

CPP 23: Modeling and Simulation of Soft Matter III

Time: Wednesday 9:30–11:15

Location: H 0107

CPP 23.1 Wed 9:30 H 0107

FAIR Data Management for Soft Matter Simulations using NOMAD — ●JOSEPH F. RUDZINSKI¹, JOSÉ M. PIZARRO¹, NATHAN DAELMAN¹, LUCA M. GHIRINGHELLI², and SILVANA BOTTI³ — ¹Institut für Physik und IRIS-Adlershof, Humboldt-Universität zu Berlin, Berlin — ²Department of Materials Science and Engineering, Friedrich-Alexander-Universität, Erlangen-Nürnberg — ³RC-FEMS and Faculty of Physics, Ruhr University Bochum, Bochum

NOMAD [nomad-lab.eu][1, 2] is an open-source data infrastructure for materials science data. NOMAD already supports an array of computational codes and techniques, with over 60 parsers that automatically extract essential (meta)data from the raw output of standard calculations. Traditionally, the NOMAD repository has focused on contributions from DFT calculations, accumulating over 12.5 million such entries. More recently, this framework has been expanded considerably, now supporting classical molecular dynamics simulations, as well as complex simulation workflows. In this context, a variety of new features have been implemented into NOMAD, including a schema for defining molecular topologies and system hierarchies. In this talk, I will introduce NOMAD in the context of soft matter simulations, demonstrating some basic functionalities and its potential for improving the data management standards within the classical simulation community through its adherence to the FAIR principles (Findability, Accessibility, Interoperability, Reusability).

[1] Scheidgen, M. *et al.*, *JOSS* **8**, 5388 (2023).

[2] Scheffler, M. *et al.*, *Nature* **604**, 635-642 (2022).

CPP 23.2 Wed 9:45 H 0107

Symmetry-adapted polarization learning for vibrational spectroscopy — ●DAVID WILKINS — Centre for Quantum Materials and Technologies, School of Mathematics and Physics, Queen's University Belfast, United Kingdom

The polarization of a material system is vital in modelling its response to light, and thus in predicting the results of spectroscopic experiments. However, there are several problems in these kinds of calculations: chief among them the requirement for electronic structure theory calculations and the fact that bulk polarizations are only defined modulo the so-called "quantum of polarization".

The solution of the first of these problems using machine-learning methods, some of which I have developed in the past few years, is by now in the mainstream of computational chemistry, but the second problem is still extant: the polarization is not a continuous function of the atomic positions. In this talk, I will compare and contrast two methods for overcoming this problem, based on localized dipole moments or on localized charges.

While the former method performs best for systems in which long-ranged electrostatics are not important, learning local charges is by far the best method when these effects are important (e.g. in electrolyte solutions). I show how both methods, where they are applicable, lead to very high-quality modelling of infrared and sum-frequency generation spectroscopy.

CPP 23.3 Wed 10:00 H 0107

Vibrational Spectroscopy from Machine Learning Molecular Dynamics by Accurately Representing the Atomic Polar Tensor — ●PHILIPP SCHIENBEIN — Department of Physics, Imperial College London, London, SW72AZ, United Kingdom — Department of Physics and Astronomy, University College London, London, WC1E 6BT, United Kingdom

Vibrational spectroscopy is a key technique to elucidate microscopic structure and dynamics. Without the aid of theoretical approaches, it is, however, often difficult to understand such spectra at a microscopic level. Ab initio molecular dynamics has repeatedly proved to be suitable for this purpose, but the computational cost can be daunting; in particular when electronic structure methods beyond GGA DFT are required. Here, a new route to calculate accurate IR spectra from machine learning molecular dynamics is presented, utilizing the atomic polar tensor. The latter can be trained a posteriori on existing molecular dynamics simulations using the E(3)-equivariant neural network e3nn and is a most fundamental physical observable. The introduced methodology is therefore general and transferable to a broad range of systems. Besides benchmarking the method against explicit ab initio

molecular dynamics, I will also present applications utilizing a atomic polar tensor neural network at the hybrid DFT level. These demonstrate that it has the potential to significantly contribute toward novel physical findings, especially where large-scale molecular dynamics simulations or expensive electronic structure calculations are required.

