

CPP 58: Computational Physics of Soft Matter II

Time: Wednesday 15:45–18:30

Location: PC 203

Invited Talk

CPP 58.1 Wed 15:45 PC 203

The Tricontinuous 3ths(5) Phase: A New Morphology in Copolymer Melts — ●GERD SCHROEDER-TURK^{1,2,3}, MICHAEL FISCHER^{1,2,4}, LILIANA DE CAMPO^{2,5}, JACOB KIRKENGAARD⁶, and STEPHEN HYDE² — ¹Theor. Physik, Fried.-Alex. Univ. Erlangen-Nürnberg, Germany — ²Applied Maths, Res. School of Physics & Eng., Australian National Univ., Canberra, Australia — ³School of Engineering & Inform. Technol., Murdoch Univ., Western Australia — ⁴Adolphe Merkle Institute, Fribourg, Switzerland — ⁵Australian National Science & Technol. Organisation, Bragg Institute, NSW, Australia — ⁶Niels Bohr Institute, Univ. of Copenhagen, Denmark

Self-assembly remains the most efficient route to the formation of ordered nanostructures, including the double gyroid network phase in diblock copolymers based on two intergrown network domains. Here we use self-consistent field theory to show that a tricontinuous structure with monoclinic symmetry, called 3ths(5), based on the intergrowth of three distorted ths nets, is an equilibrium phase of triblock star-copolymer melts when an extended molecular core is introduced. The introduction of the core enhances the role of chain stretching by enforcing larger structural length scales, thus destabilizing the hexagonal columnar phase in favor of morphologies with less packing frustration. This study further demonstrates that the introduction of molecular cores is a general concept for tuning the relative importance of entropic and enthalpic free energy contributions, hence providing a tool to stabilize an extended repertoire of nanostructured phases.

Reference: Fischer *et al*, *Macromolecules* **47**, 7424-7430 (2014)

CPP 58.2 Wed 16:15 PC 203

Coarse-grained electrostatic interactions for symmetric disk-shaped molecules — ●THOMAS HEINEMANN¹, KAROL PALCZYNSKI², JOACHIM DZUBIELLA², and SABINE H. L. KLAPP¹ — ¹Institut für Theoretische Physik, TU Berlin, 10623 Berlin — ²Institut für Physik - Komplexe Systeme, HU Berlin, Newtonstr. 15, 12489 Berlin

In the present study we introduce a novel route for calculating temperature-dependent effective pair potentials of disk-shaped molecules with van der Waals and electrostatic interactions. An ideal candidate for this study is the coronene molecule. For this molecule, an anisotropic and temperature-dependent coarse-grained model describing van der Waals interactions already exists [1]. Here we extend the approach towards electrostatic interactions. The electrostatic potential can be described through a set of differently charged rings [2]. However, the evaluation of the intermolecular ring-ring interactions is numerically quite involved. Therefore we introduce two strategies to cope with the ring-ring interactions. The first strategy aims at representing the entire electrostatics, given by the concentric set of rings, through a linear point quadrupole. In the second strategy, we fit the combined potential consisting of van der Waals and ring-ring-contributions with an extended van der Waals model. We discuss the applicability of both models and compare many-particle simulation results for crystalline structures with experimental data.

[1] T. Heinemann, K. Palczynski, J. Dzubiella and S. H. L. Klapp, *J. Chem. Phys.*, in press, arXiv: 1407.4352

[2] Obolensky et al., *Int. J. Quantum Chem.* **107**, 1335 (2007).

CPP 58.3 Wed 16:30 PC 203

Brownian Dynamics simulations on soft patchy particles made of isotropic spheres — ●MALTE LÜTJE — Universität Tübingen

Patchy Particles are widely used as a model system for proteins in a solution. We propose a dynamical model for patchy colloidal particles emerging from a binary mixture of particles of different size, "colloids" and "bonds". Both colloids and bonds interact with isotropic, but non-additive hard-sphere interactions, additionally the colloid-bond interaction contains a short-range, isotropic attraction. Our model resembles a soft version of the model studied by E. Zaccarelli et al. [1]

Equilibrium and dynamical properties of the mixture are investigated by Brownian Dynamics simulations. Due to the repulsion among the bonds a finite number of binding sites on a colloid is achieved.

We focus on a maximum number of four binding sites on each colloid, rendering possible a tetrahedral structure. Upon varying bond concentration, temperature and packing fraction ϕ of the colloids, we equilibrate the system and examine the resulting structures. Of par-

ticular interest is the transition from a locally clustered state at small volume fractions of colloids to the emerging tetrahedral network that was found in Ref. [1] for $\phi \geq 0.22$.

