Location: MOL 213

CPP 24: Complex Fluids and Soft Matter I (joint session DY/CPP)

Time: Tuesday 10:00-13:00

CPP 24.1 Tue 10:00 MOL 213 How are mobility and friction related in viscoelastic fluids? — •JULIANA CASPERS¹, NIKOLAS DITZ², KARTHIKA KR-ISHNA KUMAR², FELIX GINOT², CLEMENS BECHINGER², MATTHIAS FUCHS², and MATTHIAS KRÜGER¹ — ¹Institute for Theoretical Physics, Georg-August Universität Göttingen, 37073 Göttingen, Germany — ²Fachbereich Physik, Universität Konstanz, 78457 Konstanz, Germany

The motion of a colloidal probe in a viscoelastic fluid is described by friction or mobility, depending on whether the probe is moving with a velocity or feeling a force. While the Einstein relation describes an inverse relationship valid for Newtonian solvents, both concepts are generalized to time-dependent memory kernels in viscoelastic fluids. We theoretically and experimentally investigate their relation by considering two observables: the recoil after releasing a probe that was moved through the fluid and the equilibrium mean squared displacement (MSD). Applying concepts of linear response theory, we generalize Einstein's relation and thereby relate recoil and MSD, which both provide access to the mobility kernel. With increasing concentration, however, MSD and recoil show distinct behaviors, rooted in different behaviors of the two kernels. Using two theoretical models, a linear two-bath particle model and hard spheres treated by mode-coupling theory, we find a Volterra relation between the two kernels, explaining differing timescales in friction and mobility kernels under variation of concentration.

Can liquid-state theory predict jamming of hard particles? — •CARMINE ANZIVINO¹, MATHIAS CASIULIS², AMGAD MOUSSA³, STEFANO MARTINIANI², and ALESSIO ZACCONE¹ — ¹Department of Physics "A. Pontremoli", University of Milan, via Celoria 16, 20133 Milan, Italy — ²Center for Soft Matter Research, Department of Physics, New York University, New York 10003, USA — ³Syngenta AG, 4058 Basel, Switzerland

By generalizing the notion of maximally random jammed (MRJ) state [1,2] to that of MRJ-line, we show [3] that it is reasonable to assume the most random branch of jammed states to undergo crowding in a way qualitatively similar to an equilibrium liquid. We then prove that, for hard-sphere systems, liquid-state theories can be succesfully used to estimate the RCP density, when the latter is identified with the densest isostatic point, i. e. the densest among the MRJ states with z=6.

Our finding is further enforced by the analysis of polydisperse systems. Either in the case of bidisperse and polydisperse hard spheres our prediction of the RCP density is in very good agreement with simulations, for a large values of size ratios and polydispersity.

[1] S. Toquato, T. M. Truskett, and P. G. Debenedetti, Phys. Rev. Lett. 84, 2064 (2000). [2] S. Torquato and F. H. Stillinger, Reviews of Modern Physics 82, 2633 (2010). [3] C. Anzivino, M. Casiulis, T. Zhang, A. S. Moussa, S. Martiniani and A. Zaccone, "Estimating RCP as the densest isostatic packing in bidisperse and polydisperse hard spheres", manuscript submitted (2022).

CPP 24.3 Tue 10:30 MOL 213

2D crystals of squares and the tetraic phase — •PETER KEIM — Max-Planck-Institut für Dynamik und Selbstorganisation, Göttingen

Squares (5 micron edge-length) were manufactured from a photo-resist using a 3D nanoprinter (nanoscribe GT). In aqueous solution, particles sediment by gravity to a thin cover slide where they form a mono-layer of Brownian particles. The curvature of the cover slide can be adjusted from convex to concave, which allows to vary the area-density of the mono-layer from 700 to 1500 particles in the field of view. For low densities, the squares are free to diffuse and form a 2D fluid while for high densities they form a quadratic crystal. In analogy to 2D-melting by topological defects with an intermediate hexatic phase for isotropic particles (KTHNY-theory and Nobel-price 2016), a four-folded bondorder correlation function is used to resolve a tetratic phase with quasilong-range orientational order but short rang translational order.

CPP 24.4 Tue 10:45 MOL 213

Transient microrheology unveils the presence of two relaxation processes in viscoelastic fluids — •Félix Ginot¹, Juliana CASPERS², LUIS FRIEDER REINALTER¹, KARTHIKA KRISHNA KUMAR¹, MATTHIAS KRÜGER², and CLEMENS BECHINGER¹ — ¹Fachbereich Physiks Universität Konstanz, 78457 Konstanz, Germany — ²Institute for Theoretical Physics, Georg-August Universität Göttingen, 37073 Göttingen, Germany

We experimentally investigate the transient dynamics of a colloidal probe particle in a viscoelastic fluid after the driving force acting on the probe is suddenly removed. In this situation, the probe exhibits a strong backward recoil, with two distinct timescales. While the first timescale naturally originates from the viscoelastic properties of the fluid, the second timescale arises from the coupling between the probe and the bath. These experimental observations are in excellent agreement with a microscopic model which considers the probe particle to be coupled to two bath particles via harmonic springs. Interestingly, this model exhibits two sets of eigenmodes corresponding to reciprocal and non-reciprocal force conditions, and which can be experimentally confirmed in our experiments. We expect our findings to be relevant under conditions where particles are exposed to non-steady shear forces as this is encountered e.g. in microfluidic sorting devices or the intermittent motion of motile bacteria within their natural viscoelastic surroundings.

