MM 10: Methods in Computational Materials Modelling (methodological aspects, numerics)

Sessions: Atomic Potentials and Phonons

Time: Monday 15:45–18:45   Location: H45

MM 10.1   Mon 15:45   H45

Accuracy vs. efficiency? Towards ACKS2-based polarization in force fields — •Patrick Gütlein[1], Karsten Reuter[1], Harald Oberhofer[1], and Jochen Blumberger[2] — [1]Technische Universität München, Germany — [2]University College London

Electronic polarization poses a minor energy contribution compared to the formation and arrangement of covalent bonds, yet it crucially influences both geometric and electronic properties of soft condensed matter and molecular crystals. Frequently, effective force field techniques employed to sample the phase space of such systems insufficiently cover the complex dielectric response to external perturbations. Here, the recently proposed atom-condensed Kohn-Sham density functional theory approximated to second order (ACKS2) approach promises to be a computationally undemanding, still accurate electronic polarization extension to conventional force field methods. It rests on an atom-centered basis set expansion of the linear responses of both the electron density and the Kohn-Sham potential.

In order to provide a generally applicable ACKS2 extension to force fields, we develop a new minimalistic basis set representation composed of s- and p-type Gaussian functions. We demonstrate the efficacy of ACKS2 with the new basis set for a range of molecular model systems including anthracene and tetracene, which are relevant building blocks in common organic semiconductors. Screening a range of external electrostatic perpurbations, we find very good agreement with density functional theory references. This marks an important first step to employ ACKS2 as polarization correction in force fields.

MM 10.2   Mon 16:00   H45

Validation and transferablity of interatomic potentials — •Yury Lysogorskiy, Thomas Hammerschmidt, and Ralf Drautz — ICAMS, Ruhr University Bochum, Germany

Interatomic potentials (IP) are widely used in computational materials science, in particular for simulations that are too computationally expensive for density functional theory (DFT). A large number of IPs is available for a wide range of chemical elements and their mixtures. Most IPs have a limited application range and often there is little information available regarding their performance for specific simulations.

We performed extensive tests for the majority of the potentials for unaries that are available from the OpenKIM and NIST repositories as well as from other sources. The following properties were considered: energy-volume curves, equilibrium bulk modulus, elastic constants, phonon spectrum and density of states, vacancy formation energies, transformation paths, surface energies, thermodynamic properties in the quasiharmonic approximation and thermal expansion. For accessing the transferability of IPs we cover a wide range of atomic environments by considering special random structures with one and two atoms in the unit cell and evaluated their energetic, geometrical and elastic properties. Two typical cases of IP behavior - high accuracy at the expense of low transferability versus low accuracy but higher transferability were identified. The results of our calculations are collected in a specially designed database for further analysis.

MM 10.3   Mon 16:15   H45

Parametrization of classical force fields for single molecule junctions — •Narendra P. Arasu and Hector Vazquez — Institute of Physics of the Czech Academy of Sciences, Cukrovarnicka 10, Prague, Czech Republic

In molecular junctions, where single molecule bridges the gap between metal electrodes, interface geometry under the influence of thermal fluctuations can be explored from Molecular Dynamics (MD) simulations. Ab-initio methods are accurate but their computational cost restricts the time scale that can be probed in the simulations. Thus, simulations based on Density Functional Theory (DFT) can be limited to rather restricted phase-space. On the other hand, Molecular Mechanics (MM) simulations are computationally faster than DFT. However, careful parametrization of the MM energy functions is required for realistic representation of an MD ensemble. In this work, we propose a method to parametrize Optimized Potentials for Liquid Simulations (OPLS-AA) forcefield to run a long time scale (~ nanoseconds) MD simulations. The parameters are obtained by fitting the model energy

functions of short-range interactions to the energies from DFT calculations. As a prototypical molecular junction, we consider a biphenyl molecule with three different linker groups having different chemical binding: Au-C, Au-NH2, and Au-SCH3 to test our parametrization technique. We compare the statistics of changes in bond lengths, angles and dihedrals obtained from classical MD trajectories to that of the ab-initio MD simulations. The results show that the parameters obtained using our method describe well DFT MD simulations.

MM 10.4   Mon 16:30   H45

Parametrization of a bond-order potential for Ti — •Alberto Ferrari, Malte Schröder, Yury Lysogorskiy, Jutta Rogal, Matous Mrovec, and Ralf Drautz — Interdisciplinary Centre for Advanced Materials Simulation, Ruhr-Universität Bochum, 44801 Bochum, Germany

Ti alloys are attractive structural materials, featuring remarkable stiffness/weight and strength/weight ratios, exceptional corrosion resistance, good creep properties, and biocompatibility. In addition, many Ti alloys exhibit shape-memory and superelastic properties, and the recently discovered gum-metals are also based primarily on Ti. Atomic-scale studies of Ti-based metals often employ first-principles methods such as density functional theory, with maximum system sizes that are limited to a few hundreds atoms. To perform large-scale and/or finite temperature simulations, empirical potentials or machine-learning potentials are usually fitted. However, the transferability of these potentials to properties or environments not included in the training is limited. In this contribution, we present a newly developed bond-order potential (BOP) for Ti derived by coarse-graining the tight-binding approximation to the electronic structure. The BOP model shows an unprecedented transferability to various bulk as well as defective Ti structures. We demonstrate the excellent predictive power of our potential for a number of structural and thermodynamic properties that are relevant for the structural stability, phase transformations, and defect behavior in Ti-based materials.

