

CPP 2: Modeling and Simulation of Soft Matter I

Time: Monday 9:30–13:00

Location: MER 02

Invited Talk

CPP 2.1 Mon 9:30 MER 02

Molecular Theories meet Explainable Machine Learning – Novel Concepts for Advanced Drug Formulations — ●JENS SMIAŁEK — Institute for Computational Physics, University of Stuttgart, Germany — Boehringer Ingelheim Pharma GmbH & Co. KG, Biberach (Riss), Germany

Pharmaceutical processes and drug formulations are based on a variety of molecular mechanisms and principles. Significant advances have been made in recent years in terms of basic understanding. Despite these advances, a large number of mechanisms are not yet fully understood, which would be of great advantage, especially for the optimization of development processes as well as the quality and stability of drugs. In this talk I will present fundamental molecular theories of electronic interactions in solutions. Due to the large number of components and the complexity of the interactions, these theories can only be used for a qualitative understanding. However, by combining it with explainable machine learning, the basic molecular mechanisms can be identified and the corresponding thermodynamic properties of the solutions can be predicted. I will present some examples and discuss the underlying benefits and additional challenges for future developments.

CPP 2.2 Mon 10:00 MER 02

Revealing the relation between structure and dynamics using unsupervised machine learning — ●MOUMITA MAITI¹, ANAND NARAYANAN KRISHNAMOORTHY², YOUSSEF MABROUK², DIDDO DIDDENS², and ANDREAS HEUER¹ — ¹Institute of Physical Chemistry, University of Münster, Corrensstrasse 28/30, 48149 Münster, Germany — ²Forschungszentrum Jülich GmbH, Helmholtz-Institute Münster (IEK-12), Corrensstraße 46, 48149 Münster, Germany

Molecular dynamics trajectories of Li-ions in electrolytes with different anions are analysed using an unsupervised machine learning algorithm to understand the relation between structure and dynamics. The trajectories are generated using a polarizable force-field model instead of the standard OPLS force-field, thereby generating excellent agreement of structural and dynamical properties with experiment. The high-dimensional feature space is constructed from the distances and angles between Li-ions and its neighbours, reflecting the structural information of local environments. It can be reasonably reduced to 2 dimensions using the dimension reduction algorithm umap. This is performed for a large number of Li-ions. After the mapping distinct clusters can be identified. Although they are based on structural properties, they also reflect different dynamical properties with respect to diffusivity and ion-correlations. We finally show how the total ionic conductivity can be expressed in terms of the structural information of the first solvation shell.

CPP 2.3 Mon 10:15 MER 02

Data reweighting in metadynamics simulations — TIMO SCHÄFER¹ and ●GIOVANNI SETTANNI^{1,2} — ¹Department of Physics, Johannes-Gutenberg University Mainz, Germany — ²Faculty of Physics and Astronomy, Ruhr University Bochum, Germany

The data collected along a metadynamics simulation can be used to recover information about the underlying unbiased system by means of a reweighting procedure. We analyze the behavior of several reweighting techniques in terms of the quality of the reconstruction of the underlying unbiased free energy landscape in the early stages of the simulation and propose a simple reweighting scheme that we relate to the other techniques. We show[1] that the free energy landscape reconstructed from reweighted data can be more accurate than the negative bias potential depending on the reweighting technique, the stage of the simulation, and the adoption of well-tempered or standard metadynamics. While none of the tested reweighting techniques from the literature provides the most accurate results in all the analyzed situations, the one proposed here, in addition to helping simplifying the reweighting procedure, converges quickly and precisely to the underlying free energy surface in all the considered cases, thus allowing for an efficient use of limited simulation data.

