

MM 33: Computational Materials Modelling: Process Schemes / Oxides

Time: Thursday 15:45–18:30

Location: H44

MM 33.1 Thu 15:45 H44

How to Speed up First-Principles Based Geometry Optimization with Small Numerical Basis Sets — ●ELISABETH KELLER, JOHANNES T. MARGRAF, and KARSTEN REUTER — Fritz-Haber-Institut der Max-Planck-Gesellschaft, Berlin, Germany

First-principles based geometry optimizations are often the most expensive part of high-throughput virtual screening studies for functional materials. This is particularly true for large systems, i.e. when studying complex surface reconstructions or nanoparticles. Here, the computational cost is strongly influenced by the size of the basis set. Large, converged basis sets result in precise equilibrium geometries, yet demand high computational cost and thus limit the simulation scale. Semiempirical methods using minimal basis sets offer a much lower computational cost, but may yield unacceptably large and uncontrolled errors. Furthermore, the availability of adequate parameterizations is rather sparse across the periodic table.

In this presentation, we will discuss the potential of using near-minimal basis sets for accelerating and enabling large-scale geometry optimizations at the DFT level. For this purpose, we studied how the size of the numeric atom-centered orbital (NAO) basis set in FHI-aims impacts the accuracy of bulk geometries. We recover equilibrium geometries at a nearly converged level with a highly compact basis by employing a simple short-ranged pair-potential correction. We show the scheme's ability to treat different systems across the periodic table ranging from small molecules and clusters to large-scale bulk and surface structures as well as complex molecule-surface interactions.

MM 33.2 Thu 16:00 H44

A machine-learned interatomic potential for crystalline and amorphous silica — ●LINUS ERHARD¹, JOCHEN ROHRER¹, KARSTEN ALBE¹, and VOLKER DERINGER² — ¹Institute of Materials Science, Technische Universität Darmstadt, Otto-Berndt-Strasse 3, 64287 Darmstadt, Germany — ²Department of Chemistry, Inorganic Chemistry Laboratory, University of Oxford, Oxford OX1 3QR, United Kingdom

Fitting an interatomic potential for silicon oxide that can be used for both the amorphous and numerous crystalline phases has proven to be difficult. This is already shown by the large number of interatomic potentials published in the last decades. Here, we present a machine-learned interatomic potential for silica, which is highly transferable between different crystalline polymorphs and the amorphous phase. It predicts the thermodynamics of the system accurately and is able to generate low-defect amorphous models by melt and quench simulations. We also discuss the importance of choosing an appropriated exchange-correlation functional for density-functional data input, which is particularly important for silica. Since the generation of realistic amorphous structure models by melt-quench simulations is highly dependent on the quench rate, we show new ways via hybrid simulations that combine the speed of classical interatomic potentials with the accuracy of machine-learning potentials. We also investigate the extrapolation behavior of our machine-learning potential using high-pressure simulations. Finally, we show first steps towards an interatomic potential for mixed Si-SiO₂ systems.

MM 33.3 Thu 16:15 H44

An all-functionals automatic workflow for IR and Raman spectra — ●LORENZO BASTONERO¹ and LORENZO BASTONERO^{1,2} — ¹University of Bremen, Bremen, Germany — ²EPFL, Lausanne, Switzerland

IR and Raman spectroscopies are among the best methods for the characterisation of materials at small scales, thanks to their fast measurement and high sensibility to local composition and configuration. Theoretical calculations are fundamental for the interpretation of experimental results and for the assessment of thermal properties. DFT has been employed in the last decades as a reliable tool for the analysis of these spectra, although the calculation of vibrational properties has been limited to the use of few functionals. Here, we devise an automatic user-friendly workflow for IR and Raman calculations within the AiiDA infrastructure, which exploits the finite displacements and finite fields to allow application to any complex functional. The package provides at the same time easy access and full customisability, relevant both for less experienced users and more elaborate purposes such as

high-throughput searches.

MM 33.4 Thu 16:30 H44

A Workflow for Obtaining Robust Density Functional Tight Binding Parameters Across the Periodic Table — ●MENGHAN CUI, JOHANNES T. MARGRAF, and KARSTEN REUTER — Fritz-Haber-Institut der MPG, Berlin, Germany

The Density Functional Tight Binding (DFTB) approach allows electronic structure based simulations at length and time scales far beyond what is possible with first-principles methods. This is achieved by using minimal basis-sets and empirical approximations. Unfortunately, the sparse availability of parameters across the periodic table is a significant barrier to the use of DFTB in many cases.

