DY 5: Machine Learning in Dynamics and Statistical Physics I

Time: Monday 9:30–13:00 Location: BH-N 243

DY 5.1 Mon 9:30 BH-N 243

Active-Learning Training of Accurate Machine-Learned Interatomic Potentials for Strongly Anharmonic Materials — •Kisung Kang, Thomas A. R. Purcell, Christian Carbogno, and Matthias Scheffler — The NOMAD Laboratory at the FHI of the Max-Planck-Gesellschaft and IRIS-Adlershof of the Humboldt-Universität zu Berlin

Machine-learned interatomic potentials (MLIP) promise numerically efficient access to long time and large length scales in molecular dynamics (MD) simulations while retaining an accuracy that is on par with *ab initio* MD. To this end, it is necessary that MLIPs provide reliable predictions even for geometries that are largely unaccounted for in the original training, e.g., for rare dynamic events. For instance, this is required for thermal transport calculations, for which the creation of defects and phase transition precursors can profoundly affect anharmonic effects [1]. To this end, we propose an active learning ($\mathcal{AL}$) technique, in which uncertainty estimates are used to iteratively incorporate strongly anharmonic configurations into the MLIP training. At variance with traditional approaches, this $\mathcal{AL}$ method is thereby able to accurately capture those (meta-stable) configurations that are only seldom explored, as demonstrated in the cases of CuI and $AgGaSe_2$. Eventually, we show that this approach results in improved training and data acquisition efficiency for strongly anharmonic materials, whereas virtually no overhead is needed for more harmonic compounds.

[1] F. Knoop, *et al.*, *Phys. Rev. Lett.* **130**, 236301 (2023).

DY 5.2 Mon 9:45 BH-N 243

Machine-learned Potentials for Vibrational Properties of Acene-based Molecular Crystals — •Shubham Sharma and Mariana Rossi — Max Planck Institute for the Structure and Dynamics of Matter

Machine-learning potentials (MLPs) have allowed the efficient modelling of complex atomistic systems with ab-initio accuracy. Normally, the construction of sufficiently large and diverse reference datasets, using first-principles calculations, is a bottleneck for training. Therefore, several active-learning strategies have been proposed, which aim to make the training more efficient, especially when used together with molecular-dynamics techniques [1]. In this work, we explore building protocols for training sets of high-dimensional neural network potentials (HDNNPs), targeting specifically an accurate description of the vibrational properties of weakly-bound condensed-phase systems. For that, we show how we can use and augment the committee-model framework within the i-PI code [2]. We show results for acene-based molecular crystals and discuss the advantages and limitations of different learning strategies to treat different crystal polymorphs, at various thermodynamic conditions. [1] C. Schran et. at., J. Chem. Phys. 153, 104105 (2020). [2] V. Kapil et. al., Comput. Phys. Commun. 236, 214 (2019).

DY 5.3 Mon 10:00 BH-N 243

Sampling free energies with deep generative models — •Maximilian Schebek[1], Michele Invernizzi[2], Frank Noé[1,2,3,4], and Jutta Rogal[5,1] — [1]Department of Physics, Freie Universität Berlin, 14195 Berlin, Germany — [2]Department of Mathematics and Computer Science, Freie Universität Berlin, 14195 Berlin, Germany — [3]Department of Chemistry, Rice University, 77005 Houston, Texas, USA — [4]AI4Science, Microsoft Research, 10178 Berlin, Germany — [5]Department of Chemistry, New York University, New York, NY 10003, USA

Evaluating free energy differences is a computationally demanding task, that requires a rigorous sampling of the phase space. Here, we train generative machine learning models based on normalizing flows to map between probability distributions of condensed phase systems at different thermodynamic conditions. Using the trained model, uncorrelated configurations can easily be generated. The model architecture incorporates permutation invariance and periodic boundary conditions, which improves convergence and enables the treatment of solid and liquid systems on the same footing. Training the flow model in a conditional way allows us to assess free energy differences over a wide range of temperatures and pressures, needed to evaluate the relative stability of different phases and reconstruct phase diagrams. The developed approach is applied to determine the coexistence line between liquid and solid as well as two different solid phases of a Lennard-Jones system. Our results are in excellent agreement with state-of-the-art methods, while the computational costs are significantly reduced.