[1]Schäfer TM, Settanni G. *J Chem Theory Comput.* 2020 Apr 14;16(4):2042-2052. doi: 10.1021/acs.jctc.9b0086

CPP 2.4 Mon 10:30 MER 02

findR: An automatized workflow from molecular dynamic simulation to quantum chemical methods for reaction discovery — ●GUNNAR SCHMITZ¹, ÖZLEM YÖNDER², VANESSA ANGENENT³, CHRISTOF HÄTTIG², and ROCHUS SCHMID³ — ¹Ruhr-Universität Bochum, Lehrstuhl für Theoretische Chemie II, Bochum, Germany, 44801 Bochum — ²Ruhr-Universität Bochum, Lehrstuhl für Theoretische Chemie I, Bochum, Germany, 44801 Bochum — ³Ruhr-Universität Bochum, Lehrstuhl für Anorganische Chemie II, Bochum, Germany, 44801 Bochum

Computer-guided reaction discovery is still an open issue, which will be vital for applications in science and industry. For this purpose, we present an automatized workflow that, starting from molecular dynamics simulations (MD), identifies reaction events based on graph theory, filters them, and prepares them for accurate quantum chemical calculations using e.g. Density Functional Theory (DFT) or Coupled Cluster methods. The capabilities of the automatized workflow are demonstrated by the example of simulations for the combustion of polycyclic aromatic hydrocarbons. A limiting factor of the reaction sampling by means of MD simulations is the required simulation time to observe reactions. In order to enhance this process, we compare different acceleration techniques like metadynamics and a nano reactor setup, in which a spherical confinement is combined with periodically applied forces directed to the simulation box center. We demonstrate that our workflow can be used to identify reactions without prior chemical knowledge and can give new insights into the combustion process.

CPP 2.5 Mon 10:45 MER 02

Data-driven analysis and prediction of the performance of cleansing solutions — ●TAKAHIRO YOKOYAMA¹, HIDEKI MIWAKE², RYOICHI NAKATAKE², NORIYOSHI ARAI¹, and ARASH NIKOUBASHMAN^{1,3} — ¹Department of Mechanical Engineering, Keio University, Yokohama, Japan — ²Research Institute, Fancl Corporation, Yokohama, Japan — ³Institute of Physics, JGU Mainz, Germany

Cleansing solutions are complex multi-component liquids, which are commonly used to wash excess sebum, dirt, and make-up cosmetics directly from the skin. Their formulations typically contain a large variety of ingredients, including surfactants, viscosity modifiers, and polyols to meet the desired product requirements, such as high skin compatibility, moisture retention, and long shelf life. The large number of different components makes it, however, challenging to understand and predict the physical properties of such cleansing solutions, since the majority of established theories have been developed for pure systems. Machine learning (ML) methods are promising methods to solve these kinds of problems, as they can reveal non-trivial trends and interactions between the various components. In this work, we predict the cleansing performance of a large number of formulations using various ML methods, such as random forest regression and (recurrent) neural networks. Although this work focuses primarily on cleansing solutions, the applied methodology should be transferable to predict the physical properties of other multi-component liquids as well.

CPP 2.6 Mon 11:00 MER 02

Multiscale Simulation Framework for the electromechanical behavior of PEDOT:PSS — STEFFEN KAMPMANN¹, ALEXANDER CROY², ●AREZOO DIANAT¹, and GIANAURELIO CUNIBERTI¹ — ¹Chair of Materials Science and Nanotechnology, Faculty of Mechanical Science and Engineering, TU Dresden, Dresden, Germany — ²Chair of Theoretical Chemistry, Faculty of Chemistry and Earth Sciences, Friedrich Schiller University Jena, Jena, Germany

Functionally mechanically resilient polymer films, such as poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEDOT:PSS), play an important role for strain gauges or organic light-emitting diode displays [1-3]. Typically, the respective material behavior results from an interplay of mechanisms across multiple scales. The modeling and simulation workflow presented here enables the generation of disordered polymers and the linking of their mechanical and their electronic properties from the atomistic to the microscopic scale. In particular, we focus on the relationship between deformation and conductivity by combining density functional tight binding calculations, molecular dynamics simulations, and finite element calculations. The in-situ processing, evaluation as well as the exchange of the generated data across the simulation methods is performed using a Python framework which

provides a computationally efficient assessment of material properties at different scales. Exemplarily, we present results for a strain gauge based on PEDOT:PSS. [1] R. Luo, et al., *Progress in Organic Coatings*, 162, 106593 (2022) [2] M. Cinquino et al., *JS: AMD*, 7, 1,100394 (2022) [3] L. Liu, et al., *Organic Electronics*, 89, 106047 (2021)