In this contribution, we therefore propose a workflow which allows the robust and consistent parameterization of DFTB across the periodic table. Importantly, the approach requires no element-pairwise parameters and can thus easily be extended to new elements. Specifically, the parameters defining the band energy and repulsive potential are obtained via Bayesian Optimization on a set of elemental solids. In this way, robust baseline parameters can be obtained for arbitrary element combinations. The transferability of the parameters and applications in hybrid DFTB/Machine Learning models will be discussed.

MM 33.5 Thu 16:45 H44

ChemiTEM - optimized solutions and workflows for electron microscopy in materials science and chemistry — ●WALID HETABA^{1,2}, ROBERT IMLAU³, LISETH DUARTE-CORREA², MAXIMILIAN LAMOTH², STEPHAN KUJAWA³, and THOMAS LUNKENBEIN² — ¹Max-Planck-Institut für Chemische Energiekonversion, Mülheim/Ruhr, Deutschland — ²Fritz-Haber-Institut der MPG, Berlin, Deutschland — ³Thermo Fisher Scientific, Eindhoven, Niederlande

Transmission electron microscopy (TEM) is an important and versatile method for investigating materials on the nanoscale. Information about the elemental composition and electronic structure can be obtained while imaging the sample with atomic resolution. Such investigations are usually performed by TEM-experts. However, enabling non-expert TEM users to perform such measurements would tremendously improve the efficiency of TEM investigations in both, materials science and chemistry. We therefore developed ChemiTEM: a set of standardized workflows for data acquisition and analysis which are integrated in an app for tablets and smartphones to provide easy access for all TEM users, irrespective of their level of experience. We tested the ability of ChemiTEM in helping non-expert TEM users to collect high quality data by having non-expert and expert TEM users investigate the same sample. Using ChemiTEM, the data acquired by the non-expert users were of similar quality to that of the data recorded by the TEM-expert. Thus, we were able to show that using the ChemiTEM app, TEM can be made available to everyone working in materials science.

15 min. break

MM 33.6 Thu 17:15 H44

Determination of Formation Energies and Phase Diagrams of Transition Metal Oxides with DFT+U — ●DANIEL MUTTER¹, DANIEL URBAN¹, and CHRISTIAN ELSÄSSER^{1,2} — ¹Fraunhofer IWM, Freiburg, Germany — ²Freiburger Materialforschungsinstitut (FMF), Freiburg, Germany

Knowledge about formation energies of compounds is essential to derive phase diagrams of multicomponent phases with respect to elemental reservoirs. The determination of formation energies using (semi-)local exchange-correlation approximations of the density functional theory exhibits well-known systematic errors if applied to oxide compounds containing transition metal elements. We generalize and reevaluate a set of approaches proposed and widely applied in the literature to correct for errors arising from the over-binding of the O₂ molecule and from correlation effects of electrons in localized transition-metal orbitals. The DFT+U method is exemplarily applied to iron oxide compounds, and a procedure is presented to obtain the U values, which lead to formation energies and electronic band gaps comparable to the experimental values. Using such corrected formation energies, we derive phase diagrams for LaFeO₃, Li₅FeO₄, and NaFeO₂,

which are promising materials for energy conversion and storage devices. A scheme is presented to transform the variables of the phase diagrams from the chemical potentials of elemental phases to those of precursor compounds of a solid-state reaction, which represents the experimental synthesis process more appropriately. The workflow and methods can directly be applied to other transition metal oxides.

MM 33.7 Thu 17:30 H44

A comparative study of the bulk properties of iron oxides calculated using empirical potentials and ab-initio calculations

— ●AHMED ABDELKAWY, MIRA TODOROVA, and JÖRG NEUGEBAUER — Max-Planck-Institut für Eisenforschung, Max-Planck-Str.1, 40470 Düsseldorf

Striving toward a green economy requires re-evaluating industrial processes and looking for new routes to, e.g., obtaining iron from iron ore. This necessitates understanding the steps involved in the reduction of iron oxides and leading to several phase transitions the final product of which is iron. The optimization of the involved processes will hugely benefit from an atomistic level understanding. Atomistic simulation techniques are versatile but are dependent on the quality of the used underlying empirical potentials. We therefore first evaluate the applicability of interatomic potentials for this study by comparing them to ab initio calculations. Specifically, we focus on a reactive forcefield (ReaxFF) parametrized for these materials, which accounts for the different oxidation states of each species (Fe and O) and enables their variation depending on the local environment and coordination, bond breaking, and making. We assess the reliability of the force field by assessing various bulk properties of the relevant iron as compared to density functional theory calculations. We find that a universal forcefield that is able to accurately describe the three main iron oxides (Magnetite, Hematite, and Wüstite) is difficult to obtain.

