

## MM 62: Methods in Computational Materials Modelling (methodological aspect, numerics)

QM-based interaction models

Time: Thursday 15:45–17:15

Location: TC 006

MM 62.1 Thu 15:45 TC 006

**Uncertainty Quantification for Classical Effective Potentials** — ●SARAH LONGBOTTOM and PETER BROMMER — University of Warwick, Coventry, UK

Effective potentials are an essential ingredient of classical molecular dynamics simulations. Little is understood of the errors incurred in representing the complex energy landscape of an atomic configuration by an effective potential containing considerably fewer parameters. The probabilistic sloppy model method [1] has been implemented in the potfit force matching code [2]. This introduces uncertainty quantification into the interatomic potential generation process. Uncertainties in the effective potential are propagated through MD to obtain uncertainties in quantities of interest, which are a measure of the confidence in the model predictions.

We demonstrate the technique using three potentials for nickel: two simple pair potentials, Lennard-Jones and Morse, and a local density dependant EAM potential. A sloppy model fit to DFT reference data is constructed for each potential to calculate the uncertainties in lattice constants, elastic constants and thermal expansion. These can be used to show the unsuitability of pair potentials for nickel. In contrast, with EAM we observe a decreased uncertainty in the model predictions. This shows that our method can capture the effects of the error incurred in the potential generation process without resorting to comparison with experiment or DFT.

[1] S. L. Frederiksen, et al., Phys. Rev. Lett., 93 (16), 2004.

[2] P. Brommer, et al., Modell. Simul. Mater. Sci. Eng., 23 (7), 2015.

MM 62.2 Thu 16:00 TC 006

**Combination of machine learning and high-throughput DFT calculations for the prediction of thermodynamic stability**

— ●JONATHAN SCHMIDT<sup>1</sup>, JINGMING SHI<sup>4</sup>, PEDRO BORLIDO<sup>2</sup>, LIMING CHEN<sup>3</sup>, SILVANA BOTTI<sup>2</sup>, and MIGUEL MARQUES<sup>1</sup> — <sup>1</sup>Institut für Physik, Martin-Luther-Universität Halle-Wittenberg, Germany — <sup>2</sup>Institut für Festkörpertheorie und -optik, Friedrich-Schiller-Universität Jena and European Theoretical Spectroscopy Facility, Germany — <sup>3</sup>Département MI, Laboratoire ICTT, Ecole Centrale de Lyon, France — <sup>4</sup>Institut Lumière Matière, Université de Lyon, France

We perform a large-scale benchmark of machine learning methods for the prediction of the thermodynamical stability of solids. We start by constructing a data set that comprises density functional theory calculations of around 250,000 cubic perovskite systems. Incidentally, around 500 of these are thermodynamically stable but are not present in crystal structure databases. This data set is then used to train and test a series of machine learning algorithms to predict the distance to the convex hull of stability. In particular, we study the performance of ridge regression, random forests, extremely randomized trees (including adaptive boosting), and neural networks. We find that extremely randomized trees give the best results and use this method in combination with DFT to explore ternary compounds with the  $AB_2C_2$  composition. By using machine learning we reduce the overall calculation cost by around 75% and find that there may be 10 times more stable compounds in these phases than previously known.

MM 62.3 Thu 16:15 TC 006

**Improving the training of force-fields based on neural networks** — ●MÁRIO MARQUES and MIGUEL MARQUES — Institut für Physik, Martin-Luther-Universität Halle-Wittenberg, 06099 Halle (Saale), Germany

The evaluation of potential energy surfaces lies at the heart of many problems in materials science. Density functional theory is often used for this task, but it quickly becomes impractical for systems with hundreds or thousands of atoms.

In this contribution we describe a methodology, based on Behler-Parrinello approach for artificial neural networks, to solve this problem. We obtain training and test sets from a fully unbiased approach based on global structural prediction techniques.

We extend the back propagation method to consider the error in the forces and stress and we discuss the suitability of a few activation functions. Finally, we develop force fields for Si and Ge, and present some applications to the calculation of point defects and phase diagrams.

