CPP 42: Focus: Multiscale Simulations for Soft Matter: The Challenge of Dynamics (joint session CPP/DY, organized by CPP)

Organizers: Tristian Bereau , Joseph F. Rudzinski, Kurt Kremer (all MPI Polymerforschung, Mainz)

Multiscale simulations have gained increasing interest in soft matter, due to their ability to better reach the many underlying length and timescales spanning these systems. While significant development of coarsegraining methodologies which aim to accurately describe static equilibrium properties has led to a variety of successful applications, obtaining an accurate description of dynamics remains challenging. This focus session aims at bringing together researchers making various contributions to improving the description of dynamics in coarsegrained models, whether providing a theoretical background, improving parametrization protocols, or studying the limits of existing models.

Time: Wednesday 15:00–18:15   Location: H51

CPP 42.1   Wed 15:00   H51
Reactive molecular dynamics simulations of NaOH solutions — •Matti Hellström and Jörg Behler — Lehrstuhl für Theoretische Chemie, Ruhr-Universität Bochum, 44780 Bochum, Germany

Sodium hydroxide (NaOH) is soluble in water up to very high concentrations and has many applications in chemical industry. Still, surprisingly little is known about the structural and dynamical properties of its aqueous solutions. Using a high-dimensional neural network potential for NaOH(aq) based on dispersion corrected density-functional theory calculations, we have performed large-scale molecular dynamics simulations with close to ab initio quality. First results on many different phenomena like ligand exchange, ion clustering and proton transfer as well as their dependence on concentration are presented and discussed.

CPP 42.2   Wed 15:15   H51
Benchmark of a Reparametrized OPLS Force Field for Chlorinated Hydrocarbons Using Molecular Dynamics Simulations — •Zhu Liu, Jakob Timmermann, Karsten Reuter, and Christoph Scheurer — Theoretical Chemistry, Technische Universität München

The dielectric permittivity sensitively reflects the unique microscopic characteristics of a material [1]. To our knowledge no flexible non-polarizable force field for chlorinated hydrocarbons reliably reproduces this important physical property. We address this situation by reparametrizing an OPLS-AA (Optimized Parameters for Liquid Simulations, All-Atom) force field [2] to optimize the description of dynamic and dielectric properties. Specifically, we refitted the torsional potential energy profile and the atomic partial charges for several molecules from this class to match quantum chemical data. We find the new parametrization to yield a range of physical properties (especially the dielectric constant, isothermal compressibility, and thermal expansion coefficient) in excellent agreement with experimental data for a range of chlorinated hydrocarbon solvents, in addition to getting the surface tension and heat capacity at least as well as the original OPLS-AA model.

[1] C.J.F. Böttcher, Theory of Electric Polarization, Vol. I: Dielectrics in Static Fields, Elsevier B.V., Amsterdam (1973).
[2] W.L. Jorgensen *et al.*, Proc. Natl. Acad. Sci. **102**, 6665 (2005).

CPP 42.3   Wed 15:30   H51
Thermodynamics of polymer nematics: particle-based simulations versus mean-field calculations — •Cristina Greco, Kurt Kremer, and Kostas Daoulas — Max Planck Institute for Polymer Research, Mainz, Germany

Conjugated liquid crystalline (LC) polymers have emerged as promising materials for organic electronics. Predicting their large-scale morphology requires the implementation of mesoscale models. The statistical mechanics of such models is often addressed using mean-field (MF) approaches. These have been very successful in polymer physics and are attractive because of their computational efficiency and straightforward description of thermodynamic properties. For LC polymers, however, deviations between MF predictions and experimental results have been reported, e.g. for the molecular weight dependence of the nematic-isotropic transition. The question then arises: are the discrepancies due to the MF approximation or do they reveal fundamental limitations of the model, e.g. transferability?

To address this issue, we consider a model representing polymer nematics as worm-like chains interacting through soft anisotropic potentials and investigate its phase behaviour by (i) MF calculations based on partial enumeration of conformations, (ii) Monte Carlo simulations. In the latter, accurate free energies are obtained via a special thermodynamic integration scheme which avoids thermodynamic singularities. By comparing the results of the two methods, we can evaluate the effect of fluctuations and local correlations on the macroscopic behaviour.

CPP 42.4   Wed 15:45   H51
Comparing atomistic and coarse-grained simulations of P3HT — •Jonathan Gross, Momchil Ivanov, and Wolfhard Janke — ITP Uni Leipzig

Poly(3-hexylthiophene) (P3HT) is a key material used in organic photovoltaics (OPVs). In this study we assess the validity of two coarse-grained models of P3HT. We compare coarse-grained Monte Carlo simulations to fully atomistic molecular dynamics simulations. Structural properties of single polymer chains of short to medium lengths are compared between the three representations.

Invited Talk   CPP 42.5   Wed 16:00   H51
A coarse-grained model for DNA: dynamics of self-assembling biological systems and nanostructures. — •Ard Louis — Theoretical Physics, University of Oford

DNA is a particularly promising candidate for large-scale self-assembly because the specific binding of DNA bases can be accurately designed to build dynamic structures on the nanoscale with atomic precision. Inside the cell, DNA not only stores information in a digital code, but controls its readout by subtle modulation of its dynamic properties.

