MM 14: Development of Computational Methods: Thermodynamics and Local Chemistry, Electronic Structure

Time: Tuesday 10:15–13:00 Location: SCH A 251

MM 14.1 Tue 10:15 SCH A 251

Performance of pseudopotentials in chemically diverse materials — •Andris Gulans[1] and Stefan Goedecker[2] — [1]University of Latvia, Riga, Latvia — [2]University of Basel, Switzerland

We present a novel framework for assessing quality of pseudopotentials. It comprises a database of atomization energies and new highly precise norm-conserving pseudopotentials. The database covers more than 700 molecules with 65 elements from six rows of the periodic table. It accounts for chemical diversity by considering every atom in a variety of oxidation states. For each molecule, we provide highly precise scalar-relativistic atomization energies obtained with linearized augmented plane waves obtained using PBE exchange-correlation functionals. The new pseudopotentials reproduce the reference data within the chemical accuracy limit of 1 kcal/mol. They are crucial for assessing other pseudopotentials, as we use them for decomposing the total error in atomization energies into individual atomic contributions. Our calculations reveal that even state-of-the-art pseudopotential families are still not fully compliant with chemical accuracy, and the largest errors arise in molecules with atoms in high oxidation states.

MM 14.2 Tue 10:30 SCH A 251

Pushing to the limits of machine-learning potentials for high-entropy alloys — •Konstantin Gubaev, Victor Zaverkin, Prashanth Srinivasan, and Blazej Grabowski — University Stuttgart

Chemically complex multicomponent alloys have garnered widespread interest owing to their exceptional properties coming from a sheer inexhaustible compositional space. The complexity poses severe challenges for atomistic modelling and interatomic potential development. Here, we explore the limits of two complementary state-of-the-art machine-learned potentials—the moment tensor potential (MTP) and the Gaussian moment neural network (GM-NN)—in simultaneously describing both the configurational *and* vibrational degrees of freedom in the prototype Ta-V-Cr-W alloy family. Both models are equally accurate with exceptional performance in comparison to classical potentials. A single potential is able to achieve root-mean-square-errors of 1.37-4.35 meV/atom and 0.023-0.057 eV/Å in 0 K energies and forces, respectively, across all subsystems of the alloy family, and 0.156-0.179 eV/Å in high-temperature molecular dynamics forces for the disordered quaternary. MTPs achieve faster convergence with the training size than the GM-NNs, whereas GM-NNs are faster in execution. Active learning is partially beneficial and has to be complemented with a conventional human-based training set generation.

MM 14.3 Tue 10:45 SCH A 251

Extracting free energies from local composition fluctuations in solids: Theoretical background and atomistic simulations — Daniel Bitter[1], Marvin Poul[2], Guido Schmitz[1], and •Sebastian Eich[1] — [1]Institute of Materials Science, University of Stuttgart, Heisenbergstraße 3, D–70569 Stuttgart, Germany — [2]Max-Planck-Institut für Eisenforschung GmbH, Max-Planck-Straße 1, D–40237 Düsseldorf, Germany

While thermodynamic fluctuation theory has been applied to liquids for decades in order to obtain direct thermodynamic information, i.e. Gibbs free energies, from local composition fluctuations, the present work offers an extension for the application in solids. It is demonstrated that composition fluctuations are affected by an additional elastic work term which arises only in solids, thus covering liquids as a special case. The extended fluctuation model is verified through atomistic simulations in an exemplary Cu–Ni embedded-atom system for which Monte Carlo simulations have been carried out in the semi-grandcanonical ensemble at fixed temperature over the entire composition range. Composition fluctuations were monitored in a subvolume over time and statistically evaluated in terms of the variance, demonstrating perfect agreement with the prediction from the extended model. This method has been developed primarily for the application to experimental atom probe data, where three-dimensional chemical information is available with sub-nanometer accuracy, allowing a direct measurement of local composition fluctuations to extract thermodynamic information.

