DY 15: Condensed-matter simulations augmented by advanced statistical methodologies (joint session DY/CPP)

Time: Monday 15:30–18:15   Location: HÜL 186

DY 15.1 Mon 15:30 HÜL 186

Funnel Hopping Monte Carlo: An efficient method to overcome broken ergodicity — •Jonas Alexander Finkler and Stefan Goedecker — University of Basel, Switzerland

Monte Carlo simulations are a powerful tool in the investigation of thermodynamic properties of atomic systems. In practice however sampling of the complete configuration space is often hindered by high energy barriers between different regions of configuration space which can make ergodic sampling completely infeasible within accessible simulation time.

We present a novel method called Funnel Hopping Monte Carlo (FHMC) to overcome broken ergodicity in Monte Carlo simulations. The method requires a priory knowledge of the funnel structure of the potential energy surface. This information does not have to be obtained from expensive Monte Carlo simulations but can be obtained very cheaply from a Minima Hopping [1] run. By approximating the Boltzmann distribution around local minima using Gaussian mixtures high quality Monte Carlo moves can be proposed that allow the simulation to directly jump between different funnels, circumventing the energy barriers in between.

The methods performance is demonstrated on the example of the 38 and 75 atom Lennard-Jones clusters which are well known for their double funnel energy landscapes that completely prevent ergodic sampling with conventional Monte Carlo simulations.

[1] S. Goedecker, J. Chem. Phys. 120, 9911 (2004)

DY 15.2 Mon 15:45 HÜL 186

Second-principles investigation of the electrocaloric properties of $PbTiO_3$ — •Monica Graf[1] and Jorge Iñiguez[1,2] — [1]Luxembourg Institute of Science and Technology - LIST — [2]University of Luxembourg

Electrocaloric effects attract increasing attention as they provide an ecologically-friendly alternative for solid-state cooling [1]. Here we investigate the electrocaloric response of prototype ferroelectric $PbTiO_3$, using a second-principles approach that has already proved its ability to reproduce qualitatively the properties of this compound in bulk [2] and when embedded in various heterostructures [3]. More precisely, we perform Monte Carlo simulations at finite temperature and under applied electric field to obtain the first- and second-order electrocaloric tensor of $PbTiO_3$, in a temperature range including the ferroelectric transition. The first-order electrocaloric tensor can be computed from the thermal fluctuations of polarization and energy, as obtained from simulations in absence of electric field, taking advantage of Maxwell's relations in the same way that is done in "indirect" experimental measurements of the electrocaloric effect. To access the second-order tensor, we simulate the material subject to electric fields of varying magnitude, parallel and perpendicular to the polarization. In this talk we will present and discuss our results.

References:
1. Nat Commun 9, 1827 (2018)
2. Phys. Rev. Lett. 112, 247603 (2014)
3. Nature 534, 524 (2016)

DY 15.3 Mon 16:00 HÜL 186

Exploring Chemical Reaction Space with Machine Learning — •Sina Stocker[1], Gábor Csányi[2], Karsten Reuter[1], and Johannes T. Margraf[1] — [1]Chair of Theoretical Chemistry, Technical University Munich, Germany — [2]Department of Engineering, University of Cambridge, United Kingdom

Reaction networks are essential tools for the analysis, visualization and understanding of chemical processes in such diverse fields as catalysis, combustion and the origin of life. For complex processes, the number of individual reaction steps in such a network is so large that an exhaustive first-principles calculation of all reaction energies and rates becomes prohibitively expensive. In this contribution, we use machine learning (ML) to accelerate the exploration of chemical reaction space, in analogy to the more established ML-based exploration of chemical space. To this end, we generated a new *reactive* reference database of open- and closed-shell organic molecules. This allows us to apply "chemical space" ML methods to predict the thermochemistry of reaction networks. We also develop explicitly "reaction space" based ML

approaches to directly predict reaction properties. The performance of these methods confirms the potential of ML for the high-throughput screening of large reaction networks.

DY 15.4 Mon 16:15 HÜL 186

Kernel-based machine learning for efficient molecular liquid simulations — •Christoph Scherer[1], René Scheid[1], Tristan Bereau[1,2], and Denis Andrienko[1] — [1]Max-Planck-Institut für Polymerforschung, Mainz, Germany — [2]University of Amsterdam, Netherlands

Most current force fields based on machine learning (ML) techniques result in high computational cost at every integration time step of an MD simulation. We describe a number of practical and computationally-efficient strategies to parametrize force fields for molecular liquids with kernel-based ML. We employ a particle decomposition ansatz to two- and three-body force fields and covariant kernels. Binning techniques allow to incorporate significantly more training data. Tabulation of the kernel predictions lead to MD simulations with the same computational cost than analytic three-body potentials. Results are presented for model molecular liquids: pairwise Lennard-Jones and three-body Stillinger-Weber systems, as well as an example from bottom-up coarse-graining of liquid water [1]. Many-body representations, decomposition, and kernel regression schemes are implemented in the open-source software package VOTCA [2].

