CPP 9: French-German Session: Simulation Methods and Modeling of Soft Matter I

Time: Monday 15:00–17:00 Location: ZEU/LICH

Invited Talk CPP 9.1 Mon 15:00 ZEU/LICH

Why Polymers Swell or Collapse: Molecular Insights into Cosolute Effects — •Nico van der Vegt — Department of Chemistry, Technical University of Darmstadt

Water-soluble amphiphilic polymers are widely used as stimuli-responsive materials. Their performance is governed by the coil-globule transition, characterised by the theta temperature, which can be tuned by adding small organic molecules (cosolutes). Although related phenomena - such as cononsolvency and salting-out - are well understood at a phenomenological level, their underlying molecular mechanisms remain unclear. This talk explores the solvation physics that determine how different classes of cosolutes influence the coil-globule transition of both generic hydrophobic polymers and realistic systems such as poly(N-isopropylacrylamide), using computer simulations. We identify two distinct molecular driving forces that regulate cosolute-induced hydrophobic interactions, leading to either polymer swelling or collapse. These driving forces depend sensitively on the relevant length scales and expose the interplay between microscopic and macroscopic solvation physics, underscoring the key role of solvent entropy in determining the properties of responsive soft-matter systems. [1] Comm. Chem. 3, 165 (2020), [2] PCCP 24, 2080-2085 (2022), [3] J. Chem. Phys. 160, 164902 (2024)

CPP 9.2 Mon 15:30 ZEU/LICH

THz Pump Pulse Drives Nonlinear H-bond Dynamics in Liquid Water — •Philipp Schienbein — Lehrstuhl für Theoretische Chemie II, Ruhr-Universität Bochum, 44780 Bochum, Germany — Research Center Chemical Sciences and Sustainability, Research Alliance Ruhr, 44780 Bochum, Germany

Light-matter interactions are central to processes in nature, chemical engineering, and fundamental research. Of particular interest is the frequency-dependent excitation of molecular systems by incident light. Here, we present machine-learning-accelerated molecular dynamics simulations of water subjected to an intense, monochromatic THz pump pulse, generating a total of 32 ns of trajectory data. The light pulse is explicitly modeled via a time-dependent external electric field, extending our previous framework for field-driven machine learning MD simulations, allowing us to observe the excitation in realtime. Our simulations reproduce key experimental observables, including transient dichroism and birefringence, and reveal that the pulse transiently modulates hydrogen-bond dynamics: H-bonds become markedly shorter-lived compared to unperturbed bulk water. This response is nonlinear, ruling out pure heating and resolving the experimental controversy. These results demonstrate that our machinery to incorporate electric fields in machine learning MD simulations can directly connect microscopic dynamics with measurable observables, further paving the way to controlling water dynamics - and potentially chemical reactivity - through tailored light pulses, a long-standing goal in chemistry.

CPP 9.3 Mon 15:45 ZEU/LICH

Fluctuating Solvent Fields and Vibrational Shifts Revealed by Machine Learned Force Fields — •Florian Brünig and Alexandre Tkatchenko — Department of Physics and Materials Science, University of Luxembourg, Luxembourg

Fluctuating solvent fields is one of the driving mechanism behind vibrational shifts observed in solvatochromic experiments [1]. Previously, experimental infrared spectra have been correlated with mean electric fields obtained from empirical force field molecular dynamics simulations [2]. However, vibrational dynamics of these force fields are insufficient to reproduce the experimental line shape properties.

Here, machine learned force fields (MLFFs) offer multiple opportunities: they are highly accurate, efficient, and can be interpretable if based on physical models. We explore the capabilities of modeling solvatochromism of vibrational probes in solvent and protein environments employing the SO3LR [3] MLFF. SO3LR combines the SO3krates architecture with pairwise analytical electrostatic and dispersion potentials in the long range, and was pretrained on a diverse data set of molecular complexes. We show how fluctuating solvent electrostatic and dispersion fields can be related to vibrational bands in a self consistent manner due to the versatility of MLFFs.

[1] Brünig, FN, Netz, RR, et al. JPCB 126, 1579 (2022).

[2] Weaver, JB, Kozuch, J, Kirsh, JM & Boxer, SG. JACS 144, 7562 (2022).

[3] Kabylda, A, Frank, JT, Müller, KR, Tkatchenko A, et al. JACS 147, 33723 (2025).