[1] E. Zaccarelli, F. Sciortino, P. Tartaglia, *J. Chem. Phys.* **127**, 174501 (2007)

CPP 58.4 Wed 16:45 PC 203

Self organization of asymmetric dipolar particles in external fields — ●ARZU BAHAR YENER and SABINE H. L. KLAPP — Institut für Theoretische Physik, Technische Universität Berlin, Hardenbergstr. 36, 10623 Berlin, Germany

A colloidal system of spherical particles with centred dipolar moments shows layer formation in the presence of an external rotating magnetic field [1]. This non-equilibrium phenomenon requires synchronization of the particles with the driving field [1]. Here, we investigate a model system of spherical particles with off-centred, i.e. laterally shifted dipole moments. The pair potential is modeled by a short-range repulsive potential and a point dipole potential. In a first step, we analytically and numerically determine the ground state structures of up to four particles. Then, we quantify the self assembly behaviour at non-zero temperature for several shifts by using Molecular Dynamics simulations and show that our model reproduces features observed for synthesized particles with magnetic caps [2] or Janus particles [3]. Finally, we apply an external rotating magnetic field and ask if layer formation emerges as in the centred system, and if synchronization is still a condition for layer formation.

[1] S. Jäger, and S. H. L. Klapp, *Soft Matter* **7**, 6606 (2011)

[2] D. Zerrouki, J. Baudry, D. Pine, P. Chaikin, and J. Bibette, *Nature* **455**, 380 (2008)

[3] J. Yan, M. Bloom, S. C. Bae, E. Luijten, and S. Granick, *Nature* **491**, 578 (2012)

CPP 58.5 Wed 17:00 PC 203

Assessing the Quality of Ab Initio Molecular Dynamics Simulations of Water by High-Dimensional Neural Network Potentials — TOBIAS MORAWIETZ¹, ANDREAS SINGRABER², CHRISTOPH DELLAGO², and ●JÖRG BEHLER¹ — ¹Lehrstuhl für Theoretische Chemie, Ruhr-Universität Bochum, D-44780 Bochum, Germany — ²Computational Physics, Universität Wien, Sensengasse 8/9, A-1090 Vienna, Austria

Ab initio molecular dynamics simulations have become a standard tool to study the properties of water, but they are severely hampered by the computational costs if long simulations of large systems are required. In recent years, interatomic potentials based on artificial neural networks (NNs) have been shown to provide very efficient and accurate potential-energy surfaces close to the reliability of electronic structure methods. Here we present a set of NN potentials for water based on density-functional theory. We demonstrate that these potentials can be used to assess the quality of different exchange-correlation functionals and the importance of van der Waals interactions in the description of liquid water.

CPP 58.6 Wed 17:15 PC 203

Proton disorder in cubic ice: Effect on the electronic and optical properties — VIVIANA GARBUIO¹, OLIVIA PULCI¹, MICHELE CASCELLA², IGOR KUPCHAK³, and ●ARI PAAVO SEITSONEN^{4,5} — ¹MIFP, ETSF, Dipartimento di Fisica, Università di Roma Tor Vergata, Italy — ²Department of Chemistry and Centre for Theoretical and Computational Chemistry, University of Oslo, Norway — ³MIFP, V. Lashkarev Institute of Semiconductor Physics of National Academy of Sciences of Ukraine, Kiev — ⁴Institut für Chemie, Universität Zürich, Switzerland — ⁵Département de Chimie, École Normale Supérieure, Paris, France

The proton disorder in ice has a key role in several properties such as the growth mode, thermo-dynamical properties and ferroelectricity. While structural phase transitions from proton disordered to proton ordered ices have been extensively studied, much less is known about their electronic and optical properties. Here, we present ab-initio many body perturbation theory based calculations of the electronic and optical properties of cubic ice at different levels of proton disorder. We compare our results with liquid water, as an example of fully (proton

and oxygen) disordered system. We find that, by increasing the disorder, a shrinking of the electronic gap occurs. Simultaneously, the excitonic binding energy decreases, so that the final optical gaps result to be almost independent on the degree of disorder. We explain these findings as an interplay between the local dipolar disorder and the electronic correlation.

CPP 58.7 Wed 17:30 PC 203

Molecular dynamics simulation of the dielectric permittivity tensor at the water/1,2-dichloroethane liquid-liquid interface — •ZHU LIU, HENDRIK H. HEENEN, KARSTEN REUTER, and CHRISTOPH SCHEURER — Technische Universität München

The liquid-liquid interface between Two Immiscible Electrolyte Solutions (ITIES) has been the focus of scientific interest for decades [1,2]. One important continuum response property of the liquids at the interface is expressed by the dielectric permittivity tensor, which sensitively reflects the unique microscopic characteristics in the interfacial region. The dielectric permittivity is known to change across the liquid-liquid interface, varying between the isotropic values for the pure liquids in the limit of the bulk far away from the interface. Recent work by Netz *et al.* based on Molecular Dynamics (MD) simulations for water at a solid-liquid interface revealed a rather unexpected behavior of the dielectric permittivity tensor as a function of the distance from the interface [3]. To our knowledge no comparable investigations are currently available for liquid-liquid interfaces. We present results for the prototypical water/1,2-dichloroethane (DCE) liquid-liquid interface making use of our recent implementation of a suitable polarizable force field in the Large-Scale Atomic/Molecular Massively Parallel Simulator (LAMMPS).