MM 10.5   Mon 16:45   H45

Learning to use the force: DFTB repulsion with Gaussian Process Regression — Artur Engelmann, •Chiara Panosetti, Johannes T. Margraf, and Karsten Reuter — Chair for Theoretical Chemistry, Technische Universität München, Germany

Density-Functional Tight Binding (DFTB) is increasingly popular among computational modellists as it provides comparable accuracy to DFT at a fraction of the cost, enabling large scale simulations while retaining direct access to electronic structure properties. Yet, a bottleneck to this day remains the difficulty to parametrize the interactions for large subsets of atoms across the periodic table, let alone an extensive, universal parametrization. Especially challenging is the parametrization of the pairwise repulsion: an unescapable N^2 effort, and a cumbersome one. Most schemes involve fitting the repulsion to some analytical potential by *e.g.* minimizing force residues. However, this presents a number of limitations, such as constraints on the reference geometries, and the necessity of projecting forces along the bonds. Further, any predefined functional form not only carries a certain degree of arbitrary bias in its very choice, but may also lack the flexibility to capture subtle features around equilibrium distances (where the "repulsive" potential may as well be attractive). We thereby propose to rather machine-learn the repulsive force, using Gaussian Process Regression similarly to the generation of GAP potentials [1]. We discuss a proof-of-principle application on carbon, showing how such an approach removes all the above limitations at once.
[1] A.P. Bartók *et al.*, PRL 104, 136403 (2010)

MM 10.6   Mon 17:00   H45

A Neural Network Potential for Lithium Manganese Oxides — •Marco Eckhoff[1], Peter Blöchl[2], and Jörg Behler[1] — [1]Universität Göttingen, Institut für Physikalische Chemie, Theoretische Chemie, Tammannstraße 6, 37077 Göttingen, Germany — [2]Technische Universität Clausthal, Institut für Theoretische Physik, Leibnizstraße 10, 38678 Clausthal-Zellerfeld, Germany

The lithium manganese oxide spinel $Li_xMn_2O_4$, with $0 < x < 2$, is an

important cathode material in lithium ion batteries. Its accurate description by density functional theory (DFT) is far from trivial due to several energetically close electronic and magnetic states. In extensive benchmark studies we find that the hybrid functionals PBE0, HSE06, and PBE0r yield energetic, structural, electronic, and magnetic properties in good agreement with experiment. Building on such hybrid DFT data, we are able to extend the time and length scales of molecular dynamics simulations of $Li_xMn_2O_4$ using a high-dimensional neural network potential, which provides a first-principles quality description of the potential energy surface at a fraction of the computational costs.

15 min. break

MM 10.7   Mon 17:30   H45

Machine learning for training lattice based models — •Mattias Ångqvist, Erik Fransson, J. Magnus Rahm, and Paul Erhart — Chalmers University of Technology, Department of Physics, Gothenburg, Sweden

Creating lattice based models for studying configurational and vibrational effects has now become possible with little technical effort. The basic idea is that the models will be trained using reference data which commonly comes from expensive density functional theory calculations. If the training goes well the model will both accurately predict the training data but may also predict unseen data and do so at a fraction of the computational cost. The training is evaluated by cross validation by splitting up the reference data into testing and training sets. The number of optimization algorithms that one may choose to do the training with are very numerous. In this talk I will present how the different optimization algorithms perform when training these so called lattice based models both with respect to error estimation and to the prediction of e.g. thermodynamic quantities.

MM 10.8   Mon 17:45   H45

Reliable methods for combining tight binding models based on maximally localized Wannier functions — •Jae-Mo Lihm and Cheol-Hwan Park — Department of Physics, Seoul National University, Seoul 08826, Korea

Maximally localized Wannier functions (MLWFs) form a set of localized orthogonal basis functions of a periodic system which accurately represent given Bloch states. [1] MLWFs are commonly used as building blocks for large-scale tight-binding calculations. For example, MLWFs generated from ab initio calculations on the bulk and a thin slab of a given material may be used to construct a tight-binding model for a thicker slab, or a semi-infinite slab. However, the corresponding MLWFs of different systems are not identical to their partners. This discrepancy may cause artifacts in the physical quantities calculated from the combined tight-binding model if not properly taken care of.

