

MM 4: Methods in Computational Materials Modelling (methodological aspects, numerics)

Sessions: Automated workflows and High throughput searches

Time: Monday 10:15–13:15

Location: H45

MM 4.1 Mon 10:15 H45

Formation enthalpies for automated computational materials design — ●RICO FRIEDRICH¹, DEMET USANMAZ¹, COREY OSES¹, ANDREW SUPKA², MARCO FORNARI², MARCO BUONGIORNO NARDELLI³, CORMAC TOHER¹, and STEFANO CURTAROLO⁴ — ¹Dept. of Mech. Eng. and Mat. Sci., Duke Univ. — ²Dept. of Phys. and Science of Advanced Materials Program, Central Michigan Univ. — ³Dept. of Phys. and Dept. of Chem., Univ. of North Texas — ⁴Mat. Sci., Elec. Eng., Phys. and Chem., Duke Univ.

The accurate calculation of formation enthalpies is crucial for computational materials design. For compounds chemically similar to their reference phases such as metal alloys, standard semi-local approximations to density functional theory (DFT) lead to accurate results [1]. When the phases are chemically dissimilar as in the case of oxides, DFT suffers from a lack of error cancellation leading to deviations of several hundred meV/atom compared to experimental values [2]. We use the automated computational materials design framework AFLOW [3] to validate correction schemes for *ab-initio* formation enthalpies [2, 4]. These empirical methods can improve DFT predictions by a factor of 4 to 7. Zero-point vibrational and thermal contributions to the formation enthalpy are found to largely cancel each other.

[1] S. Curtarolo *et al.*, *Calphad* **29**, 163-211 (2005).

[2] V. Stevanović *et al.*, *Phys. Rev. B* **85**, 115104 (2012).

[3] S. Curtarolo *et al.*, *Comput. Mater. Sci.* **58**, 218 (2012).

[4] L. Wang *et al.*, *Phys. Rev. B* **73**, 195107 (2006).

MM 4.2 Mon 10:30 H45

Accelerating high-throughput searches for new alloys with active learning of interatomic potentials — ●KONSTANTIN GUBAEV¹, EVGENY PODRYABINKIN¹, GUS HART², and ALEXANDER SHAPEEV¹ — ¹Skolkovo Institute of Science and Technology, Skolkovo Innovation Center, Nobel str. 3, Moscow 143026, Russia — ²Department of Physics and Astronomy, Brigham Young University, Provo, UT 84602, USA

We propose an approach to materials prediction that uses a machine-learning interatomic potential to approximate quantum-mechanical energies and an active learning algorithm for the automatic selection of an optimal training dataset.

Our approach significantly reduces the amount of density functional theory (DFT) calculations needed, resorting to DFT only to produce the training data, while structural optimization is performed using the interatomic potentials.

Our approach is not limited to one (or a small number of) lattice types (as is the case for cluster expansion, for example) and can predict structures with lattice types not present in the training dataset. We demonstrate the effectiveness of our algorithm by predicting the convex hulls for the following three systems: Cu-Pd, Co-Nb-V, and Al-Ni-Ti. Our method is three to four orders of magnitude faster than conventional high-throughput DFT calculations and explores a wider range of materials. In all three systems, we found unreported stable structures compared to the AFLOW database.

MM 4.3 Mon 10:45 H45

Automated error analysis and control for ab initio calculations — ●JAN JANSSEN, TILMANN HICKEL, and JOERG NEUGEBAUER — Max-Planck-Institut für Eisenforschung, Düsseldorf, Germany

Over the last years methodological and computational progress in atomistic simulations have substantially improved the predictive power in materials design. A critical prerequisite to ensure a reliable comparison between the ab initio computed data with experimental data is to quantify the various sources of uncertainty present in the ab initio calculations. These include systematic errors due to insufficient convergence, statistical or numerical errors due to incomplete sampling and model errors for derived quantities. A well-known example is the determination of the equilibrium lattice constant and bulk modulus, which requires a careful analysis of the fit of the ab initio data on an approximate analytic form such as the Murnaghan equation of state.

To automatize the complex analysis we have developed an algorithm which takes the precision in the derived quantity as a convergence goal and automatically determines the convergence parameter to achieve it.

This algorithm is implemented using pyiron (<http://pyiron.org>) - an integrated development environment (IDE) for computational material science. This tool provides an efficient and user friendly environment to implement complex simulation protocols and allows to run them as high-throughput simulations over the periodic table. Our investigations revealed that many of the commonly used rules of the thumb for fitting ground state materials properties become invalid for high precision calculations.

