

DY 27: Complex Fluids and Colloids, Micelles and Vesicles (joint session CPP/DY)

Time: Wednesday 9:30–12:45

Location: H14

Invited Talk

DY 27.1 Wed 9:30 H14

Microstructural transitions and characterization of capillary suspensions — SEBASTIAN BINDGEN¹, FRANK BOSSLER², IRENE NATALIA¹, and •ERIN KOOS¹ — ¹Department of Chemical Engineering, KU Leuven, Leuven, 3001, Belgium — ²Institute for Mechanical Process Engineering, Karlsruhe Institute of Technology, Gotthard-Franz-Str. 3, 76131 Karlsruhe, Germany.

Suspensions can exhibit a wide range of rheological behaviors that are closely linked to both the bulk particle structure as well as the microstructure including direct particle contacts. Graph theory offers methods and parameters that can be used to analyze complex structures. This method is demonstrated using ternary liquid-liquid-solid systems, which exhibit a wide variety of different morphologies depending on the ratio of the three components. We analyze these networks using the coordination number and clustering coefficient. These parameters are compared to the measured storage and loss moduli.

These capillary suspension networks also exhibit atypical rheological behavior. For instance, a negative normal stress difference is observed from re-orientation of the flocs into the vorticity direction during shearing. Typically, systems with negative normal stress differences have either high volume concentrations and are shear thinning, or are shear thickening with very low particle concentrations. In contrast, the capillary suspensions we report here have a 25% solid concentration and are shear thinning; a combination that has never before been reported in literature.

DY 27.2 Wed 10:00 H14

Flow of silica rods in microfluidic channels — •HANSOL JEON¹ and DIRK AARTS² — ¹Max Planck Institute for dynamics and self-organization, Göttingen, Germany — ²University of Oxford, Oxford, United Kingdom

This project presents the flow of dense liquid crystal phases of colloidal silica rods in microfluidic channels. We describe the synthesis of silica rods that exhibit three different phases (isotropic, nematic and smectic). Using microfluidic channels of novel designs, we observe the flow behaviour of silica rods in the channels by confocal microscopy. We then conduct image analysis to track the dyed rods and measure the flow profile. We compare the flow profiles with the theory and provide future outlook to this experiment.

DY 27.3 Wed 10:15 H14

Dynamics during the formation of an arrested protein phase separation studied using X-ray photon correlation spectroscopy — •NAFISA BEGAM¹, ANITA GIRELLI¹, ANASTASIA RAGULSKAYA¹, HENDRIK RAHMANN², KEVIN LOPATA², FABIAN WESTERMEIER³, CHRISTIAN GUTT², FAJUN ZHANG¹, and FRANK SCHREIBER¹ — ¹Universität Tübingen, Germany — ²Universität Siegen, Germany — ³Petra III, DESY, Germany

Phase separation in aqueous protein solutions is of primary interest in the field of many biological and chemical processes. It is observed that the glass line often interrupts the phase separation [1] and leads the solution to a dynamically arrested state which results in a drastic reduction of the microscopic relaxation rate. Kinetics of phase separation of such a system of bovine γ -globulin in the presence of polyethylene glycol studied using ultra-small angle X-ray scattering [2] shows an arrest for a quench to temperatures below $\sim 2.5^\circ\text{C}$. Here, our goal is to study the dynamics of the solution while approaching such an arrested state. We have used X-ray photon correlation spectroscopy which provides the time evolution of dynamics in the form of two time correlation (TTC) functions. A systematic study of the evolving dynamics of protein solutions during arrest at different quench temperatures using TTC exhibits initially an exponential growth of the relaxation time and at the later stage, an ageing heterogeneous dynamics. This study reveals a comprehensive picture of the protein dynamics that leads the solution to an arrested state. [1] Cardinaux et. al, *Phys. Rev. Lett.*, **99**, 118301, (2007) [2] S. Da Vela et. al, *Soft Matter*, **13**, 8756, (2017)

DY 27.4 Wed 10:30 H14

Electron-Atom duality in DNA-programmable assembly — •MARTIN GIRARD¹, ANINDITA DAS^{2,4}, SHUNZHI WANG^{2,4}, JINGSHAN DU^{3,4}, BYEONGDU LEE⁵, CHAD A. MIRKIN^{2,4}, and MONICA OLVERA DE LA CRUZ^{3,6} — ¹Max Planck Institute for Polymer Physics, Mainz,

Germany — ²Department of Chemistry, Northwestern University, Evanston, IL, USA — ³Department of Material Science and Engineering, Northwestern University, IL, USA — ⁴International Institute for Nanotechnology, Evanston, IL, USA — ⁵X-ray Science Division, Advanced Photon Source, Argonne National Laboratory, IL, USA — ⁶Department of Physics and Astronomy, Northwestern University, Evanston, IL, USA

A powerful method for designing colloidal crystals involves the use of DNA as a particle-directing ligand. With such systems, DNA-nanoparticle conjugates are considered programmable atom equivalents (PAEs), and design rules have been devised to engineer complex crystallization outcomes. Here, we report a new property of PAEs, a type of electron-atom duality. When reduced in size and DNA grafting density, PAEs can behave as electron equivalents (EEs) and move through lattices defined by larger PAEs. In such mixtures, the EEs roam through the crystals as electrons do in metals, holding the large PAEs in specific lattice sites. As the number of strands increases or the temperature decreases, the EEs localize yielding a transition from a metal to a compound. This concept of electron-atom-equivalent duality changes the way we think about colloidal systems and helps define new routes to metallic, intermetallic, and compound phases.