MM 33.8 Thu 17:45 H44

Uncertainty in Predicting Thermodynamic Properties of TiO₂ Polymorphs

— ●OLGA VINOGRADOVA, PIN-WEN GUAN, SIYING LI, and VENKATASUBRAMANIAN VISWANATHAN — Carnegie Mellon University, Pittsburgh, USA

Polymorphism of crystals directly leads to materials with vastly different chemical and physical properties. However the lowest energy polymorphs often differ by only small amounts of energy. This makes it challenging to predict relative properties using first-principles density functional theory (DFT), which is significant in designing a material for the desired application. In this work we apply computational uncertainty within DFT to quantify the accuracy of stability and phase transition predictions under finite temperature and pressure. We study six polymorphs of TiO₂ using a set of six exchange-correlation functionals to present a detailed sensitivity analysis using uncertainty capabilities within the Bayesian Error Estimation Functional. We show that a prediction confidence metric is particularly important for comparing the stability of numerically close predictions. We show how the choice of functional significantly affects predictions of phase transitions and identify which structures and properties that have inherently large uncertainties. From the trends observed in stability, finite-temperature, and phase transition pressure predictions we propose that uncertainty quantification provides a valuable insight in problems where drawn

conclusions are highly sensitive to the choice of the functional.

MM 33.9 Thu 18:00 H44

Self-consistent phonon calculations of lattice dynamical properties in cubic EuTiO₃ comparing with experimental thermal conductivity

— ●CHEN SHEN¹, WENJIE XIE¹, XINGXING XIAO¹, ANKE WEIDENKAFF¹, TERUMASA TADANO², and HONGBIN ZHANG¹ — ¹Institute of Materials Science, Technical University Darmstadt, Darmstadt 64287, Germany — ²Research Center for Magnetic and Spintronic Materials, National Institute for Materials and Science, Tsukuba, Japan

We investigate the role of the quartic anharmonicity in the lattice dynamics and thermal transport of the cubic EuTiO₃ by combining ab initio self-consistent phonon theory combined with compressive sensing techniques experimental thermal conductivity determination measurement. The antiferromagnetic G-type magnetic structure is used to mimic the para-magnetic EuTiO₃. We find that the strong quartic anharmonicity of oxygen atoms plays an essential role in the phonon quasiparticles free from imaginary frequencies in EuTiO₃, causing the hardening of vibrational frequencies soft modes. The hardened modes thereby affect calculated lattice thermal conductivity significantly, resulting in an improved agreement with experimental results, including the deviation from $\kappa_L \propto T^{-1}$ at high temperature. The calculated thermal conductivity of 8.2 W/mK at 300 K matched the experimental value of 6.1 W/mK. When considering the boundary scattering, the calculated thermal conductivity is reduced to 6.9 W/mK at 300 K, which agrees better with the experiment.

MM 33.10 Thu 18:15 H44

Simulated indentation on graphene oxide

— ●JAVIER ROJAS-NUNEZ¹, SAMUEL BALTAZAR¹, EDUARDO BRINGA², and ALEJANDRA GARCIA³ — ¹Physics Department and CEDENNA, Universidad de Santiago de Chile (USACH), Santiago, Chile — ²Laboratorio de síntesis y modificación de nanoestructuras y materiales bidimensionales, Centro de Investigación en Materiales Avanzados, Nuevo León, México — ³CONICET & Facultad de Ingeniería, Universidad de Mendoza, Mendoza, Argentina

The better understanding of nanomaterial properties will be a key factor to tailor and enhance properties of new materials. Graphene oxide in particular can be synthesized with different oxidation levels in order so gain similar properties to its deoxidized counterpart, graphene. Through the molecular dynamic simulations, the atomistic behavior of a tri-layer graphene membrane under mechanical indentation will be studied in this work.

This work will study a highly oxidized graphene oxide tri-layer that will be indented with a repulsive spherical indentator. The modeling of the membrane will generate single layer graphene oxide candidates to pick the lowest energy configuration and later stack this layer over itself. The final tri-layer was used for the indentation simulation, where the young modulus was reproduced with decent similarity to experimental results.

The atomistic analysis of the indentation process suggest an important role of epoxide groups in the mechanical deformation of the membrane.