In this work, we propose two post-processing methods that correct the differences between the MLWFs in two different systems. First, we describe a minimal correction which is simple and efficient, but fails in complex systems. Next, we propose a more accurate and generally applicable correction method based on the hopping parameters. We demonstrate the utility of our correction methods by applying them to real materials. We also compare the corrected MLWFs with the Wannier functions constructed without maximal localization.

[1] N. Marzari et al., Rev. Mod. Phys. 84, 1419 (2012)

MM 10.9   Mon 18:00   H45

Phonon-Related Properties in Metal-Organic Frameworks from First Principles — •Tomas Kamencek[1], Sandro Wieser[1], Egbert Zojer[1], and Natalia Bedoya-Martínez[1,2] — [1]Institute of Solid State Physics, NAWI Graz, Graz University of Technology, Austria — [2]Materials Center Leoben, Austria

Metal-organic frameworks (MOFs) are crystals consisting of metallic and organic constituents, which form open and porous structures. These materials have been extensively studied during the last years due to their numerous possible applications exploiting the large amount of internal surface area (e.g. catalysis, storage, capture and separation of gases). Phonon-related properties of MOFs, despite their importance for describing practically relevant quantities as thermal conductivity or thermal expansion, are still largely unexplored. Moreover, the huge number of building blocks and ways of connecting them to assemble a MOF opens the possibility to design materials with tailor-made properties. This calls for developing an in-depth understanding of phonons and phonon-related properties in MOFs. Therefore, we studied the influences of different constituents on the (an)harmonic phonon properties by means of atomistic simulations. Starting from MOF-5 we have systematically varied metallic nodes and organic linkers in order to deduce reliable structure-to -property relationships for phonon properties in MOFs. Our simulations have been performed in the framework of density functional tight binding theory with a focus on predicting elastic constants, phonon dispersions relations, and deduced quantities.

MM 10.10   Mon 18:15   H45

Phonon filters in molecular junctions — •Alvaro Rodriguez Mendez[1,2], Leonardo Medrano Sandonas[1,2], Rafael Gutierrez[1], Jesus Ugalde[3], Vladimiro Mujica[4], and Gianaurelio Cuniberti[1,5] — [1]Institute for Materials Science and Max Bergmann Center of Biomaterials, TU Dresden, 01062 Dresden, Germany — [2]Max Planck Institute for the Physics of Complex Systems, 01187 Dresden, Germany — [3]Kimika Fakultatea, Euskal Herriko Unibertsitatea (UPV/EHU) and Donostia International Physics Center (DIPC); P.K.1072; 20080 Donostia; Euskadi (Spain) — [4]Arizona State University, School of Molecular Sciences, Tempe, AZ 85287, USA. — [5]Dresden Center for Computational Materials Science (DCMS), TU Dresden, 01062 Dresden, Germany

We provide computational support to the intriguing idea that heterogeneous molecular junctions, consisting of molecular wires bridging two different nano-contacts, can act as a phonon filter. The filtering effect is ultimately due to the asymmetric nature of the contacts that translate into different interfacial energy transferring properties. The most important finding is the appearance of gaps on the phonon transmittance spectrum, which are strongly correlated to the properties of the vibrational spectrum of the molecule in the junction. This filtering effect may have very important technological implications for the design of smart nano-devices and nano-materials that can be used to control the asymmetric transmission of heat.

MM 10.11   Mon 18:30   H45

Phonon spectrum, phase stability and thermodynamic properties of the cubic pi-phase in the IV-VI monochalcogenides - formation and effect of a phonon band gap — •Uri Argaman, Ran Abutbul, and Guy Makov — Materials Engineering Department, Ben-Gurion University of the Negev, Beer Sheva 84105, Israel.

The pi phase is a recently discovered cubic phase in the IV-VI monochalcogenides with theoretical and technological interest (ref. 1). In this work, the phonon frequencies were calculated ab-initio in the cubic pi-phase of the monochalcogenides in four materials: SnS, SnSe, GeS and GeSe. All four were found to be mechanically stable, and a phonon band gap appears in their spectra. The phonon spectra of all these systems exhibit phonon band gaps in the optical phonon spectrum with widths of 0.7-2 THz. The origin of the phonon band gap was examined through an analysis of the normal modes. The contributions of the geometry of the bonds and mass differences to the phonon spectrum and, in particular, to the band gap are discussed and analyzed. We found differences in the relative motion of the two types of atoms and the rigidity of the bonds in this low symmetry structure. The effect of the phonon band gap on the thermodynamic properties is discussed. In addition, Raman experiment supported the existence of the phonon band gap in pi-SnS and pi-SnSe.

1. Abutbul, R. E., Segev, E., Argaman, U., Makov, G. and Golan, Y. (2018), Monochalcogenide Semiconductors: π*Phase Tin and Germanium Monochalcogenide Semiconductors: An Emerging Materials System. Adv. Mater., 30: 1870310. doi:10.1002/adma.201870310