DY 27.5 Wed 10:45 H14

Interaction forces in model cement systems — •SIMON BECKER and REGINE VON KLITZING — Soft Matter at Interfaces, Institut für Festkörperphysik, TU Darmstadt

For the workability of concrete and cement as highly abundant construction materials the flow behaviour in the early stages of these materials is of special interest. This can be influenced using additives such as super plasticizers to decrease the viscosity of cement pastes, yielding an enhanced workability. A better understanding of the interactions on the nanoscopic level and the impact on the macroscopic rheology is of great importance in the development of higher performing additives.

The interaction between model particles is mapped via colloidal-probe atomic force microscopy (CP-AFM) in sphere-sphere geometry. Combining CP-AFM with optical microscopy allows to arrange colloidal particles coaxially and determine the interaction forces between these particles upon approach. Spherical silica beads with diameters in the micrometer range serve as model cement grains due to the restriction of the AFM technique to either spherical particles or particles with a defined roughness. The influence on the interaction forces of these model grains in presence of different pH and different salts such as KCl and CaCl_2 and their concentrations is investigated. Furthermore the change of the interaction between the model particles in presence of different concentrations of PCE is examined.

DY 27.6 Wed 11:00 H14

Computing the 3D Radial Distribution Function from Particle Positions: An Advanced Analytic Approach — •BERND F. A. KOPERA and MARKUS RETSCH — Department of Chemistry, University of Bayreuth, Universitätsstraße 30, 95447 Bayreuth

The radial distribution function, $g(r)$, is ubiquitously used to analyze the internal structure of particulate systems. However, experimentally derived particle coordinates are always confined to a finite sample volume. This poses a particular challenge on computing $g(r)$: Once the radial distance, r , extends beyond the sample boundaries in at least one dimension, substantial deviations from the true $g(r)$ function can occur. State of the art algorithms for $g(r)$ mitigate this issue for instance by using artificial periodic boundary conditions. However, ignoring the finite nature of the sample volume distorts $g(r)$ significantly. Here, we present a simple, analytic algorithm for the computation of $g(r)$ in finite samples. No additional assumptions about the sample are required. The key idea is to use an analytic solution for the intersection volume between a spherical shell and the sample volume. In addition, we discovered a natural upper bound for the radial distance that only depends on sample size and shape. This analytic approach will prove to be invaluable for the quantitative analysis of the increasing amount of experimentally derived tomography data.

15 min. break

DY 27.7 Wed 11:30 H14

A classical density functional from machine learning and a convolutional neural network — ●SHANGCHUN LIN and MARTIN OETTEL — Institut für Angewandte Physik, Universität Tübingen, Tübingen, Deutschland

We use machine learning methods to approximate a classical density functional. The functional *learns* by comparing the density profile it generates with that of simulations. As a study case, we choose the model problem of a Lennard Jones fluid in one dimension where there is no exact solution available and training data sets must be obtained from simulations. After separating the excess free energy functional into a "repulsive" and an "attractive" part, machine learning finds a functional in weighted density form for the attractive part. The density profile at a hard wall shows good agreement for thermodynamic conditions beyond the training set conditions. This also holds for the equation of state if it is evaluated near the training temperature.

DY 27.8 Wed 11:45 H14

Semi-dilute mixtures of circular and linear polymers - towards novel separation techniques — ●LISA B. WEISS¹, CHRISTOS N. LIKOS¹, and ARASH NIKOUBASHMAN² — ¹Faculty of Physics, University of Vienna, Boltzmanngasse 5, A-1090 Vienna, Austria — ²Institute of Physics, Johannes Gutenberg University Mainz, Staudingerweg 7, 55128 Mainz, Germany

Linear polymers have been extensively studied in various concentration regimes, at rest and when exposed to a variety of flow fields. Recently, this interest extended to ring polymers, focusing on melts and dilute solutions. When diluted and at rest, rings show the same scaling behavior as chains, but when exposed to non-equilibrium conditions astonishing differences become apparent uniquely related to their topology. Furthermore, entangled rings show in equilibrium as well as when sheared marked differences compared to their linear counter part. However, the semi-dilute regime of pure ring polymer solutions and topological mixtures of rings and chains have been sparsely investigated so far. To resolve this, we simulate pure linear and ring polymer solutions and their mixtures around the overlap concentration at rest and under Hagen-Poiseuille flow. To correctly take into account hydrodynamic interactions, we employ the multi-particle collision algorithm. Mixtures of chains and rings show a distinct cross stream migration behavior at any investigated concentration, offering a promising route towards novel separation techniques. In particular, we observed a pronounced propensity of the rings to focus at the channel center while the chains populate the region close to the confining walls.

