the description of polar molecules and materials (particularly in non-isotropic environments). To overcome these issues, we re-

cently proposed an ML framework for predicting charge distributions in molecules termed Kernel Charge Equilibration (kQEq) [1]. Here, atomic charges are derived from a physical model using environment-dependent atomic electronegativities. In this contribution, strategies for creating kQEq interatomic potentials are discussed, including the combination of short-ranged Gaussian Approximation Potentials with kQEq.

[1] M. Vondrak, K. Reuter, and J.T. Margraf, J. Chem. Phys. **159**, 054109 (2023).

MM 44.7 Wed 17:30 C 264

Sampling-free computation of finite temperature material properties in isochoric and isobaric ensembles using the mean-field anharmonic bond model — •Raynol Dsouza[1], Marvin Poul[1], Liam Huber[2], Thomas D. Swinburne[3], and Jörg Neugebauer[1] — [1]Max-Planck-Institut für Eisenforschung GmbH, Düsseldorf, 40327, Germany — [2]Grey Haven Solutions, Victoria, BC, Canada — [3]Aix-Marseille Universite, CNRS, CINaM UMR 7325, Campus de Luminy, Marseille 13288, France

The recently introduced mean-field anharmonic bond model has shown remarkable accuracy in predicting finite temperature free energies for certain potential models of fcc crystals without thermodynamic sampling. In this work, we extend the model to treat modern machine learning potentials in both isochoric and isobaric ensembles. Testing against molecular dynamics simulations of Al and Cu, we find meV/atom accuracy in free energies up to the melting temperature under typical operating pressures, with similar accuracy for the thermal expansion. Our sampling-free estimation is universally superior to the quasi-harmonic approximation for a computational cost that is nearly two orders of magnitude lower and many orders of magnitude more efficient than thermodynamic integration. We discuss applications of the method in modern computational materials science workflows.

MM 44.8 Wed 17:45 C 264

Exploring Alternative Dispersion Corrections for the BEEF-vdW Functional — •Elisabeth Keller[1], Johannes T. Margraf[1], Volker Blum[2], and Karsten Reuter[1] — [1]Fritz-Haber-Institut der MPG, Berlin — [2]Department of Mechanical Engineering and Materials Science, Duke University, Durham, United States

The BEEF-vdW exchange-correlation functional was developed as a general purpose functional for chemistry and condensed matter physics. As such, BEEF-vdW gives a balanced description for molecular and solid-state systems, making it particularly well-suited to investigate surface science and catalytic processes. However, the use of a nonlocal vdW functional to describe dispersion interactions makes BEEF-vdW computationally expensive and difficult to incorporate in many electronic structure codes. In this work, we therefore explore the use of different alternative dispersion corrections to emulate the general applicability of BEEF-vdW in the FHI-aims code. We discuss the impact that the choice of dispersion correction (TS-vdW, MBD, MBD-NL, XDM) has on the accuracy and computational demand of BEEF-vdW for molecular, surface and solid-state systems.