DY 5.4 Mon 10:15 BH-N 243

Generative deep neural networks for topological defects and their microstructure reconstruction in two-dimensional spin systems — •Kyra Klos[1], Karin Everschor-Sitte[2], and Friederike Schmid[1] — [1]Institute of Physics, Johannes Gutenberg-University Mainz, Germany — [2]Faculty of Physics and Center for Nanointegration Duisburg- Essen (CENIDE), University of Duisburg-Essen, Germany

Topological defects are stable localized perturbations of an underlying ordering field characterized by their winding number. These microscopic structures have an intrinsic multi-scale character and can be described as point like quasi-particles in the macroscopic picture. Due to long range interaction patterns and their complex implications, like the phase transition induced by topological defects, the so called Berezinskii-Kosterlitz-Thouless phase transition [1] in the two-dimensional XY Model, simulations are of high interest but difficult to realize for large system sizes. To overcome this problem we develop a generative neural network tool based on an Wasserstein Generative Adversarial Network (WGAN) [2] bridging between the microscopic and macroscopic scale. Through physics induced constraints this WGAN tool provides the opportunity to construct physical realistic representative sets of spin configuration of magnetic materials from a given defect distribution and physics input parameters.

[1] J. M. Kosterlitz, Rev. Mod. Phys. 89 (2017)
[2] M. Arjovsky et al. arXiv:1701.07875v3 (2017)

DY 5.5 Mon 10:30 BH-N 243

Automated large-scale chemical breakdown simulations with machine learned reaction rates — •Joe Gilkes[1], Mark Storr[2], Reinhard J. Maurer[1], and Scott Habershon[1] — [1]University of Warwick, United Kingdom — [2]AWE plc, United Kingdom

Degradation of organic materials occurs over many years and involves rare reaction events over expansive networks of elementary processes.

Building such networks and propagating them in time to evaluate the mechanisms by which materials break down requires tackling combinatorially large chemical spaces and accurately calculating the rates at which thousands of reactions proceed. Such a process comes with a high computational cost, and kinetic simulation of such networks is made prohibitively difficult when variable experimental conditions must be considered, as reaction rates must also vary over the duration of a simulation.

We demonstrate a workflow for the automated construction and solution of chemical breakdown networks using machine-learned reaction rates and a discrete kinetic update approximation when propagating networks in time. This allows for rapid iterative exploration of chemical reaction space with arbitrary variable experimental conditions. We demonstrate this by constructing and simulating a detailed reaction network for the pyrolysis of ethane.

DY 5.6 Mon 10:45 BH-N 243

Machine learning of a density functional for anisotropic patchy particles — •Alessandro Simon[1,2], Martin Oettel[1], and Georg Martius[2] — [1]University of Tübingen, Tübingen, Germany — [2]Max Planck Institute for Intelligent Systems, Tübingen, Germany

Anisotropic patchy particles have become an archetypical statistical model system for associating fluids. Here we formulate an approach to the Kern-Frenkel model via classical density functional theory to describe the positionally and orientationally resolved equilibrium density distributions in flat wall geometries. After investigating the orientational structure of the fluid close to the wall, we bring the anisotropic part of the free energy into a kernel-form suitable for machine learning, through an expansion into orientational invariants and the proper incorporation of the tetrahedral single-particle symmetries. The mean-field kernel is constructed via machine learning on the basis of hard wall simulation data and a robust and numerically stable method that is able to condition neural networks on fixed points of their output. Results are compared to other well-known mean-field approximations,

which strongly underestimate orientational correlation. Finally, we propose more general machine-learning methods that are able to go beyond the mean-field approximation.

DY 5.7 Mon 11:00 BH-N 243

Systematic construction of velocity gradient models for turbulence — Maurizio Carbone[1,2], Vincent Peterhans[3,2], Alexander Ecker[4,2], and •Michael Wilczek[1,2] — [1]Theoretical Physics I, University of Bayreuth — [2]Max Planck Institute for Dynamics and Self-Organization, Göttingen — [3]Faculty of Physics, University of Göttingen — [4]Institute of Computer Science and Campus Institute Data Science, University of Göttingen

The dynamics and statistics of small-scale turbulence can be described in terms of velocity gradients, which makes them an appealing starting point for low-dimensional modeling approaches. Modeling velocity gradients in turbulence requires formulating closures for nonlocal pressure contributions and viscous effects based on modeling hypotheses about the small-scale dynamics and statistics of turbulence.