[1] A.G. Volkov *et al.*, *Liquid-Liquid Interfaces: Theory and Methods*, CRC Press, New York (1996).

[2] Z. Samec, *Electrochimica Acta* **84**, 21 (2012).

[3] D.J. Bonthuis *et al.*, *Langmuir* **28**, 7679 (2012).

CPP 58.8 Wed 17:45 PC 203

Quantum-Classical Adaptive Coupling in Grand-Canonical like Adaptive Resolution Simulations — •ANIMESH AGARWAL and LUIGI DELLE SITE — Institute for Mathematics, Arnimallee 6, D-14195, Freie Universität, Berlin, Germany

We have extended the recently developed Grand Canonical AdResS (GC-AdResS)[1,2] to quantum-classical adaptive coupling where the quantum delocalisation of an atom is described by the path integral formalism. Compared to standard adaptive coupling approaches [3], the advantage of GC-AdResS is that there is no need to obtain a coarse-grained model that correctly reproduces the structural and thermodynamic properties of a full PI (path integral) system, thereby eliminating the need to run a full PI simulation before starting the adaptive simulation. In this context, we have shown that spherical molecules described by a simple generic WCA potential in the coarse-grained region, act as a particle reservoir for the PI region. The resulting Grand Canonical set up is such that the structural and dynamical properties of quantum flexible water models in the PI subregion in AdResS are

consistent with the properties obtained in the same subregion in full PI simulations.

[1] H.Wang, C.Hartmann, C.Schütte and L.Delle Site, *Phys.Rev.X* **3**, 011018 (2013)

[2] A.Agarwal, H.Wang, C.Schütte and L.Delle Site,*J.Chem.Phys.* **141**, 034102 (2014)

[3] A.B. Poma and L.Delle Site, *Phys. Rev. Lett.* **104**, 250201 (2010)

CPP 58.9 Wed 18:00 PC 203

Reactive molecular dynamics simulation of the twin polymerization process with ReaxFF — THOMAS SCHÖNFELDER¹, •JANETT PREHL¹, JOACHIM FRIEDRICH², STEFFEN SEEGER¹, STEFAN SPANGE³, and KARL HEINZ HOFFMANN¹ — ¹Computerphysik, Technische Universität Chemnitz — ²Theoretische Chemie, Technische Universität Chemnitz — ³Polymerchemie, Technische Universität Chemnitz

Twin polymerization is a new synthesis concept, which enables the formation of two different macromolecular structures from organic-inorganic hybrid materials in one single process step. To gain insights into formation processes we implement a first-principles-based ReaxFF reactive force field for C/H/O/Si for the initial electrophilic substitution of an aromatic system [1]. In this presentation we first give a brief introduction to the details of twin polymerization and afterward we present our progress [2] in establishing an appropriate reactive force field parametrization to model all partial reaction steps and the subsequent polymerization at least for small system sizes.

[1] T. Schönfelder, J. Friedrich, J. Prehl, S. Seeger, S. Spange, and K. H. Hoffmann, *Chem. Phys.* **440** (2014) 119-126

[2] T. Schönfelder, J. Prehl, J. Friedrich, and K. H. Hoffmann, to be submitted

CPP 58.10 Wed 18:15 PC 203

Development of the extended ReaxFF+ approach to accurately model ionic system — •OLIVER BÖHM, STEPHAN PFADENHAUER, and PHILIPP PLÄNITZ — AQcomputare GmbH, 09125 Chemnitz, Germany

In order to be able to describe ionic systems within a liquid environment we have extended the original bond order dependent reactive force field (ReaxFF) of the van Duin group [1]. The new approach is called ReaxFF+ [2]. It uses a new charge equilibrium scheme which is able to model ionic as well as neutral systems. The main goal is the correct description of the ionic and covalent behaviour of bonds. This is realized by a bond order dependent Coloumb screening and an electronegativity which is a function of the over- and undercoordination. This allows the simulation of reactions in basic and acid solutions as well as the charge transfer in bond breaking reactions of neutral molecules. The accuracy of the scheme is demonstrated for different ionic molecules.

[1] A. van Duin *et al.*, *J. Phys. Chem. A*, **105**, 9396 (2001); *J. Phys. Chem. A*, **107**, 3803 (2003)

[2] O. Böhm *et al.*, *J. Am. Chem. Soc.* (2015) submitted