15 min. break

CPP 2.7 Mon 11:30 MER 02

Differentiable simulation for Solar Power Plants and beyond — ●STEFAN KESSELHEIM¹, MAX PARGMANN², and JAN EBERT¹ — ¹Forschungszentrum Jülich — ²German Aerospace Center (DLR)

In Solar Power Plants, temperatures sufficient for chemical processes or the generation of electrical power are created by reflecting sunlight with thousands of mirrors ("heliostats") to a surface ("the receiver"). In operation, the temperature distribution on the receiver is critical for the performance and must be optimized. The heliostats are never perfectly flat as due to budget constraints, the construction is not optimal. We have devised a method to infer the heliostat surface from the reflection of the sun. The technique is based on an implementation of a simulation in PyTorch, where the automatic differentiation engine is used to optimize the surface. The surface is modeled as by a Non-Uniform Rational B-Spline (NURBS) and the NURBS parameters are subject to optimization. Furthermore we employ a regularization technique to mitigate the appearing challenge of ambiguous solutions. Our approach makes efficient use of GPUs based on PyTorch's linear algebra engine. We believe our approach poses an interesting example of a fruitful interaction of techniques originating from Machine Learning and simulation, that can serve as an exciting example how to integrate simulation and experimental data, for example in biomolecular simulation.

CPP 2.8 Mon 11:45 MER 02

Simulation-based inference of single-molecule force-spectroscopy — ●ROBERTO COVINO^{1,2}, LARS DINGELDEIN¹, and PILAR COSSIO³ — ¹Frankfurt Institute for Advanced Studies, 60438 Frankfurt am Main, Germany — ²International Max Planck Research School on Cellular Biophysics, 60438 Frankfurt am Main, Germany — ³Center for Computational Mathematics and Biology, Flatiron Institute, 10010 New York, United States

Single-molecule force spectroscopy (smFS) is a powerful approach to studying molecular self-organization. However, the coupling of the molecule with the ever-present experimental device introduces artifacts, that complicates the interpretation of these experiments. Performing statistical inference to learn hidden molecular properties is challenging because these measurements produce non-Markovian time series, and even minimal models lead to intractable likelihoods. To overcome these challenges, we developed a computational framework built on novel statistical methods called simulation-based inference (SBI). SBI enabled us to directly estimate the Bayesian posterior, and extract reduced quantitative models from smFS, by encoding a mechanistic model into a simulator in combination with probabilistic deep learning. Using synthetic data, we could systematically disentangle the measurement of hidden molecular properties from experimental artifacts. The integration of physical models with machine-learning density estimation is general, transparent, easy to use, and broadly applicable to other types of biophysical experiments.

CPP 2.9 Mon 12:00 MER 02

Coarse-grained modelling of liquid-liquid and liquid-gas interfaces — ●JAKOB FILSER¹, HARALD OBERHOFER², CHRISTOPH SCHEURER¹, and KARSTEN REUTER¹ — ¹Fritz-Haber-Institut der MPG, Berlin, Germany — ²Chair for Theoretical Physics VII, University of Bayreuth

Modelling of dielectric interfaces remains a central challenge in computational chemistry. We present a new method to incorporate solvation effects into density-functional theory calculations of organic adsorbates at liquid-liquid and liquid-gas interfaces.

Simulating a large number of solvent molecules explicitly at first-principles level is generally not computationally tractable. We therefore resort to an implicit solvation approach, treating the solvent as a structureless dielectric continuum. Specifically, we advance the multipole-expansion method, in which we model the interface as the boundary of two semi-infinite media with different permittivity. Gauging the limiting behaviour for moving the solute from one bulk medium into the other is straightforward by referencing experimental transfer

free energies. Complementary force-field level MD simulations with explicit solvent molecules are used as a reference for the behaviour at the interface. These calculations yield thermal distributions at atomic scale resolution, allowing for a fine assessment of the role of the solvent structure in the adsorption. In our implicit method, we employ a model for the transition of free energy terms, such as the surface tension, between the two media, aimed at reproducing the above mentioned thermal distributions extracted from an atomistic solvent model.

