

MM 22: Topical session: Integrated computational materials engineering for design of new materials V

Time: Tuesday 11:45–13:30

Location: H39

MM 22.1 Tue 11:45 H39

Structure prediction of bulk materials via compressed sensing — ●RUNHAI OUYANG, LUCA M. GHIRINGHELLI, and MATTHIAS SCHEFFLER — Fritz-Haber-Institut der Max-Planck-Gesellschaft, 14195 Berlin-Dahlem, Germany

Structure prediction of materials from only their chemical compositions can greatly accelerate the novel material discovery and design. Fast structure prediction can be made by building a structure map based on available knowledge from material database. The key for accurate prediction is to identify the physical parameters (descriptor) that determine the material structures. While most of the current methods experience the problem of a biased intuitive feature selection or having difficulty in solving ill-conditioned linear systems when considering a big feature space, we employ the rather recent compressed sensing theory from signal and image processing field for the descriptor identification. Here, we focus on the prediction of crystal structure of ternary semiconductors and insulators with covalent and ionic bonding by building a map on which the structures are classified. Machine learning is performed on first-principle materials data from the NoMaD repository (<http://nomad-repository.eu>), complemented by newly performed density-functional calculations. We show how a physically interpretable descriptor can be identified unbiasedly and systematically by using the least absolute shrinkage and selection operator (LASSO) method from a big feature space (size n) but with a relatively small sample data set (size m , $m \ll n$).

MM 22.2 Tue 12:00 H39

Sampling parameter sensitivities for kinetic Monte Carlo models — MAX J. HOFMANN², FELIX ENGELMANN³, and ●SEBASTIAN MATERA¹ — ¹FU Berlin, Germany — ²U Stanford, USA — ³TU München

We present a three-step procedure for the sampling of local sensitivities (parameter gradients) from kinetic Monte Carlo models, which severely reduces the computational cost compared with standard numerical differentiation. In the first step, we utilize the Relative Entropy Method[1] for obtaining upper/lower bounds for the derivatives and discard those parameters with vanishing sensitivity. For the remaining, we sample the derivatives from a single trajectory employing an estimator based on Linear Response Theory. It turns out that some sensitivities can very well be sampled, while others show a higher variance. Only for the later, we perform a numerical differentiation using Coupled Finite Differences (CFD)[2]. We demonstrate the approach by revisiting the sensitivity analysis of the CO oxidation on RuO₂ (110)[3]. We find that CFD is only necessary for a small fraction of the sensitivities. [1] Y. Pantazis and M. A. Katsoulakis, *J.Chem. Phys.* 138 (2013): 054115; [2] D. F. Anderson, David, *SIAM J. Num. Analysis* 50 (2012): 2237-2258; [3] H. Meskine, S. Matera, M. Scheffler, K. Reuter, K., H. Metiu, (2009). *Surf. Sci.*, 603 (2009): 1724-1730

MM 22.3 Tue 12:15 H39

An adaptive sparse grid approach for quantifying the error propagation in first principles kinetic Monte Carlo models — ●SANDRA DÖPKING¹, DANIEL STROBUSCH², CHRISTOPH SCHEURER², and SEBASTIAN MATERA¹ — ¹FU Berlin, Germany — ²TU München, Germany

First principles kinetic Monte Carlo (1p-kMC) models utilize Density Functional Theory (DFT) based rate constants (RCs) as the input for stochastic mesoscopic reactivity models. Despite the success of DFT, the employed approximations can easily lead to uncertainties in the RCs of two orders of magnitude. We quantify the error propagation to the 1p-kMC simulation results by a global sensitivity analysis (GSA), i.e. we decompose the error in the result into contributions caused by each uncertain RC. The required high dimensional integrals over the space of the RCs are numerically evaluated using locally and dimension-adaptive sparse grids. With this approach, we can exploit that, typically, strongly non-linear behavior only appears in small sub-domains (local adaptivity) and on low-dimensional subspaces (dimension-adaptivity) of the whole parameter space. As a prototypical example, we consider the 1p-kMC model for the CO oxidation on a RuO₂(110) surface. Comparing with local (linear) sensitivity analysis, we find qualitative differences between both approaches, but

also in the global setting only a small portion of all RCs has an impact at all. Possible applications of the approach are the reduction of the number of expensive first principles simulations or the identification of descriptor sets in computational materials screening.