DY 27.9 Wed 12:00 H14

Ring polymers in confined geometries with mixed boundary conditions — ●ZORYANA USATENKO¹, PIOTR KUTERBA², and JOANNA HALUN³ — ¹Institute of Physics, Cracow University of Technology, Cracow 30-084, Poland — ²Jagiellonian University, Cracow 30-348, Poland — ³Institute of Nuclear Physics PAN, Cracow 31-342, Poland

Investigation of a dilute solution of phantom ideal ring polymers and ring polymers with excluded volume interactions in a good solvent confined in a slit geometry of two parallel walls with mixed boundary conditions which corresponds to the case of one repulsive and the other one inert wall is performed. Besides, taking into account the Derjaguin approximation the investigation of a dilute solution of ring polymers confined in a solution of mesoscopic colloidal particles of big size with different radii and different adsorbing or repelling properties in respect for polymers is performed. The calculation of the dimensionless depletion interaction potentials, the depletion forces and the monomer density profiles were performed in the framework of the massive field

theory approach at fixed space dimension $d < 4$ up to one loop order. The density-force relation in the case of phantom ideal ring polymer chains is analyzed and the respective universal amplitude ratio is calculated. The obtained results indicate the interesting and nontrivial behaviour of ring polymers in confined geometries and give possibility better to understand the complexity of physical effects arising from confinement and chain topology, especially in the case of interaction of λ bacteriophages with E.coli bacteria cells.

DY 27.10 Wed 12:15 H14

Multi-scale simulations of polymeric nanoparticle fabrication through rapid solvent exchange — ●ARASH NIKOUBASHMAN¹, NANNAN LI², and ATHANASSIOS PANAGIOTOPOULOS² — ¹Institut für Physik, Johannes Gutenberg Universität Mainz, Mainz, Deutschland — ²Department of Chemical and Biological Engineering, Princeton University, Princeton, USA

Tailored nanoparticles are increasingly sought after for many scientific and technological applications, such as optoelectronic devices and selective catalysts. However, both research and commercialization of these materials has been impeded by the lack of suitable fabrication techniques. One promising approach for overcoming this hurdle is flash nanoprecipitation, where (soft) nanoparticles are assembled through rapid micromixing of polymers in solution with a miscible poor solvent. This continuous process allows for high yields as well as precise control over particle size and morphology. We employed multiscale simulations of this process to understand its underlying mechanisms and to efficiently explore parameter space. We first performed explicit solvent molecular dynamics (MD) simulations of a bead-spring polymer model to study the microscopic properties of the fabrication process. Then, we fed the MD data into a kinetic Monte Carlo algorithm to reach macroscopic length- and timescales. We discovered that the nanoparticle size can be reliably tuned through the initial polymer concentration and the mixing rate. Further, we were able to fabricate a wide variety of structured colloids, such as Janus and core-shell particles, when polymer blends were used in the feed stream.

DY 27.11 Wed 12:30 H14

Molecular Simulation of Thermodynamic Properties with the Osmotic Equilibrium Approach: Accessing Activities in Complex Concentrated Liquid Phases — ●MICHAEL BLEY^{1,2}, MAGALI DUVAİL¹, PHILIPPE GUILBAUD³, and JEAN-FRANÇOIS DUFRÊCHE¹ — ¹ICSM, CEA, CNRS, ENSCM, Univ Montpellier, BP 17171, F-30207 Bagnols-sur-Cèze, France — ²Applied Theoretical Physics-Computational Physics, Physikalisches Institut, Albert-Ludwigs-Universität Freiburg, Hermann-Herder Str. 3, D-79104 Freiburg, Germany — ³Nuclear Energy Division, Research Department on Mining and Fuel Recycling Processes (SPDS/LILA), CEA, BP 17171, F-30207 Bagnols sur Cèze, France

Thermodynamic properties such as activities of complex concentrated liquid phases are a key parameter for describing chemical equilibria by means of mass action law. The mass action law and the corresponding equilibrium constant provide a measure for the direction of any chemical equilibrium. The osmotic equilibrium approach has been developed for accessing activities for a huge bandwidth of complex liquid systems. Starting from Molecular Dynamics (MD) simulations of vapor-liquid interfaces using explicit polarization provides a direct access to the activity by comparing the mean amount of evaporated molecules of a given species in the vapor phase for a mixture and for the pure species, respectively. Obtained structures and thermodynamic properties showed a very good agreement with previous experimental and theoretical studies on various aqueous electrolyte solutions and organic solvent phases up to high solute concentrations.