MM 14.4 Tue 11:00 SCH A 251

Extracting free energies from local composition fluctuations in solids: Application on the atom probe data obtained from TAPSim simulations — •Jianshu Zheng[1], Marvin Poul[2], and Sebastian Eich[1] — [1]Institute of Materials Science, University of Stuttgart, Heisenbergstr. 3, D-70569 Stuttgart, Germany — [2]Max-Planck-Institute für Eisenforschung GmbH, Max-Planck-Straße 1, D-40237 Düsseldorf, Germany

This work proposes a novel methodology to extract direct thermodynamic information, i.e. Gibbs free energies, from local composition fluctuations found in atom probe data based on statistical mechanics. The proof-of-concept is demonstrated with an exemplary simulated Cu–Ni alloy, which has first been equilibrated using Monto Carlo techniques with an embedded-atom potential and subsequently field evaporated by TAPSim[1]. It is shown that the variance of the local composition on the reconstructed data reveals a significant dependence on the size of the chosen (sub-) volume used for the evaluation, but nevertheless the extrapolation to an infinitely large volume unveils a remarkable link to the curvature of the Gibbs free energy. Given the composition range is explored at several points, the Gibbs excess free energy of mixing could be recovered in a CALPHAD-style parametrization. This methodology promises to improve the accuracy of thermodynamic information from direct atom probe measurements.

[1] C. Oberdorfer, S.M. Eich, G. Schmitz. Ultramicroscopy 128, (2013), 55.

MM 14.5 Tue 11:15 SCH A 251

A general-purpose framework for kinetic Monte-Carlo simulations — •Roya Ebrahimi Viand and Sebastian Matera — Fritz Haber Institute of the Max Planck Society, Berlin, Germany

The kinetic Monte Carlo (kMC) method has gained popularity in the last years for the simulation of problems whose dynamics are controlled by rare events. Applications range from atomic-scale models for heterogeneous catalysis or charge transport to disease modeling, and a number of specialized codes have been developed for different classes of models. However, specialization is often the bottleneck when trying to extend the codes to a more general setting or to implement new algorithms. We present our first steps towards a flexible general-purpose framework for implementing kMC models to overcome these limitations. This assumes nothing more than that the model can be regarded as a continuous time, discrete state Markov process, which is common among all kMC models. The specific physical model must be provided by the user in terms of update functions for the transition rates of the different possible elementary processes. Besides illustrating the usage on simple and well investigated models, we demonstrate how the tool can be used to implement models with long range interactions, e.g., for the simulation of charge transport in solid materials.

15 min. break

MM 14.6 Tue 11:45 SCH A 251

Pressure-driven tunable properties of the small-gap chalcopyrite topological quantum material ZnGeSb2: A first-principles study — •Surasree Sadhukhan[1], Banasree Sadhukhan[2], and Sudipta Kanungo[1] — [1]Indian Institute of Technology Goa, 403401 Ponda, India — [2]KTH Royal Institute of Technology, Stockholm

Search for new topological quantum materials is the demand to achieve substantial growth topological phase of matter. In this search process, theoretical prediction is crucial besides the obvious experimental verification. The divination of topological properties in already well-known narrow gap semiconductors is flourishing in quantum material science. We revisited the semiconductor compound in the chalcopyrite series, some of which were potential topological materials. Using this density functional theory-based first-principles calculations, we report a strong topologically nontrivial phase in chalcopyrite ZnGeSb 2, which can act as a model system of strained HgTe. The estimates reveal the non-zero topological invariant (Z 2), Dirac cone crossing in the surface spectral functions with spin-momentum locked spin texture. We also report the tunable topological properties from nontrivial to trivial phases under

moderate hydrostatic pressure within *7 GPa. A minor modification of a lattice parameter is enough to achieve this topological phase transition easily accomplished in an experimental lab. We have incorporated the discontinuity in the tetragonal distortion of non-centrosymmetric ZnGeSb 2 to drive the topological quantum phase transition.

MM 14.7 Tue 12:00 SCH A 251

electronic structure of a non-symmorphic kondo lattice system CeAgSb2 — •Sawani Datta[1], Khadiza Ali[2], Rahul Verma[1], Saroj p. Dash[2], Bahadur Singh[1], Arumugam Thamizhavel[1], and Kalobaran Maiti[1] — [1]Tata institute of fundamental research, Mumbai, India — [2]Chalmers university of technology, Goteborg, Sweden

Topological Dirac semimetal with non-symmorphic symmetry (NSS) is one of the most recent discoveries in the field of novel topological quantum materials [1]. The uniqueness of this system is the presence of robust (including spin-orbit coupling) Dirac line nodes with NSS protection. CeAgSb2, a Kondo system, exhibits a large anisotropic resistivity and a complex magnetic ground state at a lower temperature [2]. Employing angle-resolved photoemission spectroscopy (ARPES), we find that CeAgSb2 possesses several non-trivial crossings near the Fermi level protected by the NSS. The ARPES data collected at different photon energies suggest a quasi-2-D behavior which is consistent with the transport results. In addition, we observe energy bands of the surface states in the near-Fermi-level region along with the non-trivial bulk bands that make the electronic structure complex. Our study shows the CeAgSb2 family of materials as a new platform to study the robust non-symmorphic symmetry-protected Dirac semi-metallic systems with high spin-orbit coupling.

