DY 41: Modeling and Simulation of Soft Matter II (joint session CPP/DY)

Time: Thursday 9:30–12:00   Location: H13

Invited Talk   DY 41.1   Thu 9:30   H13
Systematic Dynamic Coarse-Graining with Memory — Gerhard Jung, Martin Hanke, and •Friederike Schmid — Johannes Gutenberg Universität Mainz, Germany

In soft matter, systematic coarse-graining (CG) approaches often face the problem that the separation of time scales is incomplete, and memory effects become important. One challenge is to extract the CG dynamical equations, namely the memory kernel, from equilibrium all-atom simulations. Another challenge is to devise an algorithm that efficiently deals with pair-memory contributions to the dynamical equations. Such pair-memory interactions may become important, e.g., in dispersions of nanocolloids when the frequency dependence of hydrodynamic interactions cannot be neglected.

The talk will address these two problems. We propose a "generalized Langevin Dynamics" model, which has the form of a generalized Langevin equation with distance-dependent two-particle contributions to the self- and pair-memory kernels. A simulation algorithm is developed that scales linearly with the number of coarse-grained particles. Furthermore, we present a robust iterative method for the accurate reconstruction of memory kernels from dynamic correlation functions.

We apply the method to a suspension of nanocolloids with frequency-dependent hydrodynamic interactions. The GLD simulations perfectly reproduce the dynamics of the underlying fine-grained system and accelerate the simulation by a factor of roughly 10.000.

DY 41.2   Thu 10:00   H13
Accurate structure-based coarse-graining leads to consistent barrier-crossing dynamics — •Tristan Bereau and Joseph F. Rudzinski — MPI for Polymer Research, Mainz

Structure-based coarse-graining of molecular systems offers a systematic route to reproduce the many-body potential of mean force. Unfortunately, common strategies are inherently limited by the molecular-mechanics force field employed. Here we extend the concept of multi-surface dynamics, initially developed to describe electronic transitions in chemical reactions, to accurately sample the conformational ensemble of a classical system in equilibrium. In analogy to describing different electronic configurations, a surface-hopping scheme couples distinct conformational basins beyond the additivity of the Hamiltonian. The incorporation of more surfaces leads systematically toward improved cross-correlations. The resulting models naturally achieve consistent long-time dynamics for systems governed by barrier-crossing events. [Bereau and Rudzinski, arXiv:1808.05644]

DY 41.3   Thu 10:15   H13
Mapping multiple timescales in heterogeneous melts with predictive and adaptive subdomains — •Horacio V Guzman[1] and Hideki Kobayashi[2] — [1]Max Planck Institute For Polymer Research, Ackermannweg 10, 55128 Mainz, Germany — [2]Theory Research Interest Group, University of Cambridge Lensfield Road Cambridge

Heterogeneous molecular systems can be modeled by means of advanced simulations methods, as it is done with multiscale concurrent representations and non-equilibrium simulations. Interestingly, the heterogeneity of the mentioned systems has a huge potential to map and span time and length scales beyond fully atomistic simulations, since in most cases a subdomain of the simulation box can be tackled with slowly diffusive regime, while other remains in a faster diffusive regime. From this description, a crucial question arises on how to map those heterogeneous time scales without losing the theoretical speedup planned from the method development perspective. Here, we introduce the heterogeneous time-spatial domain decomposition approach which is a combination of an heterogeneity sensitive spatial domain decomposition with a time evolution average of particles' diffusion domainwise estimated. Within this approach, we present the theoretically modeled and results in scaling-laws for the force calculation time, while time-wise the subdomains with different diffusivity are adapted by means of the number of neighboring shells to a unique frequency of neighbour list updates. We explore the new approach capabilities, by comparing it with the state-of-the-art spatial domain decomposition techniques.

DY 41.4   Thu 10:30   H13
Automated detection of many-particle solvation states for

accurate characterizations of diffusion kinetics — •Joseph Rudzinski, Marc Radu, and Tristan Bereau — Max Planck Institute for Polymer Research, Mainz, Germany

Markov state models are powerful tools for reducing the complexity of molecular dynamics trajectories, but require configuration-space representations that accurately characterize the relevant dynamics. Well-established, low-dimensional order parameters for constructing this representation have led to widespread applications to study conformational dynamics of biomolecules. On the contrary, applications to characterize single-molecule diffusion have been scarce and typically employ system-specific, higher-dimensional order parameters to characterize local solvation states. In this work, we propose an automated method for generating a coarse configuration-space representation, using the coordination numbers about each particle. To overcome the noisy behavior of these low-dimensional observables, we treat the features as indicators of a latent Markov process. The resulting hidden Markov models filter the trajectories of each feature into the most likely latent solvation state at each time step. The filtered trajectories are then used to construct a configuration-space discretization, which accurately describes the diffusion kinetics. The method is validated on a standard model for glassy liquids, where particle jumps between local cages determine the diffusion properties. The resulting models provide quantitatively accurate characterizations of the diffusion constant and also reveal a mechanistic description of diffusive jumps.