Here, we discuss an alternative, data-driven approach to derive a velocity gradient model that captures given velocity gradient statistics by construction. By analyzing the velocity gradient PDF equation, we distinguish contributions to the single-time statistics from those that impact temporal correlations. We then systematically construct a closure to reproduce a given velocity gradient PDF by design. We use the 'normalizing flow' machine learning approach to estimate the full eight-dimensional velocity gradient PDF from direct numerical simulation (DNS) data. Comparisons with Lagrangian velocity gradient data from DNS confirm that statistical features of small-scale turbulence statistics can be quantitatively captured by our low-dimensional dynamical model.

15 min. break

DY 5.8 Mon 11:30 BH-N 243

Quantum Phase Transitions with Neural Network Quantum States and a Lee-Yang Method — •Pascal M. Vecsei, Jose L. Lado, and Christian Flindt — Aalto University, Otakaari 1, 02150 Espoo, Finland

Predicting the phase diagram of interacting quantum many-body systems is a central problem in condensed matter physics and related fields. A variety of quantum many-body systems, ranging from unconventional superconductors to spin liquids, exhibit complex competing phases whose theoretical description has been the focus of intense efforts. Here, we show that neural network quantum states can be combined with a Lee-Yang method to investigate quantum phase transitions and predict the critical points of strongly correlated spin lattices [1,2]. Specifically, we implement our approach for quantum phase transitions in the transverse-field Ising model on different lattice geometries in one, two, and three dimensions. We show that the Lee-Yang method combined with neural network quantum states yields predictions of the critical field, which are consistent with large-scale quantum many-body methods. As such, our results provide a starting point for determining the phase diagram of more complex quantum many-body systems, including frustrated Heisenberg models.

[1] Pascal M. Vecsei, J. L. Lado, C. Flindt, Phys. Rev. B **106**, 054402 (2022)
[2] Pascal M. Vecsei, C. Flindt, J. L. Lado, Phys. Rev. Research **5**, 033116 (2023)

DY 5.9 Mon 11:45 BH-N 243

A Study of Quantum Non-Equilibrium Relations with Imaginary Path Integrals — •Jorge Castro, Eszter Pos, and Mariana Rossi — Max Planck Institute for the Structure and Dynamics of Matter, Hamburg, Germany

Several processes at the atomic scale occur in strongly out-of-equilibrium conditions. When such processes involve atomic motion at low temperatures or with light nuclei, it is expected that nuclear quantum effects play a pronounced role. However, theories that can rigorously treat quantum dynamics out of equilibrium are typically impossible to apply to explicit atomistic simulations of systems with many degrees of freedom. In this contribution, we study the protocols proposed in Ref.[1], where path-integral molecular dynamics simulations are used to evaluate quantum free energy differences through the Jarzynski and Crooks relations. We developed a code capable of generating non-equilibrium trajectories from path integral molecular dynamics. With this code, we investigated the efficiency and realm of validity of the work calculation employing driven path-integral molecular dynamics simulations. Our findings, based on 1D potentials, compare performance at various switching rates and varying coordinate shifts between the minima of starting and ending anharmonic potentials. We investigate the extend to which machine-learning methods targeting the inference of work distribution can speed up the statistical convergence of these protocols.

