

## MM 62: Development of Calculation Methods III

Time: Thursday 15:45–18:00

Location: C 264

MM 62.1 Thu 15:45 C 264

**Phase transitions in radial distribution biased Molecular Dynamics simulations** — ●LARS DAMMANN<sup>1,2</sup>, PATRICK HUBER<sup>1,2</sup>, and ROBERT H. MEISSNER<sup>1,3</sup> — <sup>1</sup>Hamburg University of Technology, Hamburg, Germany — <sup>2</sup>Deutsches Elektronen-Synchrotron DESY, Hamburg, Germany — <sup>3</sup>Helmholtz-Zentrum Hereon, Geesthacht, Germany

Molecular Dynamics (MD) simulations are crucial for the study of phenomena at the atomic level. However, simulating the transition from liquid to solid states remains a challenge due to the energy barrier between phases, that makes the observation of spontaneous phase transitions in unbiased simulations computationally infeasible. Radial distribution functions (RDFs) provide valuable insights into atomic structures but are not unambiguous and therefore hard to interpret. I will present an algorithm that biases molecular dynamics simulations to reproduce a target RDF while minimizing the amount of information with which the original force field is biased. This is achieved through the application of the principle of maximum relative entropy. Following this principle, it is possible to use the structural information about the system contained in the RDF while conserving as much information as possible about the atomic interactions contained in the original force field. I will demonstrate that this strategy can be used to promote liquid-solid phase transitions in MD simulations. In the future, this method could potentially aid in the investigation of complex phase transitions, the analysis of metastable states and for the interpretation of structural information from experimental data.

MM 62.2 Thu 16:00 C 264

**Finite temperature electronic structure calculations for heavy element tetrahedral semiconductors using a dynamic tight-binding model** — ●SHAOMING ZHANG, MARTIN SCHWADE, and DAVID A. EGGER — Physics Department, TUM School of Natural Sciences, Technical University of Munich, Germany

Performing first-principles calculations of electronic properties at finite temperatures typically involves a substantial computational effort. Recent advancements in machine learning force-field molecular dynamics have expanded our ability to simulate large systems over longer timescales. But accurately computing the electronic structure in large, thermally disordered materials, particularly with non-negligible spin-orbit coupling effects when heavy elements are involved, remains difficult. To tackle these computational challenges, our approach adapts the tight-binding formalism for enhanced efficiency. Central to our method[1] is the use of hybrid-orbital basis functions, along with the incorporation of spin-orbit coupling. This yields a dynamic tight-binding model defined by a small set of parameters, optimized through density functional theory calculations. We show that our model is able to describe the temperature-dependent electronic properties of tetrahedral semiconductors with heavy elements, demonstrating alignment with first-principles results.

[1] M. Schwade, M. J. Schilcher, C. Reverón Baecker, M. Grumet, D. A. Egger, arXiv:2308.08897 [cond-mat.mtrl-sci] (2023)

MM 62.3 Thu 16:15 C 264

**Machine Learning Potentials for Multi-State Systems: Predicting Photoluminescence Spectra from Molecular Dynamics** — CHRISTOPHER LINDERÄLV<sup>1,2</sup>, ●NICKLAS ÖSTERBACKA<sup>2</sup>, JULIA WIKTOR<sup>2</sup>, and PAUL ERHART<sup>2</sup> — <sup>1</sup>University of Oslo — <sup>2</sup>Chalmers University of Technology

Divacancy defects in 4H-SiC show potential as single-photon emitters, which are important devices in quantum information technology. The photoluminescence spectra of these defects are crucial for this application, and accurate prediction of such spectra can aid in both understanding of the underlying defect physics and in device optimization.

We introduce a method for predicting photoluminescence spectra from the energy difference between the ground and excited state of defects sampled from molecular dynamics (MD) simulations, for which we employ machine learning potentials (MLPs) to extend the accessible length and time scales. Standard MLP construction approaches lead to exponential divergence in absorption and emission energies with increasing system size, however. To circumvent this, we introduce a method for the construction of MLPs capable of simultaneously describing both states.