DY 41.5   Thu 10:45   H13
Simulating liquid alkanes from first principles with machine learning potentials — •Max Veit — École Polytechnique Fédérale de Lausanne, 1015 Lausanne, CH — Department of Engineering, University of Cambridge, Cambridge CB2 1PZ, UK

The reliable prediction of the macroscopic properties of molecular liquids requires potential energy surface (PES) models that are not only accurate, but computationally efficient enough to handle large systems and reach long time scales typically inaccessible to explicit quantum-mechanical methods. This work introduces a new approach to the systematic approximation of the first-principles PES of a molecular liquid using the GAP machine learning method [A. Bartók, M. Payne, R. Kondor, and G. Csányi, Phys. Rev. Lett. 104, 136403 (2010)]. By applying machine learning to separately approximate each physical component of the interaction energy in a full many-body framework and with high and controllable accuracy, we can simulate the liquid accurately across a wide range of temperatures and pressures (with the inclusion of quantum nuclear effects) while gaining physical insight into the inner workings of the fluid. Following the recent success of this approach on predicting the equation of state of compressed fluid methane [M. Veit, S. K. Jain, S. Bonakala, I. Rudra, D. Hohl, and G. Csányi, arXiv:1810.10475], I will discuss how this approach can be extended to other molecular liquids with the help of emerging techniques in machine learning potential development, and how these ideas can be applied to other important molecular materials.

DY 41.6   Thu 11:00   H13
Effective interaction between fillers and their effect on phase separation of polymer blends. — •Alexander Chervanyov — Institut für Theoretische Physik, Westfälische Wilhelms-Universität Münster, 48149 Münster, DE

Effective interactions between fillers immersed in polymer blends are shown to arise from the interplay between the conventional compressibility mechanism and the mechanism relying on the polymer compositional variations in the vicinity of fillers. By developing and making use of the liquid state theory, we calculate the potentials of the effective interactions acting between fillers having different affinities for polymer species comprising the blend. Further, we study the effect of interacting fillers on phase separation in polymer blends. The calculated shift of the spinodal point and critical temperature are shown to significantly depend on the interactions between fillers and polymers.

15 min. break

DY 41.7   Thu 11:30   H13

Relative resolution: A multipole approximation at appropriate distances — ●Aviel Chaimovich[1], Kurt Kremer[2], and Christine Peter[3] — [1]Max Planck Institute of Colloids and Interfaces, 14476 Potsdam — [2]Max Planck Institute for Polymer Research, 55128 Mainz — [3]University of Konstanz, 78464 Konstanz

Recently, we introduced Relative Resolution as a hybrid formalism for fluid mixtures [1]. The essence of this approach is that molecular resolution switches in terms or relative separation: While near neighbors are characterized by a detailed fine-grained model, far neighbors are characterized by a simplified coarse-grained model. In this presentation, we notably show the comprehensive mathematics of our multiscale algorithm: We cast our Hamiltonian in terms of a multipole approximation at appropriate distances, which allows us for an analytical parameterization between the fine-grained and coarse-grained models. We consequently test the ability of Relative Resolution in describing various nonpolar liquids, in turn, capturing correctly and efficiently the statics and dynamics of many structural correlations and thermal properties across state space. Furthermore, we show that our multiscale approach works best if we switch between the fine-grained and coarse-grained potentials between the primary and secondary coordination shells: At this location, all orientations become negligible in the Hamiltonian. We conclude by discussing how Relative Resolution is the inherent variant, for molecular simulations, of the famous "cell-multipole" approach.

[1] A. Chaimovich, C. Peter, and K. Kremer (JCP, 2015).

DY 41.8   Thu 11:45   H13

Cononsolvency of thermoresponsive polymers: theory and simulation — ●Nico van der Vegt, Swaminath Bharadwaj, Cahit Dalgicdir, and Divya Nayar — Technische Universität Darmstadt, Germany

Cononsolvency of thermoresponsive polymers is an intriguing phenomenon whose molecular-scale mechanism remains elusive ever since it was first reported in the late 1980s. I will present theoretical results and computer simulations which provide a new angle on this question. Significantly, direct cosolvent binding to the polymer is shown to be an entropy-driven process, which leads to polymer dehydration resulting in coil-globule collapse and phase separation in poly(N-isopropyl acrylamide)/water/methanol solutions. Specific cosolvent interactions with the polymer play no role in this mechanism. Instead of that, cosolvent enrichment around hydrophobic groups frustrates hydration of the amide group and induces phase separation. Theoretical arguments and simulation results are furthermore presented which illustrate that direct cosolvent binding is not a necessary requirement for cononsolvency.

D. Nayar, N.F.A. van der Vegt, J. Phys. Chem. B 122, 3587-3595 (2018) N.F.A. van der Vegt, D. Nayar, J. Phys. Chem. B 121, 9986-9998 (2017) C. Dalgicdir, F. Rodriguez-Ropero, N.F.A. van der Vegt, J. Phys. Chem. B 121, 7741-7748 (2017)