CPP 2.10 Mon 12:15 MER 02

Geometrical frustration causes self-limiting assembly in systems of bipods — ●ASWATHY MUTTATHUKATTIL and MICHAEL ENGEL — Institute for Multiscale Simulation, IZNF, Friedrich-Alexander Universität Erlangen-Nürnberg, 91058 Erlangen, Germany

Geometric frustration emerges in self-assembly if local interactions between building blocks are incompatible to form uniform (bulk) materials. Examples for geometrically frustrated assemblies with self-limiting size include protein filament bundles, twisted molecular crystals, bent core liquid-crystals and viral capsids. Despite of significant efforts to develop a general theory on geometrical frustration, it remains as a challenge to use it as a design principle to engineer finite-size equilibrium assemblies in soft matter. In this study, we control geometrical frustration deliberately and systematically in system of bipods to demonstrate self-limiting crystallization. Our minimal computational model consists of a central sphere that connect to two attractive rigid rods diametrically via a flexible hinge. Tuning the flexibility of the hinge and rod attraction, anisotropic fibrillar assemblies are observed. The fibrils do not grow wider than a fixed limit, which can be controlled by the misfit introduced by the radius of central sphere. Our model bipods can be realized in experiments as nanoparticles with tethered polymer bundles, partially unfolded polymer globules (including biopolymers like proteins) and organic molecules.

CPP 2.11 Mon 12:30 MER 02

Pair-Reaction Dynamics in Water: Competition of Memory, Potential-Shape and Inertial Effects — ●FLORIAN BRÜNIG, JAN DALDROP, and ROLAND NETZ — Freie Universität Berlin, Department of Physics, 14195 Berlin, Germany

When described by a one-dimensional reaction coordinate, pair-reaction rates in a solvent depend, in addition to the potential barrier height and the friction coefficient, on the potential shape, the effective mass and the friction relaxation spectrum, but a rate theory that accurately accounts for all these effects does not exist. We show how to extract all parameters of the generalized Langevin equation (GLE) and in particular the friction memory function from molecular dynamics (MD) simulations of two prototypical pair reactions in water, the dissociation of NaCl and of two methane molecules. Simulations of the GLE by Markovian embedding techniques accurately reproduce the pair-reaction kinetics from MD simulations without any fitting parameters, which confirms the accuracy of the approximative form of the GLE and of the parameter extraction techniques. By modification of the GLE parameters, we investigate the relative importance of memory, mass and potential-shape effects. Neglect of memory slows down NaCl and methane dissociation by roughly a factor of 2, neglect of mass accelerates reactions by a similar factor, the harmonic approximation of the potential shape gives rise to slight acceleration. This error cancellation explains why Kramers' theory, which neglects memory effects and treats the potential shape harmonically, describes reaction rates better than more sophisticated theories.

CPP 2.12 Mon 12:45 MER 02

The Crucial Role of Solvation Forces in the Steric Stabilization of Nanoplatelets — ●NANNING PETERSEN, MARTIN GIRARD, ANDREAS RIEDINGER, and OMAR VALSSON — Max Planck Institute for Polymer Research, Ackermannweg 10, D-55128 Mainz

The precipitation of ligand coated cadmium selenide nanoplatelets is linked to the formation of nanoplatelet stacks. The exact nature of nanoplatelets' interaction is an open question, as the van der Waals attraction is too weak to be the cause of stack formation. CdSe nanoplatelets combine a large facet to particle size ratio, and a very dense ligand shell. Both features are in favor of interactions in the form of solvation forces.

We use coarse-grained molecular dynamics simulations of ligand coated nanoplatelets in different alkane solvents to investigate the role of solvation forces in nanoplatelet interactions [1]. We demonstrate that solvation forces resulting from solvent layering are sufficiently strong to stabilize nanoplatelet stacks. We examine the dependence

of solvation forces on the nanoplatelets' ligand shell, size, and other parameters. In particular, we demonstrate that for sufficiently large nanoplatelets, solvation forces are proportional to the interacting facet area. We show that their strength is intrinsically tied to the softness of the ligand shell, depends on the isomer of the alkane solvent, and is

increasing with the alkane molecule length.

[1] N. Petersen, M. Girard, A. Riedinger, and O. Valsson, ChemRxiv, doi:10.26434/chemrxiv-2022-mw1cs-v3 (2022), accepted for publication in Nano Letters