[1] L. M. Schoop et. al., Nat. Commun. 7, 11696 (2016). [2] Y. Inada et. al., Philos. Mag. B, 82, 1867 (2002).

MM 14.8 Tue 12:15 SCH A 251

An amplitude expansion of magnetic phase field crystal: A continuum model for magnetically driven dislocation networks — •Rainer Backofen, Marco Salvalaglio, and Axel Voigt — Institute of Scientific Computing, TU Dresden

The amplitude expansion of the phase-field-crystal (APFC) model enables a convenient coarse-grained description of crystalline structures. It allows the description of length scales that are orders of magnitude larger than the lattice spacing while retaining microscopic features, such as dislocations. Adaptive finite elements are used to treat the different involved length scales efficiently [1].

Here, an APFC model is presented that captures the basic physics of magneto-structural interactions: magnetostriction and magnetic anisotropy. The free energy of APFC is combined with a continuum field representing local magnetization [2]. The proposed coupling does not increase the partial differential equation to solve compared with the basic APFC model. Still, it is flexible enough to adapt the magnetic anisotropy of the model to basic ferromagnetic materials such as Fe and Ni. Exploiting this model, the influence of external magnetic fields on the evolution of defects and grain boundaries is discussed.

[1] Salvalaglio, Backofen, Voigt (2018) PRMat, 2(5), 053804. [2] Backofen, Salvalaglio, Voigt (2022) Model Simul Mat Sci Eng

MM 14.9 Tue 12:30 SCH A 251

Multi-orbital models within the ghost Gutzwiller approximation — •Carlos Mejuto-Zaera and Michele Fabrizio — SISSA, Trieste, Italy

In the pursuit towards targeted material design leveraging strong electronic correlation, computationally inexpensive yet qualitatively reliable methods play a fundamental role. These approaches should allow for a rapid mapping of phase space, unveiling a first impression of possible phases of matter, which can then be explored in selected regions of parameter space with more accurate yet involved techniques. Recently, the ghost Gutzwiller Approximation (gGA) has been shown to be an interesting candidate for this kind of phenomenological search. Based on a self-consistency condition for the simple one-body reduced density matrix of a discretized impurity model, this method can capture spectral features of both coherent and incoherent nature in the one-body Green's function. In this work we extend its applicability to the multi-orbital regime employing a truncated solver based on selected configuration interaction. This allows for increasing the number of correlated orbitals while keeping the size ratio of bath to impurity constant. We explore the reliability of the gGA for describing the phase diagram of multi-band models, comparing to more sophisticated embedding methods. We shall assess the potential of gGA for modelling of complex materials, possibly in combination with ab-initio methods.

MM 14.10 Tue 12:45 SCH A 251

Understanding the success of mGGAs for band gaps. Is it the orbital dependence? — •Péter Kovács, Peter Blaha, and Georg K. H. Madsen — Institute of Materials Chemistry, Technical University of Vienna

Density functional theory has shown remarkable success in predicting various properties of solids, such as lattice parameters and cohesive energies, yet with most functionals it is known to heavily underestimate band gaps. mGGA functionals tend to result in better band gap predictions than LDA or GGAs, but the best results are still achieved at the cost of accuracy for the other properties. Recently using a systematic search in the space of mGGA functionals we were able to find functionals, where this tradeoff is small.[Péter Kovács et al., J. Chem. Phys. 157, 094110 (2022)]

While the failure of LDA and GGA functionals for gaps are often attributed to their lack of the derivative discontiunity, the success of mGGAs can not be explained solely based on their ability to exhibit discontinuous behaviour. On a database of 440 solids, we analyze how two specialized functionals, TASK and our own mGGA23, are able to better predict gaps. We aim to understand what parts of the functional shape is responsible for their success and how these affect their prediction of structural properties, such as lattice parameter. These findings can be used in functional design and also to understand failure cases of already existing functionals.