MM 29: Data Driven Materials Science: Design of Functional Materials

Time: Thursday 10:15–11:30 Location: H45

MM 29.1 Thu 10:15 H45

Investigations of the Polysulfide Conversion Mechanism via Gaussian Approximation Potentials — •Xu Han[1,2], Carsten G. Staacke[1], Hendrik H. Heenen[1], Xuefei Xu[2], and Karsten Reuter[1] — [1]Fritz-Haber-Institut der MPG, Berlin, Germany — [2]Tsinghua University, Beijing, China

Lithium-sulfur (Li-S) batteries have been regarded as promising energy storage systems with ultra-high theoretical energy density. During a charging cycle Li_2S is converted to S_8 and vice-versa, where intermediate Li polysulfides (LiPS) are formed in a complex reaction mechanism which is still under debate. The theoretical exploration of the involved Li-S chemistry is challenged by an extended reaction network making it intractable for first principles methods. In contrast, machine learning interatomic potentials (MLIPs) which potentially retain predictive accuracy at a fraction of the computational cost are ideally suited for this task.

Here, we establish a training protocol for a Gaussian approximation potential (GAP) to simulate the chemistry of LiPS. Our training is based on a constrained on-the-fly exploration of the LiPS chemical space. In that, we enumerate the connectivity of (poly)cyclic LiPS and explore their stability via global optimization procedures with iteratively refined MLIPs. We use the final, sufficiently accurate MLIP to sample the LiPS phase space and to compute charging/discharging curves which we can directly compare to experimental data. Our MLIP calculations are expected to provide more fundamental insights into the LiPS conversion mechanism in Li-S batteries.

MM 29.2 Thu 10:30 H45

Accelerating the High-Throughput Search for new Thermal Insulators with Symbolic Regression — •Thomas Purcell[1], Matthias Scheffler[1,2], Luca M. Ghiringhelli[1,2], and Christian Carbogno[1] — [1]The NOMAD Laboratory at Fritz-Haber-Institut der Max-Planck-Gesellschaft — [2]FAIRmat at Humboldt Universität zu Berlin, Berlin, Germany

Reliable artificial-intelligence models are key to accelerate the discovery of new functional materials for various applications. Here, we present a general, data-driven framework that combines symbolic regression with sensitivity analysis to create hierarchical workflows. We illustrate the power of this new framework by screening for new thermally insulating materials. We first use the sure-independence screening and sparsifying operator (SISSO) [1] to build an analytical model that describes the thermal conductivity of a material and then extract out the most important input properties using a variance-based sensitivity analysis [2]. Using the information gained from the analysis we screen over a set of 732 materials and find the region of space most likely to contain strong thermal insulators. Finally we confirm these predictions by calculating thermal conductivities using the *ab initio* Green-Kubo technique [3].

[1] R. Ouyang, *et al.*. *Phys. Rev. Mat.* **2**, 083802 (2018)
[2] S. Kucherenko, S. Tarantola, and P. Annoni. *Comput. Phys. Commun.* **183**, 937 (2012)
[3] C. Carbogno, R. Ramprasad, and M. Scheffler. *Phys. Rev. Lett.* **118**, 175901 (2017)

MM 29.3 Thu 10:45 H45

Uncertainty Modelling for Property Prediction of Double Perovskites — •Simon Teshuva[1], Mario Boley[1], Felix Luong[1], Lucas Foppa[2], and Matthias Scheffler[2] — [1]Monash University, Melbourne, Australia — [2]Fritz Haber Institute, Berlin, Germany

Statistical predictive models for double perovskite properties are of high interest, because the perovskite structure allows relatively accurate property prediction and at the same time provides enough flexibility to yield a huge number of different materials of which some are likely relevant for important applications. Existing results published for this class of materials typically refer only to the predictive performance as, e.g., measured by the root mean squared error. However, active learning strategies for effective materials screening also rely on adequate uncertainty estimates as provided by probabilistic models.

Here, we study the predictive performance of two popular machine learning models, Gaussian processes and random forests, together with the quality of their uncertainty estimates. This study is based on a dataset of over 800 single (ABO_3) and double (A_2BB'O_6)cubic perovskite oxides with computed bulk modulus, cohesive energy, and bandgap. We show that Gaussian processes, while providing sound Bayesian uncertainty estimates, can have inferior performance when their assumption of isometric smoothness of the target property is not met. In this case, as exemplified by the double perovskite bandgaps, random forests provide a better alternative, despite their rather ad-hoc uncertainty estimates. Improving these estimates thus appears to be a promising direction for future research.

MM 29.4 Thu 11:00 H45

Automated effective Hamiltonian construction and active sampling of potential energy surface by Bayseian optimization — •Mian Dai, Yixuan Zhang, and Hongbin Zhang — Institute of Materials Science, Technical University of Darmstadt, Darmstadt, 64287, Germany

A first-principles effective Hamiltonian method can be used to simulate the phase transition sequences. In practice it is quite tedious to express the total energy surfaces and estimate reasonable parameters for high-order polynomials. We implemented Bayesian optimization (BO) to sample the total energy surfaces based on active learning and fit the set of parameters for constructing the effective Hamiltonians. Taking $BaTiO_3$ as a case study, we found that less than 30 sampling configurations with automated generated structures by BO are enough to determine a new set of parameters. The hyperparater in our BO process is tuned to show the improvement of the convergence for all fitted parameters. Using the new set of parameters, we perform Monte Carlo simulations which produce comparable phase transition temepratures with experimental values and previous results. Our BO algorithm has a great potential for future application in construction the effective Hamiltonians with more complicated subspace and effective atomic potentials describing the full lattice dynamics.

MM 29.5 Thu 11:15 H45

Predicting oxidation and spin states by high-dimensional neural networks — •Knut Nikolas Lausch[1], Marco Eckhoff[1], Peter Blöchl[2], and Jörg Behler[1] — [1]Georg-August-Universität Göttingen, Institut für Physikalische Chemie, Theoretische Chemie, Göttingen, Germany — [2]Technische Universität Clausthal, Institut für Theoretische Physik, Clausthal-Zellerfeld, Germany

Machine learning potentials (MLP) such as high-dimensional neural network potentials (HDNNP) provide first-principles quality energies and forces enabling large-scale molecular dynamics simulations at low computational costs. However, most current MLPs do not provide any information about the electronic structure of the system, which is often important for a detailed understanding of complex systems such as transition metal oxides. The lithium intercalation compound $Li_xMn_2O_4$ ($0 \leq x \leq 2$), a commercially used cathode material in lithium ion batteries, is such a system since the manganese ions adopt different oxidation states based on the lithium content and distribution. Here, we propose a high-dimensional neural network (HDNN) that can predict atomic oxidation and spin states as a function of the local atomic environments in $Li_xMn_2O_4$. The HDNN can complement HDNNP-driven MD simulations giving insights into the underlying electronic processes that give rise to complex phenomena such as a charge ordering transition, and electrical conductance.