To study processes on these time and length-scales we employ oxDNA[1], a nucleotide level coarse-grained model that can model dynamical DNA structures with thousands of nucleotides. We are able to accurately reproduce the dynamics of exchange reactions, a key component of dynamic DNA nanotechnology, and also make detailed predictions for the hybridization dynamics of DNA duplexes. Larger structures such as a DNA nanobot can also be studied. For the dynamics of large-scale addressable assemblies, we develop a new multi-scale technique. As is the case for many other coarse-grained systems, correctly interpreting dynamical results is subtle[2].

[1] http://dna.phyiscs.ox.ac.uk [2] J.T. Padding and A.A. Louis, Phys, E 74, 031402 (2006)

15 min. break

CPP 42.6   Wed 16:45   H51
Cycle representatives for the coarse-graining of systems driven into a non-equilibrium steady state — •Fabian Knoch and Thomas Speck — Institut für Physik, Johannes Gutenberg-Universität Mainz, Staudingerweg 7-9, 55128 Mainz, Germany

A major current challenge in statistical mechanics poses the systematic construction of coarse-grained Markov State Models [1] that are dynamically consistent, and, moreover, might be used for systems driven out of thermal equilibrium. Here we present a novel prescription that extends the Markov state modeling approach to driven systems violating detailed-balance [2]. In more detail, we decompose a given Markov State Model in cycles and introduce the concept of cycle representatives, which stand for many cycles that share similar properties. The coarse-graining involves the renormalization of transition rates that preserves the entropy production of the original Markov State Model. We illustrate our new methodology by an intuitive example: A particle trapped in a 2-dimensional double well potential and driven by a non-conservative force.

[1] Prinz, J.-H., Wu, H., Sarich, M., Keller, B., Senne, M., Held, M.,

Chodera, J. D., Schütte, C. and Noe, F. Markov models of molecular kinetics: Generation and validation. JCP 134(17), 2011
[2] Knoch, F. and Speck, T. Cycle representatives for the coarse-graining of systems driven into a non-equilibrium steady state. New Journal of Physics 17(11), 2015

CPP 42.7 Wed 17:00 H51

Improving the kinetics from molecular simulations using biased Markov state models — •Joseph Rudzinski, Kurt Kremer, and Tristan Bereau — Max Planck Institute for Polymer Research, Mainz, Germany

Molecular simulations can provide microscopic insight into the physical and chemical driving forces of complex molecular processes. Despite continued advancement of simulation methodology, model errors may lead to inconsistencies between simulated and experimentally-measured observables. This work presents a robust and systematic framework for reweighting the ensemble of dynamical paths sampled in a molecular simulation in order to ensure consistency with a set of given kinetic observables. The method employs the well-developed Markov state modeling framework in order to efficiently treat simulated dynamical paths. We demonstrate that, for two distinct coarse-grained peptide models, biasing the Markov state model to reproduce a small number of reference kinetic constraints significantly improves the dynamical properties of the model, while simultaneously refining the static equilibrium properties.

Invited Talk   CPP 42.8 Wed 17:15 H51

Principle of Maximum Caliber and its application in biology — •Kingshuk Ghosh — University of Denver

We will describe the principle of Maximum Caliber (MaxCal), a variational approach to model dynamical fluctuations. MaxCal is similar to the Maximum Entropy principle but applied in the trajectory space, natural language in describing several biological systems rang-ing from macromolecular dynamics to networks. We will briefly describe the general formalism and show the equivalence between Max-Cal and Markov processes. Next we will present application of MaxCal to model stochastic dynamics in biological systems, in particular genetic networks. These applications will highlight the role of MaxCal in describing complex systems with cooperativity, non-linearty and feed-back in a coarse-grained manner and yet provide us with quantitative tools to analyze data and gain valuable insights.

Invited Talk   CPP 42.9 Wed 17:45 H51

Coarse-graining of conservative and non-conservative interactions in molecular liquids — •Nico van der Vegt — Technische Universität Darmstadt, Darmstadt, Germany

In my talk, I will discuss two central challenges in multiscale simulations of soft matter: How can we improve the transferability of bottom-up coarse grained models? How can we improve the coarse-grained model's dynamical properties? Both questions are important, in particular when transport and nonequilibrium processes are studied. I will present a bottom-up coarse-graining procedure for constructing conservative and non-conservative (dissipative and stochastic) interactions for Dissipative Particle Dynamics (DPD) models of molecular liquids. This procedure is based on the Conditional Reversible Work (CRW) method.[1] I will address the chemical and state-point transferability of conservative potentials for coarse-grained molecular liquids in bulk and at interfaces,[2,3] and discuss challenges that remain in modelling dynamical properties of molecular liquids with CRW-DPD models.[4]

References: (1) E. Brini, V. Marcon, N. F. A. van der Vegt, PCCP 13, 10468-10474 (2011). (2) E. Brini, N. F. A. van der Vegt, J. Chem. Phys. 137, 154113 (2012). (3) V. R. Ardham, G. Deichmann, N. F. A. van der Vegt, F. Leroy, J. Chem. Phys. 143, 243135 (2015) (4) G. Deichmann, V. Marcon, N. F. A. van der Vegt, J. Chem. Phys. 141, 224109 (2014)