MM 4.4 Mon 11:00 H45

Flexible and User-friendly Symmetry-Constrained Optimization and its Application to Study Different SiO₂ Polymorphs — ●SARA PANAHIAN JAND¹, MAJA-OLIVIA LENZ¹, DAVID HICKS², STEFANO CURTAROLO², MATTHIAS SCHEFFLER¹, and CHRISTIAN CARBOGNO¹ — ¹Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin — ²Duke University, Durham NC, USA

In computational solid-state physics, crystal symmetries are frequently exploited to accelerate and improve electronic-structure calculations. However, the standard techniques are usually not applicable when the global symmetry is broken, e.g. in defective materials or thermodynamic investigations. To overcome this problem, we have developed and implemented flexible geometry-constrained relaxation in the all-electron code FHI-aims. This approach preserves the *local* symmetries on a per-atom basis via user-defined symmetry-reduced parameters as presented in the AFLOW library of prototypes [1]. We demonstrate the flexibility and power of the approach by studying the stabilities of different SiO₂ polymorphs. For this compound, it is generally difficult to model structural transitions at very high temperatures and/or pressures [2]. However, our method provides a rapid way to compute the phase diagram of SiO₂, including the temperature-dependent lattice expansion. This demonstrates the ability of the developed approach to aid and accelerate the systematic search for metastable structures and the calculation of their thermodynamic properties.

[1] M. J. Mehl, *et al.*, *Comp. Mater. Sci.* **136**, S1 (2017).

[2] C. Catlow, *et al.*, *Phys. Chem. Chem. Phys.*, **12**, 786 (2010).

MM 4.5 Mon 11:15 H45

Uncertainty Quantification Framework for Force-Matched Classical Potentials — ●PETER BROMMER and SARAH LONGBOTTOM — University of Warwick, Coventry, UK

Classical effective potentials, where the energy of the system (and thus the forces on the atoms) only depend on the atomic positions, underpin a significant fraction of molecular dynamics (MD) simulations. Unfortunately, the bias, approximations and uncertainties incurred when representing the complex energy landscape by an effective potential or force field with a limited number of parameters are badly controlled, and the impact on quantities of interest (QoI) of an MD simulation is *a priori* unknown.

For potentials obtained by force matching, i.e. by adjusting their parameters to optimally reproduce forces, stresses and energies from first-principles calculations, we present an extension to the force-matching package *potfit*, where the uncertainty in the parameter set is encapsulated in an ensemble of potentials. This allows propagation of the uncertainty to QoI and makes uncertainty quantification accessible to users. We demonstrate this capability using three distinct potentials for Nickel and determine predictions and confidence intervals for lattice constants, elastic constants and thermal expansion.

15 min. break

MM 4.6 Mon 11:45 H45

On finding the dipole in the haystack of MD trajectories — ●FREDERIC FELSEN, DAVID EGGER, and CHRISTOPH SCHEURER — Theoretical Chemistry, Technical University Munich

Aiming at implicit solvation models for large scale simulations of chemical reactions, we took a first step in the development of an automated solvent characterization based on classical MD trajectories. By modifying a method for the detection of semi-rigid domains in macromolecular structures [1], we propose a coarse graining procedure for decomposing a charge neutral atomistic system into a multi-center dipole

representation. The procedure is formulated in terms of a series of linearly constrained quadratic programming problems, which describe a clustering of the particles with respect to three major properties: spatial compactness, as the multi-center expansion is in terms of ideal point dipoles, structural rigidity, to ensure a valid mapping over the whole trajectory, and local charge neutrality. Solving these optimization problems by means of an active set algorithm yields the dipole domain decomposition of the system of interest. Based on such a dipole representation, an expression for the system's polarization can be obtained which is directly related to the dielectric function of the medium [2]. Investigating molecular (liquids and macromolecules) as well as crystalline (MoS₂ sheet structures) test systems, the key features of the coarse graining will be discussed.

[1] S. Bernhard, F. Noé, PLOS ONE, 5, (2010).

[2] D. Bonthuis, S. Gekle, R. Netz, *Langmuir*, 28(20), (2012).

MM 4.7 Mon 12:00 H45

Looking for Needles in the Haystack with Fuzzy Glasses: Screening Crystal Structures with Approximate Electronic Structure Methods — ●SIMON WENGERT, CHIARA PANOSETTI, JOHANNES T. MARGRAF, and KARSTEN REUTER — Technische Universität München, Germany

Still largely a Holy Grail of modelling, the capability to reliably predict the structure of (molecular) crystals would allow for the *in silico* design of substances with desired characteristics. Application fields range from dissolution properties (and consequently the bioavailability) of drugs to optoelectronic properties in organic photovoltaics. The main challenges in this respect are the large number of possible polymorphs and the small energy differences between them. In practice, there is thus a trade-off between the ability to screen a wide range of candidates (which requires some fast evaluation of stability) and applying levels of theory that adequately describe the interplay between subtle intermolecular interactions such as H-bonding, electrostatic and dispersion effects.

Dispersion-corrected density-functional theory (DFT) is typically considered to be sufficiently accurate for this purpose, but too expensive for routine global structural screening applications. We therefore compare two approximate DFT-models, namely the Harris approximation and density-functional tight-binding (DFTB) with respect to their efficiency and accuracy. We find that DFTB is surprisingly accurate, yielding mostly correct rankings in the targeted low-energy range. The errors of both approaches are discussed in depth.