MM 22.4 Tue 12:30 H39

Machine Learning of the (Meta-)Stability of Octet Binaries — ●EMRE AHMETCIK, CHRISTIAN CARBOGNO, LUCA M. GHIRINGHELLI, and MATTHIAS SCHEFFLER — Fritz-Haber-Institut der Max-Planck-Gesellschaft, 14195 Berlin-Dahlem, Germany

Statistical learning is regarded as the most promising technique to accelerate and systematically facilitate insights into computational material science. For instance, this has been recently successfully demonstrated by using compressed sensing techniques to predict the relative stability of zincblende versus rocksalt octet binary materials from the properties of the atomic constituents alone [1]. For an application in practical material science, it is however uncertain to which extent these approaches can be generalized, e.g., to predict metastable polymorphs or thermodynamic properties. To clarify this question, we have computed the relative stability of octet binaries for several different (meta-)stable crystal structures. We discuss the applicability of statistical learning for this question, how the approach can be generalized to predict thermodynamic properties such as transition pressures, and demonstrate its robustness with respect to numerical parameters. In particular, we critically discuss to which extent further data, e.g., dimeric and/or thermodynamic descriptors, are necessary.

[1] L. M. Ghiringhelli et. al., *Phys. Rev. Lett.* 114, 105503 (2015).

MM 22.5 Tue 12:45 H39

Automated calculations for charged point defects in MgO and α -Fe₂O₃ — ●SUDARSAN SURENDRALAL, MIRA TODOROVA, and JÖRG NEUGEBAUER — Department for Computational Materials Design, Max-Planck-Institut für Eisenforschung GmbH, Max-Planck-Straße 1, 40237 Düsseldorf, Germany

The power of modern supercomputers with efficient electronic structure codes allows the rapid screening of huge numbers of systems and properties. A prerequisite for such calculations are user-friendly high-level programming interfaces that provide highly flexible automated computational tools which assist the user, for example, in setting-up or post-processing calculations, facilitate archiving a targeted accuracy or high-throughput screening of materials. Using concepts of modern object oriented programming and database management we have developed *PyIron*, a robust computational materials python library. The capabilities of *PyIron* will be demonstrated and discussed using the automation of charged point defects calculations for MgO and α -Fe₂O₃. Our density functional theory calculations are performed using the semi-local GGA functional for MgO and the GGA+U approach for α -Fe₂O₃. The FNV finite supercell charge correction [1] is applied to correct for spurious interactions resulting from the use of periodic boundary conditions. Subtleties of its automated application, the effect of the Coulombic interaction parameter U has on the band gap of α -Fe₂O₃ and on the defects' thermodynamics, will be discussed.

[1] C. Freysoldt, J. Neugebauer, and C. G. Van De Walle, *Phys. Rev. Lett.* 102, 016402 (2009).

MM 22.6 Tue 13:00 H39

Ab initio materials design — SILVANA BOTTI¹ and ●MIGUEL MARQUES² — ¹Friedrich-Schiller University of Jena, Germany — ²Martin-Luther University of Halle-Wittenberg, Germany

One of the most exciting developments in condensed matter over the past years is, without doubt, materials design. This new discipline aims at solving the called inverse problem: given a certain desired property (or properties), discover (design) the material that possesses this property under a given set of constraints. These constraints can be related to the mechanical or chemical stability of the compounds, their price, their availability, etc. The target properties can be related to applications, e.g., in photovoltaics, batteries, etc.

Theoretical approaches based and going beyond density functional theory ally today accuracy and efficiency, which make them suitable tools for predicting electronic properties of “real” materials. Thanks to

the availability of supercomputers, the powerful combination of theoretical spectroscopy with high-throughput calculations and structural prediction allows to explore the periodic table in search of new materials, providing a precious starting point for further experimental studies.

MM 22.7 Tue 13:15 H39

Bonding and electron-phonon interaction in high- T_c hydrides
— ●CHRISTOPH HEIL and LILIA BOERI — University of Technology Graz

High-temperature superconductivity in hydrogen-rich compounds has been predicted by Ashcroft almost 50 years ago [1], yet was only recently observed in experiments by Drozdov et al. [2], who measured a

critical temperature above 190 K in hydrogen-sulfide at 200 GPa pressure. In a similar experiment, the same group reported a T_c of 100 K in pressurised phosphine [3].

In this talk, I will show that the record-breaking T_c in H_3S does not originate from the heavier atoms exerting pressure on the hydrogen lattice, but that it is due to the formation of unusually strong polar covalent bonds, stabilised under extreme pressures. I will discuss the bonding, electron-phonon coupling and the superconducting properties of sulfur-hydrides and highlight possible routes to realise similar bonding conditions in other hydrogen-rich compounds [4].

[1] N.W. Ashcroft, Phys. Rev. Lett. 21, 1748 (1968)

[2] A.P. Drozdov, M.I. Erements and I.A. Troyan, Nature 525, 73 (2015)

[3] A.P. Drozdov, M.I. Erements and I.A. Troyan, arXiv:1508.06224

[4] C. Heil and L. Boeri, Phys. Rev. B 92, 060508(R) (2015)