[1] Scherer, Andrienko, PCCP, 20, 22387 (2018); [2] Rühle, Junghans, Lukyanov, Kremer, Andrienko, JCTC, 5, 3211 (2009)

DY 15.5 Mon 16:30 HÜL 186

Anharmonic phonons sampled from large scale molecular dynamics based on on-the-fly machine- learning force fields — •Jonathan Lahnsteiner and Menno Bokdam — Computational materials physics University of Vienna, Sensengasse 8/12 1090 Wien

The phonon spectrum for complex dynamic solids at elevated temperatures is often ill-described by the harmonic approximation. In this talk, I will present a molecular dynamics (MD) study of the Caesium Lead Bromide perovskite (CsPbBr3) in its orthorhombic and cubic phase. In the cubic phase, the Cs cation displays 'rattling' motion and (surrogate) structures are dynamically unstable, as indicated by imaginary phonon modes obtained in the harmonic approximation. For high accuracy and efficiency, the on-the-fly machine-learning force field (MLFF) method [1] is applied. This method generates a force-field with near first-principles accuracy and is trained to mimic the potential energy surface described by the SCAN density functional. Large scale microcanonical ensembles are generated with the MLFF and the finite-temperature dynamic structure factors are computed from the Fourier transform of the ionic density-density correlation functions. The intensities of the dynamic structure factor directly give insight in the phonon properties by taking into account phonon scattering events up to infinite order. These fully dynamic phonon spectra are compared to the phonons in the harmonic approximation. With this we are able to discuss the importance of anharmonic lattice vibrations to the stabilization of CsPbBr3 perovskite phases at finite temperature.

[1] Jinnouchi et.al. Phys.Rev.Let.10.1103/PhysRevLett.122.225701

15 min. break.

DY 15.6 Mon 17:00 HÜL 186

Edgy and Parallel – Efficient Equilibration of Anisotropic Hard Particulate Systems — •Marco Klement and Michael Engel — Institute for Multiscale Simulation, FAU Erlangen, Erlangen, Germany

Particle simulations are an important method to study the phase behavior of fluids and solids. A common task is structure prediction via thermal equilibration. Examples are crystallization or melting and the aging of glasses. Near the liquid-solid phase transition of hard spheres event driven molecular dynamics (EDMD) is known as most efficient. Correlated movement of particles in EDMD aids the equilibration. For Monte Carlo methods event-chains [1] introduce a correlation of particle updates. Particles move until the next collision. One of several collision rules determines the new particle to continue the chain and the new direction of the chain. The recently published

Newtonian event-chain (NEC) algorithm [2] is an advanced statistical methodology that assigns velocities to particles and performs collisions elastically. It is a close competitor in efficiency to EDMD. In this contribution we present the extension of NEC to polyhedral particles. We also discuss how parallelization strategies [3] can be adapted to this algorithm. Our implementation has been added to the software package HOOMD-blue. [4]

[1] Bernard et al., Phys. Rev. E 80, 056704 (2009)
[2] Klement et al., J. Chem. Phys. 150, 174108 (2019)
[3] Kampmann et al., J. Comput. Phys. 281, 864-875 (2015)
[4] http://glotzerlab.engin.umich.edu/hoomd-blue/

DY 15.7 Mon 17:15 HÜL 186

Machine-learning force fields trained on-the-fly with bayesian inference — Ryosuke Jinnouchi[1,2], Jonathan Lahnsteiner[1], Ferenc Karsai[3], Georg Kresse[1], and ●Menno Bokdam[1] — [1]University of Vienna, Vienna, Austria — [2]oyota Central R&D Labs, Inc., Aichi, Japan — [3]VASP Software GmbH, Vienna, Austria

Realistic finite temperature simulations of matter are a formidable challenge for first principles methods. Long simulation times and large length scales are required, demanding years of compute time. We present an on-the-fly machine learning scheme that generates force fields automatically during molecular dynamics simulations[1]. This opens up the required time and length scales, while retaining the distinctive chemical precision of first-principles methods and minimizing the need for human intervention. The method is widely applicable to multi-element complex systems and implemented in the VASP code. We demonstrate its predictive power on the entropy-driven phase transitions of hybrid perovskites (CH3NH3PbI3), which have never been accurately described in simulations.