We construct such a potential for a 4H-SiC divacancy defect and show that our MD-based approach yields emission spectra in good agreement with the generating function approach, which is the gold standard for such predictions from atomic structure. We also highlight the advantages of our MD-based method, emphasising synergies with MLPs.

MM 62.4 Thu 16:30 C 264

**Dynasor 2.0: From simulation to experiment through correlation functions** — ●ESMÉE BERGER, ERIK FRANSSON, FREDRIK ERIKSSON, ERIC LINDGREN, and PAUL ERHART — Department of Physics, Chalmers University of Technology, Gothenburg, Sweden

Using correlation functions, the full dynamics of a system can be accessed from atomistic simulations. This is particularly relevant for systems where perturbative approaches are not applicable, including liquids, metastable crystal structures, and anharmonic materials. The dynasor package is a flexible and efficient tool for calculating correlation functions, such as static and dynamic structure factors, both partial and total, as well as current correlation functions. Crucially, correlation functions provide a bridge to experiment, as weighing them with cross sections (or form factors) of, e.g., neutrons, X-rays or electrons, allows for the direct prediction of experimental spectra. Here, we demonstrate the new functionalities of dynasor 2.0, now featuring a Python interface, direct support for weighing, and the ability to handle trajectories from more molecular dynamics packages than before. The utility of dynasor is demonstrated through a wide range of examples, including static and dynamic structure factors, different experimental spectrum predictions, and phonon dispersions, for systems ranging from liquid Al to perovskite BZO.

MM 62.5 Thu 16:45 C 264

**Machine-learned interatomic potential for microstructure formation in Ni-rich NiAl systems** — ●ADAM FISHER<sup>1</sup>, JULIE B. STAUNTON<sup>1</sup>, HUAN WU<sup>2</sup>, and PETER BROMMER<sup>1</sup> — <sup>1</sup>University of Warwick, Coventry, UK — <sup>2</sup>TWI Ltd, Cambridge, UK

Precipitates in nickel-based superalloys form during heat treatment on a time scale inaccessible to direct molecular dynamics simulation, but can be explored using kinetic Monte Carlo (kMC) studies. This requires reliable values for the barrier energies separating distinct atomic configurations. We have previously described a method to find and validate barriers in this system and found that classical potentials such as embedded-atom method (EAM) fail to reproduce the correct ordering of barriers. Modern machine-learned interatomic potentials (MLIPs) have been shown to have an accuracy near that of density functional theory (DFT) at a fraction of the cost. In this work, we fit an atomic cluster expansion (ACE) MLIP for nickel-rich NiAl systems using ACE hyper-active learning (ACEHAL), training on a series of structures, from cubic unit cells of Ni and Ni<sub>3</sub>Al to large (> 100w atoms) NiAl solid solution cells. This is complemented by HAL runs on saddle point configurations, which improve the description of energy barriers. The MLIP barriers are then validated and compared to several traditional interatomic potentials.

MM 62.6 Thu 17:00 C 264

**Cross-Platform Hyperparameter Optimizer for Machine-Learning Potential Fitting** — ●DANIEL F. THOMAS DU TOIT, YUXING ZHOU, and VOLKER L. DERINGER — Department of Chemistry, University of Oxford, Oxford, UK

The use of machine learning interatomic potentials (MLIPs) to study materials has become increasingly popular in recent years. As the field has matured, multiple frameworks for MLIP fitting have been proposed. Here we present a Python package to optimize hyperparameters for MLIPs.

Our package, XPOT (“Cross-platform optimizer for machine learning interatomic potentials”), uses bespoke interfaces to MLIP fitting programs. XPOT enables users to use automated optimization to fit robust, accurate, and fast MLIPs. Using XPOT, we optimized hyperparameters for SNAP and ACE potentials based on existing training databases for Gaussian approximation potential (GAP) models, and demonstrated cost improvements while retaining high accuracy. We showcase the usefulness of the approach by creating optimized MLIPs across a diverse range of complex materials systems.