MM 4.8 Mon 12:15 H45

Screening of rare-earth-lean intermetallic 1-11-X, 1-12-X, and 1-13-X compounds for hard-magnetic applications — WOLFGANG KÖRNER¹, ●DANIEL F. URBAN¹, and CHRISTIAN ELSÄSSER^{1,2} — ¹Fraunhofer IWM, Freiburg, Germany — ²University of Freiburg, FMF, Germany

We report on comparative theoretical investigations of ferromagnetic rare-earth-transition-metal phases with underlying YNi₉In₂ (“1-11”), ThMn₁₂ (“1-12”), and LaCo₉Si₄ (“1-13”) crystal structures. By a combinatorial high-throughput-screening (HTS) approach based on density functional theory the intrinsic key properties of hard magnets, namely the magnetization M , energy product $(BH)_{\max}$ and uniaxial magnetocrystalline anisotropy constant K_1 are estimated [1,2]. We interpret the results in terms of the differences in the three structure types and analyze the effect of additional light interstitial atoms ($X = B, C, N$). In the HTS study, which considers Ce, Nd or Sm as rare-earth elements and Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Al, Si, and P on the transition metal sites, several promising phases are identified which partially outperform Nd₂Fe₁₄B in terms of $(BH)_{\max}$ and K_1 values, and contain significantly less rare-earth atoms.

[1] W. Körner, G. Krugel, and C. Elsässer, *Sci. Rep.* 6, 24686 (2016).

[2] W. Körner, G. Krugel, D. F. Urban and C. Elsässer, *Scripta Mat.* 154, 295 (2018).

MM 4.9 Mon 12:30 H45

A local Bayesian optimizer for atomic structures — ●ESTEFANÍA GARIJO DEL RÍO, JENS JØRGEN MORTENSEN, and KARSTEN WEDEL JACOBSEN — CAMD, Department of Physics, Technical University of Denmark

A local optimization method based on Bayesian Gaussian Processes is developed and applied to atomic structures. The method is applied to a variety of systems including molecules, clusters, bulk materials, and molecules at surfaces. The approach is seen to compare favorably to standard optimization algorithms like conjugate gradient or BFGS in most cases. The method relies on prediction of surrogate potential energy surfaces, which are fast to optimize, and which are gradually improved as the calculation proceeds. The method includes a few hyperparameters, the optimization of which may lead to further improvements of the computational speed.

MM 4.10 Mon 12:45 H45

Machine learning enhanced atomistic simulation of ZrB₂ at ultra-high temperatures — ●YANHUI ZHANG, ALESSANDRO LUNGI, and STEFANO SANVITO — School of Physics and CRANN, Trinity College Dublin, Dublin, Ireland

Machine-learned interatomic potentials (MLIP) are emerging as the tool of choice for molecular dynamics, since they exhibit robustness in large-scale atomistic simulations at a quasi-*ab initio* accuracy. Here we demonstrate the construction of such MLIP for the long-standing problem of extracting high-temperature properties of ultra-high temperature ceramics (UHTCs). Although some effort has been devoted in the past decades, the atomistic simulation at high temperatures is still sluggish. The development of a MLIP for UHTCs rises major challenges since it must simultaneously describe: 1) the constituent elements being much different from each other; 2) the nature of the bonding arising from a mixture of metallic, covalent and ionic interaction, 3) the complex response to heat and deformation loads, 4) the transferability across a wide range of temperatures and strains. All these attributes are extremely important for the accurate prediction of fundamental physical properties of UHTCs. We have developed a powerful MLIP using the SNAP model, which bears excellent temperature transferability. Thereafter, we have applied it to study the performances of UHTCs under heat and strain loads. Our simulations extend to temperature (up to 3000 K) far beyond what available in measurements. This work demonstrates that MLIPs are a very promising simulation tool in the field of ultra-high temperature materials.

MM 4.11 Mon 13:00 H45

SnP anodes for potassium-ion batteries: insights from computational structure prediction — ●MATTHEW EVANS¹, KENT GRIFFITH^{2,3}, and ANDREW MORRIS⁴ — ¹TCM Group, Cavendish Laboratory, University of Cambridge, UK — ²Department of Chemistry, University of Cambridge, UK — ³Department of Materials Science and Engineering, Northwestern University, USA — ⁴School of Metallurgy and Materials, University of Birmingham, UK

Replacing Li with more abundant alkali metals, Na and K, should decrease and stabilise the production cost of rechargeable batteries. One hurdle to overcome for Na- and K-ion batteries is the poor performance and capacity of carbonaceous anodes relative to those used in Li batteries. Alloying or conversion anodes provide an alternative route, provided destructive volume changes and resulting nanoparticle pulverisation can be mitigated.

In this talk, we shall present a computational crystal structure prediction study on the K-Sn-P ternary system, following recent experimental interest in Sn/P as conversion anodes for K-ion batteries. Using a combination of *ab initio* random structure searching (AIRSS), data mining and evolutionary approaches, several new stable crystal structures have been discovered. These structures are used to aid interpretation of ambiguous experimental results using the matador package (<https://matador-db.readthedocs.io>). Theoretical capacities of the two stable anode materials, SnP₃ and Sn₄P₃, are predicted to increase relative to the known phases by 31% and 19% respectively, with the novel ternary phases providing a route to minimise volume expansion.