[1] R. Jinnouchi et al., Phys. Rev. Lett. 122, 225701 (2019)

DY 15.8 Mon 17:30 HÜL 186

Learning effective collective variables for biasing via t-distributed stochastic neighbor embedding — ●Omar Valsson[1] and Jakub Rydzewski[2] — [1]Max Planck Institute for Polymer Research, Mainz, Germany — [2]Institute of Physics, Faculty of Physics, Astronomy and Informatics, Nicolaus Copernicus University, Torun, Poland

A common strategy to overcome the time scale problem of molecular dynamics (MD) simulations is to employ collective variable (CV) based enhanced sampling methods [1]. However, the efficiency of such approaches depends critically on the quality of the chosen CVs that must describe all the slow degrees of freedom. While physical and chemical intuition has proven generally successful in achieving this, there is a growing need for methods that can automatically find good CVs. An appealing option to accomplish this is to use ideas from the field of machine learning.

Here we show how dimensionality reduction via t-distributed stochastic neighbor embedding (t-SNE) [2] can be employed to learn effective CVs for biasing. We have implemented t-SNE directly in the PLUMED plug-in, which allows us to use it directly in numerous MD codes.

[1] O. Valsson, P. Tiwary, and M. Parrinello, Ann. Rev. Phys. Chem. 67, 159 (2016)

[2] L. van der Maaten and G. Hinton, J. Mach. Learn. Res. 9, 2579 (2008)

DY 15.9 Mon 17:45 HÜL 186

Variational autoencoders as a tool to learn collective variables from simulation snapshots — ●Miriam Klopotek and Martin Oettel — Institut für Angewandte Phsyik, Eberhard Karls Universität Tübingen, Tübingen, Germany

Variational autoencoders (VAEs) are powerful neural-network architectures capable of learning abstract representations of data (distributions of latent variables) in an unsupervised fashion. We apply a standard formulation of VAEs [1,2] to equilibrium configurations obtained by grand canonical Monte Carlo simulations and formulate a probabilistic model for the VAE which shows that the latent variables are collective variables, and their variances are generalized susceptibilities. Upon application to a lattice model with sticky rods which shows competing gas, liquid and nematic phases we find that the leading collective variables are akin to the two order parameters of the model. Furthermore, the collective variables define coarse-grained configurations. Increasing the number of latent variables leads to finer spatial resolution of the coarse-grained configurations and increasingly preciser physical observables obtained from them. Finally, we discover there is an optimal hyperparameter β in so-called β-VAEs [3] where the collective variables become disentangled with respect to structural correlation length-scales: These disentangled collective variables hence form a hierarchy of different levels-of-detail.

[1] Kingma, D. P. & Welling, M. (2013). arXiv:1312.6114. [2] Rezende, D. J., Mohamed, S., & Wierstra, D. (2014). arXiv:1401.4082. [3] Higgins, I. et al. (2017). ICLR, 2(5), 6.

DY 15.10 Mon 18:00 HÜL 186

Adversarial Reverse Mapping of Equilibrated Condensed-Phase Molecular Structures — ●Marc Stieffenhofer[1], Michael Wand[2], and Tristan Bereau[1] — [1]Max Planck Institute for Polymer Research, Ackermannweg 10, 55128 Mainz — [2]JGU Mainz, Institute of Computer Science, Staudingerweg 9, 55128 Mainz

Coarse-grained molecular dynamics simulations circumvent prohibitively long equilibration times by averaging out degrees of freedom. However to consistently link the scales a reverse mapping scheme is needed to reintroduce higher resolution details. Traditional schemes propose a rough coarse-to-fine mapping and rely on further energy minimization and molecular dynamics simulations to re-equilibrate the system. In this study we introduce DeepBackmap: A deep neural network based approach to predict equilibrated molecular structures without running molecular dynamics simulations. We use generative adversarial networks to learn the Boltzmann distribution from training data and realize reverse mapping by using the coarse-grained structure as a conditional input. Our method can be used for condensed-phase systems of arbitrary sizes. The model uses only local information and reconstructs the atomistic structure autoregressively. We test our method on syndiotactic polystyrene molecules and show that the model trained in a melt shows remarkable transferability to the crystalline phase. The learned local correlations appear to be temperature independent indicating a separation of the scales.