[1] R. van Zon, L. Hernández de la Peña, H. Peslherbe, and J. Schofield, *Phys. Rev. E* **78**, 041103 (2008)

DY 5.10 Mon 12:00 BH-N 243

Mean-field theories are simple for neural quantum states — •Fabian Ballar Trigueros, Tiago Mendes-Santos, and Markus Heyl — Universität Augsburg

The utilization of artificial neural networks for representing quantum many-body wave functions has garnered significant attention, however, quantifying state complexity within this neural quantum states framework remains elusive. In this study, we address this key open question from the complementary point of view: Which states are simple to represent with neural quantum states? Concretely, we show on a general level that ground states of mean-field theories with permutation symmetry only require a limited number of independent neural network parameters. We analytically establish that, in the thermodynamic limit, convergence to the ground state of the fully-connected transverse-field Ising model (TFIM), the mean-field Ising model, can be achieved with just one single parameter. Expanding our analysis, we explore the behavior of the 1-parameter ansatz under breaking of the permutation symmetry. For that purpose, we consider the TFIM with tunable long-range interactions, characterized by an interaction exponent α. We show analytically that the 1-parameter ansatz for the neural quantum state still accurately captures the ground state for a whole range of values for $0 \leq \alpha \leq 1$, implying a mean-field description of the model in this regime. We also comment on a potential method to identify and extract information from the neural network weight matrix that can give insight into the complexity of the state representation.

DY 5.11 Mon 12:15 BH-N 243

Tensor-network-based reinforcement learning for quantum many-body systems — •Giovanni Cemin — Max Planck Institute for the Physics of Complex Systems, Dresden, Germany

The exploration of quantum many-body systems is a widely pursued field. However, the exponential growth of the Hilbert space dimension makes it challenging to classically simulate quantum many-body systems and, consequently, to extract meaningful information. In this context, we present a novel framework for efficiently controlling quantum many-body systems based on reinforcement learning (RL). Our framework addresses the complexities of the quantum control problem by utilizing matrix product states to represent the many-body state within the trainable machine learning architecture of our RL agent. This novel methodology enables the control of systems on a scale beyond the capabilities of neural-network-only architectures, while retaining the advantages of deep learning, such as generalizability and robustness to noise. Notably, our research demonstrates the ability of RL agents to accurately handle previously unseen many-body states.

DY 5.12 Mon 12:30 BH-N 243

Derivative learning of tensorial quantities – Predicting infrared spectra from first principles — •Bernhard Schmiedmayer[1] and Georg Kresse[1,2] — [1]University of Vienna, Faculty of Physics and Center for Computational Materials Sciences, Vienna, Austria — [2]VASP Software GmbH, Vienna, Austria

In this talk, we present a novel computational framework that integrates machine learning with first-principles calculations to achieve accurate predictions of infrared spectra. Our method demonstrates its ability to reliably generate infrared spectra for complex systems at finite temperatures. The efficiency of the method is highlighted in challenging scenarios such as the analysis of water and the organic-inorganic halide perovskite MAPbI3. Our method is in good agreement with experimental results. A unique feature of our technique is the use of derivative learning, which is essential for obtaining accurate polarization data in bulk materials and facilitates the training of a symmetry-adapted machine learning framework. Using derivative learning, we are able to predict the anti-derivative with an accuracy of about 1%.

DY 5.13 Mon 12:45 BH-N 243

Stellar evolution forecasting with a timescale-adapted evolutionary coordinate and machine learning — •Kiril Maltsev and et al. — Heidelberger Institut für Theoretische Studien, Schloss-Wolfsbrunnenweg 35, 69118 Heidelberg

Many astrophysical applications require efficient yet reliable forecasts of stellar evolution tracks. One example is population synthesis, which generates forward predictions of models for statistical comparison with observations. The majority of state-of-the-art rapid population synthesis methods are based on approximate analytic fitting formulae to stellar evolution tracks that are computationally cheap to sample statistically over a continuous parameter range. The computational costs of running detailed stellar evolution codes over wide and densely sampled parameter grids are prohibitive, while stellar-age based interpolation in-between sparsely sampled grid points leads to intolerably large systematic prediction errors. In this work, we use supervised learning methods to construct an emulator of stellar evolution at a satisfactory trade-off between cost-efficiency and accuracy. We construct a timescale-adapted evolutionary coordinate and use it in a two-step interpolation scheme that traces the evolution of stars from zero age main sequence all the way to the end of core helium burning while covering a mass range from red dwarfs to very massive Wolf-Rayet stars. The feedforward neural network regression model that we train to predict stellar surface variables can make millions of predictions within tens of seconds on a 4-core CPU, with a mean prediction error that is an order of magnitude lower than typical observational uncertainties.