CPP 23.4 Wed 10:15 H 0107

Efficient construction of high-dimensional neural network potentials for the Strecker synthesis — ●ALEA MIAKO TOKITA^{1,2}, TIMOTHÉE DEVERGNE³, A MARCO SAITTA³, and JÖRG BEHLER^{1,2} — ¹Theoretische Chemie II, Ruhr-Universität Bochum, Germany — ²Research Center Chemical Sciences and Sustainability, Research Alliance Ruhr, Germany — ³Institut de Minéralogie, de Physique des Matériaux et de Cosmochimie, Sorbonne Université, Paris, France

High-Dimensional Neural Network Potentials (HDNNPs) provide potential energy surfaces with the accuracy of electronic structure calculations at strongly reduced computational costs. This enables extended molecular dynamics simulations of large systems such as organic molecules in solution. The construction of a HDNNP for such systems is not a trivial task since they have a vast configuration space which needs to be efficiently sampled. An example for such a complex system is the first step of the classic Strecker-cyanohydrin mechanism for glycine synthesis in water from formaldehyde and hydrogen cyanide. Here, we present a systematic construction of a HDNNP for this system as a showcase for molecular chemistry in solution.

CPP 23.5 Wed 10:30 H 0107

Machine learning of an implicit solvent for dynamic Monte Carlo simulations — ANKUSH CHECKERVARTY¹, JENS-UWE SOMMER^{1,2}, and ●MARCO WERNER¹ — ¹Institut Theorie der Polymere, Leibniz-Institut für Polymerforschung Dresden e.V., Dresden, Germany — ²Institut für Theoretische Physik, Technische Universität Dresden, Germany

We discuss an implicit solvent model based on an artificial neural network (NN) for dynamic Monte Carlo simulations, where the dynamics is implemented only via local particle displacements (elementary motions). The training data was obtained from coarse grained simulations using the bond fluctuation model with explicit solvent [1] for single homopolymers under variation of solvent quality. The NN based implicit solvent model takes into account only the information of the local environment of monomers in order to predict a distribution of possible acceptance rates [2] of an attempted elementary monomer move in the given configuration. We show that NN-based implicit solvent simulations reproduce the coil-globule transition, as well as dynamic properties such as the bond vector autocorrelation in time and mean square displacements of chain monomers as seen in the explicit solvent model. Furthermore, the learned NN parameters were transferable to a system of multiple homopolymers. [1] C. Jentzsch, M. Werner and J.-U. Sommer, *JCP* **138**, 094902 (2013). [2] A. Checkervarty, J.-U. Sommer and M. Werner, *JCP* **158**, 124904 (2023).

CPP 23.6 Wed 10:45 H 0107

Long-Range Descriptors in Atomistic Modeling beyond Electrostatics — ●PHILIP LOCHE, KEVIN KAZUKI HUGUENIN-DUMITTAN, and MICHELE CERIOTTI — Laboratory of Computational Science and Modeling, IMX, École Polytechnique Fédérale de Lausanne, Switzerland

Over the last decade, the use of machine learning based methods for modelling materials and molecules has developed rapidly. A key ingredient in most successful approaches has been the use of locality, also termed "nearsightedness" of electronic models. Local models truncate atomic interactions beyond some cutoff radius which allows the development of fast algorithms scaling linearly with the number of atoms. What is systematically neglected in these models are any kind of long-range interactions interactions, including electrostatics, dipole-dipole or van der Waals forces, due to the relatively high computational cost involved.

We present a general mathematical framework that is an extension of the long-range equivariant (LODE) descriptor, and can describe all long-range interactions with an inverse power law form. It can be used to predict scalar target properties (energies) but can also be used in models that need to predict gradients of those (forces). We illustrate

how this extension leads to significant improvements in both accuracy and computational cost.

CPP 23.7 Wed 11:00 H 0107

Encapsulation Of Pt-based Clusters In ZIF-8: Insights From First Principles Simulations —

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The reactivity of Pt-based catalysts dramatically depends on their size

and detailed atomic structure. Recently, atom-precise nanoparticles have been generated from molecular ligand-stabilized clusters via encapsulation within MOFs. Our work aims to rationalize the underlying encapsulation mechanism of the ligand-stabilized Pt-based clusters using first-principles simulations. Using Density Functional Theory calculations, we determine the interaction of ZIF-8 precursors with the CO ligands of the Pt-clusters and reveal chemically distinct sites of all the considered clusters, which favor the attraction of Zn²⁺ ions in solution. AIMD simulations of Pt-based clusters in an explicit methanol solvent model shed light on the competition between the attraction of Zn²⁺ and the Pt-clusters and the desorption of Zn²⁺ by solvation with methanol molecules, revealing the importance of bridging CO ligands to create a Zn-rich environment around the Pt-cluster and to guarantee the successful encapsulation.