MM 62.4 Thu 16:30 TC 006

**DFTB for lazy theorists: automatic transferable parametrization with particle swarm optimization** — ●CHIARA PANOSSETTI<sup>1</sup>, REINHARD J. MAURER<sup>2</sup>, and KARSTEN REUTER<sup>1</sup> — <sup>1</sup>Technische Universität München — <sup>2</sup>University of Warwick

In terms of numerical efficiency and accuracy, tight-binding is intermediate between first-principles and empirical methods. Among the many flavors used in materials modelling, Self-Consistent Charge Density Functional Tight Binding (SCC-DFTB)[1] is particularly geared to cover a broad range of bonding situations while retaining access to electronic properties. This renders it appealing for use, *e.g.*, as a lower-level pre-screening technique in hierarchical approaches. However, its pairwise parametrization requires a procedure which constitutes an  $N^2$  effort within the periodic table and is not only tedious, but also rather arbitrary and difficult to document and reproduce.

Building on the work of Chou *et al.*, [2] we here propose, extend and critically discuss a new parametrization strategy where all the quantities determining the SCC-DFTB interaction are optimized with a particle swarm algorithm to minimize a generalized observable-based cost function. Careful use of such a powerful tool should facilitate reproducibility and provide a straightforward recipe to fine tune the balance between transferability and accuracy.

[1] M. Elstner, Theor. Chem. Acc. 116, 316 (2006).

[2] C. P. Chou *et al.*, J. Chem. Theory Comput. 12, 53 (2016).

MM 62.5 Thu 16:45 TC 006

**Systematic development of analytic bond-order potentials for W, Re and Os**

— ●APARNA P. A. SUBRAMANYAM, JAN JENKE, THOMAS HAMMERSCHMIDT, and RALF DRAUTZ — ICAMS, Ruhr University Bochum, Bochum, Germany

The limited applicability of accurate quantum mechanical simulations to extended time and length scales calls for approximate interatomic interaction models in order to perform large-scale atomistic simulations. In this work we show the coarse graining of the electronic structure to tight binding and analytic bond-order potentials (BOPs).

We present the systematic parameterization of BOPs for the transition metals W, Re and Os. In particular we demonstrate the systematic parameterisation from downfolding the DFT eigenspectrum to a TB basis that is then solved on the BOP level. The accuracy of the BOPs is validated with respect to the structural stability, to the formation energies of point defects and the formation energies of planar defects like stacking faults and twin boundaries. We furthermore validate the transferability of the BOPs with a comprehensive set of different atomic environments.

MM 62.6 Thu 17:00 TC 006

**Charge transport in organic semiconductors: Towards an ACKS2-based polarization in force fields.**

— ●PATRICK GÜTLEIN<sup>1</sup>, HARALD OBERHOFER<sup>1</sup>, KARSTEN REUTER<sup>1</sup>, and JOCHEN BLUMBERGER<sup>2</sup> — <sup>1</sup>Technische Universität München, Germany — <sup>2</sup>University College London, UK

Charge carrier transport, an integral part of diverse reactions and devices, is generally subject to the non-negligible response of the surrounding environment. Especially in organic semiconductors the accurate theoretical treatment of dynamic electronic rearrangements in dense media is a complex task. The high anisotropy and comparatively low dielectric screening require system sizes that challenge even efficient first-principles approaches like density-functional theory (DFT). In this situation the recently proposed atom-condensed Kohn-Sham density functional theory approximated to second order (ACKS2) approach[1] could represent a computationally undemanding, yet accurate extension to conventional force field methods. In this approach, the linear response of the electron density and Kohn-Sham potential are captured by an atom-centered series expansion up to second order.

For a range of molecular model systems we systematically compute the ACKS2 parameters with DFT and discuss linear response properties obtained by a Gaussian type orbital basis set representation. This marks an important first step in making ACKS2 applicable as a polarizable force field. [1] T. Verstraelen *et al.*, J. Chem. Phys. 138, 7 (2013); *ibid* 141, 19 (2014).