MM 62.7 Thu 17:15 C 264

**Warm Dense Hydrogen as a Benchmark for Machine-Learning Potentials** — BASTIAN JÄCKL<sup>1</sup>, THOMAS BISCHOFF<sup>1</sup>, and •MATTHIAS RUPP<sup>1,2</sup> — <sup>1</sup>University of Konstanz, Germany — <sup>2</sup>Luxembourg Institute of Science and Technology, Luxembourg

Machine-learning potentials (MLPs) are fast data-driven surrogate models of potential energy surfaces that can accelerate ab-initio dynamics simulations by several orders of magnitude. The performance of MLPs is commonly measured as the prediction error in energies and forces on data not used for training. While low prediction errors on a test set are necessary, they are not sufficient for good performance in dynamics simulations. The latter requires physically motivated performance measures obtained from running accelerated simulations. The adoption of such measures, however, has been limited by the effort and domain knowledge required to calculate and interpret them. To overcome this limitation, we present data and scripts to automatically quantify the performance of MLPs in dynamics simulations of hydrogen under pressure. For this challenging benchmark system, we provide geometries, energies, forces, and stresses, calculated at the density functional level of theory for different temperatures and mass densities. We also provide scripts to automatically calculate, quantitatively compare, and visualize pressures, diffusion coefficients, stable molecular fractions, and radial distribution functions. Employing our benchmark, we show that several state-of-the-art MLPs fail to reproduce a crucial liquid-liquid phase transition, despite low test set errors in energies and forces.

MM 62.8 Thu 17:30 C 264

**Ab initio Raman spectroscopy including temperature: Theory and application for GaN and BaZrS<sub>3</sub>** — •FLORIAN KNOOP<sup>1</sup>, NIMROD BENSALOM<sup>2</sup>, MATAN MENAHEM<sup>2</sup>, OMER YAFFE<sup>2</sup>, and OLLE HELLMAN<sup>2</sup> — <sup>1</sup>Linköping University, Sweden — <sup>2</sup>Weizmann Institute of Science, Israel

Ab initio simulations of Raman spectroscopy are often performed within the harmonic approximation, which is insufficient for describing realistic spectral properties of materials. Here we present a nu-

merically efficient scheme that couples ab initio simulations, equivariant neural network potentials, and self-consistent phonon theory in the temperature-dependent effective potentials (TDEP) framework to perform precise simulations of Raman response at finite temperature. The method is presented for wurtzite GaN in comparison to latest experimental results, with a focus on rigorous treatment of direction and polarization-orientation (PO) dependence in optically anisotropic solids with LO/TO splitting, which is often neglected in computational approaches. Subsequently we discuss BaZrS<sub>3</sub>, a highly polarizable yet lead-free chalcogenide Perovskite with strong optical response to showcase the approach for complex materials. Overall, we find excellent agreement with experiment for both materials and show how the polarization information can be used e.g. to discern crystallographic orientations. We conclude by discussing limits and systematic errors in the approach with implications for other materials simulations, as well as possible strategies to overcome some of them by tighter integration of experiment and theory.

MM 62.9 Thu 17:45 C 264

**An efficient method for estimating the dynamics of full polarizability tensor in *ab initio* molecular dynamics simulations** — •POUYA PARTOVI-AZAR — Martin Luther University of Halle-Wittenberg, Halle (Saale), Germany

An efficient method is presented to approximate the dynamics of individual polarizability tensor elements, for example during *ab initio* molecular dynamics simulations. The method is based on the calculation of quadrupole moment matrix of the position operator in maximally-localized Wannier functions representation. The presented method has a wide range of applications, particularly in vibrational spectroscopy simulations, such as (resonance) Raman, Raman optical activity, sum-frequency generation, etc. It is demonstrated that this method can lead to several hundred times speedup with respect to reference linear response calculations. The predictive power of the introduced method is tested in case of various molecules as well as depolarized Raman spectra of gaseous and liquid methanol, in all of which remarkable agreements with the reference spectra are observed.