MM 64: Liquid and Amorphous Materials IV

Time: Thursday 16:45–18:00   Location: C 243

MM 64.1   Thu 16:45   C 243

Machine learning quantum Monte Carlo: application to water clusters — •Matteo Peria, Michele Casula, and Antonino Marco Saitta — IMPMC, Sorbonne Université, CNRS, MNHN, UMR 7590, 4 Place Jussieu, 75252 Paris, France

A complete understanding of the hydrogen bond and proton transfer mechanism in water is still lacking, since it requires an accurate potential energy surface (PES) and very expensive quantum mechanical simulations of the nuclear part. Reproducing this high-dimensional surface with current high-level computational chemistry methods is infeasible for the largest clusters. We test gradient-based kernel ridge regression methods and neural networks to reproduce the PES starting from a dataset of energies and forces of the protonated water clusters obtained via simulations combining classical molecular dynamics (MD) for the nuclei and quantum Monte Carlo (QMC) for the electrons. The QMC+MD approach yields very accurate results for the classical dynamics, which are however affected by the intrinsic noise inherent in the stochastic sampling of both nuclear and electronic phase space. We prove that QMC multivariate noise is not detrimental to the learning of energies and forces and that the derived machine learning force field can be used to run long and reliable quantum molecular dynamics simulations.

MM 64.2   Thu 17:00   C 243

Device-scale atomistic modelling of phase-change memory materials using a machine-learned interatomic potential — •Yuxing Zhou[1,2], Wei Zhang[2], En Ma[2], and Volker L. Deringer[1] — [1]Department of Chemistry, University of Oxford, UK — [2]Center for Alloy Innovation and Design, Xi'an Jiaotong University, China

Phase-change materials (PCM) are leading candidates for next-generation memory and neuromorphic computing chips. The Ge–Sb–Te alloys on the GeTe-Sb2Te3 tie-line (referred to as "GST") have been most widely studied and used in commercial memory products. Quantum-accurate computer simulations have played a central role in understanding complex GST alloys. However, the large computational cost has precluded simulations on the length scales of real devices. In this presentation, we describe a single, compositionally flexible machine-learning interatomic potential with a quantum-mechanical level of accuracyy. We show that our model can describe the flagship GST alloys under various practical device conditions, e.g., non-isothermal heating, and taking chemical disorder into account. The superior computing efficiency of the new approach enables the simulation of multiple thermal cycles. We also show a device-scale capability demonstration in a real device model of more than 500,000 atoms. These describe technologically relevant processes in realistic memory products. Our work demonstrates how atomistic ML-driven simulations can help study the structural and chemical properties as well as programming mechanisms of GST devices.

MM 64.3   Thu 17:15   C 243

Towards in-depth atomistic understanding of polymer-derived silicon oxycarbides using machine-learning potentials — Niklas Leimeroth, •Jochen Rohrer, and Karsten Albe — Institute of Materials Science, Technical University Darmstadt, Otto-Berndt-Strasse 3, 64287, Darmstadt, Germany

Polymer-derived amorphous silicon oxycarbides (SiOC) show promising properties for advanced applications in a variety of fields such as high-temperature coatings, biomedicine and batteries. This outstanding versatility is due to their highly tunable composition and microstructure. Simultaneously, this tunability poses a challenge for a thorough knowledge and understanding of structure-property relations in this system. In this work, we present a machine-learning

potential (MLP) for SiOCs based on the atomic cluster expansion (ACE) and trained to a diverse set of actively-learned density functional theory (DFT) data. We demonstrate the capability of the MLP to model glass-phase and microstructure formation from commonly used polymer-precursor fragments, contrast these microstructures with experimental findings and show how atomistic simulations can be used to understand complex structure-property relations on the example of Young's Moduli in relation to phase volumes and different types of bonding in the system.

MM 64.4   Thu 17:30   C 243

Modelling amorphous forms of complex hybrid-inorganic frameworks — •Thomas C. Nicholas, Daniel F. Thomas du Toit, Andrew L. Goodwin, and Volker L. Deringer — Department of Chemistry, Inorganic Chemistry Laboratory, University of Oxford, Oxford, UK

With the continued development of efficient fitting and evaluation frameworks for machine learning potentials (MLPs), modelling complex, multi-component materials is now possible. However, for systems built up of metal nodes connected by organic linkers, such as metal-organic frameworks (MOFs), the time-scales and length-scales required to model and label representative amorphous training configurations using traditional database generation strategies (for example, ab initio molecular dynamics and iterative training) remains a challenge.

We demonstrate a two-stage approach to tackle this, focusing on modelling the amorphous form of a MOF built up from Zn nodes and imidazolate linkers ($Zn[C_3N_2H_3]_2$). Firstly, by exploiting the structural analogy between silica networks and this MOF, we construct a topologically and geometrically diverse database of configurations through a back-mapping scheme whereby we decorate AB_2 networks with Zn nodes and imidazolate linkers. Secondly, we introduce an iterative training protocol whereby training configurations are generated using a Monte Carlo simulation refinement that seeks to minimise the difference between the computed and experimental data.

In this way, we demonstrate how our MLP better describes possible amorphous configurations.

MM 64.5   Thu 17:45   C 243

Thermodynamic Assessment and CALPHAD Simulation of the Ni-Pd-S Glass Forming Ternary System — •Maryam Rahimi Chegeni[1], Wenhao Ma[2], Sascha Riegler[1], Amirhossein Ghavimi[1], Magnus Rohde[2], Hans Jürgen Seifert[2], Isabella Gallino[3], and Ralf Busch[1] — [1]Saarland University, Chair of Metallic Materials, Germany — [2]Karlsruhe Institute of Technology, Institute for Applied Materials-Applied Materials Physics, Germany — [3]Department of Materials Science and Engineering, Metallic Materials, TU-Berlin, Ernst-Reuter-Platz 1, 10587 Berlin, Germany

This work conducts an experimental and computational investigation into the thermo-physical properties of the novel ternary BMG-forming Ni-Pd-S system. The simplicity of the ternary Ni-Pd-S BMG-forming system facilitates the application of the CALPHAD approach for the modeling of the underlying thermodynamics affecting the glass formation.

Experimental quantitative specific heat capacity and crystallization studies of glass-forming compositions are performed to assess thermodynamic parameters and generate input data for the calculations and modeling of the undercooled liquid. Using the two-state approach , the modeling of the undercooled liquid and glass for the pure elements and the corresponding ternary system is done.

Subsequently, the evaluation of the glass forming ability of the system is performed using the parallel tangent method for Gm of the crystalline and the liquid phases and the results are used for modeling the isothermal TTT diagrams.