CPP 9.4 Mon 16:00 ZEU/LICH

Machine learning potentials for redox chemistry in solution — •Redouan El Haouari[1,2], Emir Kocer[1,2], and Jörg Behler[1,2] — [1]Theoretische Chemie II, Ruhr-Universität Bochum, Germany — [2]Research Center Chemical Sciences and Sustainability, Research Alliance Ruhr, Germany

Most Machine-Learning Potentials (MLPs) currently in use are local, which prevents the construction of accurate potentials for systems in which long-range charge transfer is important. One example is the distinction of different oxidation states of transition metal ions such as ferrous (Fe^{2+}) and ferric (Fe^{3+}) ions in aqueous solution. In this case the near-sightedness of local MLPs cannot account for distant counter ions or non-local charge transfer. We show that 4th-Generation High-Dimensional Neural Network Potentials (4G-HDNNPs), which employ atomic charges obtained from charge equilibration as globally-determined descriptors, do not suffer from this short-coming. For aqueous ferric and ferrous chloride, the model predicts iron oxidation states correctly matching the total number of chloride ions in the system demonstrating that physical knowledge about the system of interest remains essential to construct reliable MLPs.

CPP 9.5 Mon 16:15 ZEU/LICH

Bridging Accuracy and Sampling: Insights into dAMP Solvation from ML-potentials — •Laurie Stevens[1,2], Riccardo Martina[2], Alberta Ferrarini[2], and Marialore Sulpizi[1] — [1]Chair of Theoretical Physics of Electrified Liquid-Solid Interfaces, Faculty of Physics and Astronomy, Ruhr-Universität Bochum, Germany — [2]Department of Chemical Sciences, Università degli Studi di Padova, Italy

Although nucleotides are essential biomolecular building blocks, key aspects of their structure and interactions, in particular the steps enabling their assembly and polymerization, remain unclear. Here, we investigate the conformational behavior of deoxyadenosine monophosphate (dAMP) in solution using a combined computational-experimental approach. We developed a highly accurate machine-learning potential via active learning that thoroughly samples the free-energy landscape, capturing critical conformational degrees of freedom with near-chemical accuracy. This potential enables nanosecond-scale simulations of water and nucleotides at a level previously inaccessible with hybrid DFT methods. We find that, in solution, dAMP accesses both anti and syn glycosidic conformations. We identify the anti and high anti conformers as the most stable ones, we show that they underlie the intensity variations observed in mid-IR experimental spectra.

CPP 9.6 Mon 16:30 ZEU/LICH

Fine-Tuning Unifies Foundational Machine-learned Interatomic Potential Architectures at ab intio Accuracy — •Christian Dressler, Jonas Hänseroth, Nawaz Qaisrani, and Aaron Flötotto — TU Ilmenau, Institute of Physics, Germany

Machine-learned force fields (MLFFs) have enabled ab initio quality molecular dynamics at speedups of several orders of magnitude. Foundation-model MLFFs, trained on extremely large and diverse datasets, aim to provide broadly transferable force predictions, yet their accuracy remains system-dependent. We investigate five state-of-the-art foundation models (MACE, GRACE, ORB, MATTERSIM, SEVENNET) and fine-tune them for seven chemically diverse systems.[1] Our results show that foundation models provide a useful baseline but still deviate significantly from AIMD. Fine-tuning consistently improves accuracy across all frameworks and yields predictions that closely match AIMD reference data, while also reducing performance differences between models. The final accuracy is largely independent of the underlying architecture. These findings suggest that, after fine-tuning, model choice is no longer the main bottleneck. Prioritizing inference speed and computational efficiency may therefore offer the greatest benefit for accelerating the practical use of ML-based interatomic potentials in materials modeling.

[1] Hänseroth, ... , Dreßler, Fine-Tuning Unifies Foundational

Machine-Learned Interatomic Potential Architectures at ab initio Accuracy, https://doi.org/10.48550/arXiv.2511.05337

CPP 9.7 Mon 16:45 ZEU/LICH

NMR crystallography at finite temperatures — •Matthias Kellner and Michele Ceriotti — Laboratory of Computational Science and Modeling, Institut des Matériaux, École Polytechnique Fédérale de Lausanne, 1015 Lausanne, Switzerland

In this talk I will present our latest developments in machine-learning models for predicting NMR chemical shieldings in organic molecular solids. NMR shielding-driven structure determination protocols rely on comparing experimental solid-state NMR shifts with simulations whose accuracy is often limited by computational cost. We address these limitations by combining ensembles of transformer-based shielding predictors with molecular dynamics simulations using a novel, broadly applicable machine-learning interatomic potential. This framework systematically improves agreement with experiment across many benchmark systems, removes the need for empirical corrections and makes it applicable also to disordered and amorphous materials. I will conclude by showing applications of this framework to the structure determination of amorphous active pharmaceutical ingredients.