15 min. break

 $\label{eq:CPP 24.5} Tue \ 11:15 \quad MOL \ 213$ Phason strain-free growth of quasicrystals based on purely local rules and without repair mechanism — STEFAN WOLF¹, MICHAEL ENGEL², and •MICHAEL SCHMIEDEBERG¹ — ¹Institute of Theoretical Physics, Friedrich-Alexander-Universität Erlangen-Nürnberg, 91058 Erlangen, Germany — ²Institute for Multiscale Simulation, Friedrich-Alexander-Universität Erlangen-Nürnberg, 91058 Erlangen, Germany

We introduce a simple model for the growth of colloidal quasicrystals where new particles are sequentially added according to specific local rules to the system in three dimensions. Subsequent changes to the particles are not allowed, i.e., no phasonic rearrangements can occur once a particle has been placed into the system. Our findings demonstrate that the purely local energetic rules are sufficient to obtain complex long-ranged order. Thus, phasonic rearrangements or fluctuations might be important to facilitate the formation of almost perfect quasicrystals [1-3] but they are not indispensable.

 C.V. Achim, M. Schmiedeberg, and H. Löwen, Phys. Rev. Lett. 112, 255501 (2014).

[2] A. Gemeinhardt, M. Martinsons, and M. Schmiedeberg, Eur. Phys. J. E 41, 126 (2018).

[3] K. Nagao, T. Inuzuka, K. Nishimoto, and K. Edagawa, Phys. Rev. Lett. 115, 075501 (2015).

CPP 24.6 Tue 11:30 MOL 213 Magnus force on microscopic spinning objects moving through non-Markovian baths — XIN CAO¹, •DEBANKUR DAS², NIKLAS WINDBACHER¹, FELIX GINOT¹, MATTHIAS KRUGER², and CLEMENS BECHINGER¹ — ¹Fachbereich Physik, University Konstanz, 78464 Konstanz, Germany — ²Institut für Theoretische Physik, Universität Göttingen, 37077 Göttingen Germany

When a spinning object moves through a fluid or air, its direction of motion becomes deflected due to the Magnus force that is perpendicular both to the moving direction and the spinning axis. Since the Magnus effect is caused by inertial effects within the surrounding medium, it should vanish at micro scales where viscous forces dominate over inertia. Recent experiments have observed the phenomenon similar to Magnus effect when a spinning colloids and there aggregates are externally driven through a viscoelastic fluid. Even though the deflection force shows a similar dependence on the spinning and translating velocity of the particles as in case of Magnus forces, its sign is reversed. Here, we have developed a theory of such motions which doesnot rely on the inertial effects but caused explicitly by the memory effect of the viscoelastic fluid. To better elucidate our theory we corroborate our results with experiments. Our theory successfully captures the density relaxation timescales of the viscoelastic fluid. Further we proposed viable theoretical predictions which can be verified with further experiments.

CPP 24.7 Tue 11:45 MOL 213

Preferential alignment of colloidal dumbbells with recoil direction — •KARTHIKA KRISHNA KUMAR¹, FÉLIX GINOT¹, JU-LIANA CASPERS², MATTHIAS KRÜGER², and CLEMENS BECHINGER¹ — ¹Fachbereich Physik, Universität Konstanz, Germany — ²Institute for Theoretical Physics, Georg-August Universität Göttingen, Germany

Unlike Newtonian fluids, viscoelastic fluids can store and dissipate energy on much longer timescales leading to non-Markovian dynamics. Hence, probing viscoelastic fluids using colloidal particles reveal complex dynamics in microscopic lengthscales. A consequence of this is the recoil behavior of a colloidal particle after dragging it through a viscoelastic fluid. In this work, we use a pair of colloidal particles stuck together due to depletion interactions forming a dumbbellshaped structure. This gives the advantage of resolving the orientational component in addition to the translational components of the particle motion. Surprisingly, we report that the axis of the dumbbell tends to align with the direction of the motion during a recoil. The amplitude of this orientational component follows a non-linear trend even in the regime where translational recoil amplitudes show a linear increase with shear velocity. This behavior can no longer be explained by the linear two-bath particle model which is able to explain the biexponential translational recoils. Furthermore, the amplitude of this re-orientation increases with the initial angle at which the dumbbell is dragged. This points to an asymmetric distribution of elastic energy between the two particles of the dumbbell which might cause this effect.

CPP 24.8 Tue 12:00 MOL 213

Entropic phase diagram of twisted convex particles — •POSHIKA GANDHI and ANJA KUHNHOLD — Institute of Physics, University of Freiburg, Germany

The field of liquid crystal simulations has, over the years, benefitted immensely from the study of purely entropy driven systems. Since Onsager's[1] predictions of the existence of a nematic phase in long rods, the list of known phases of hard rod-like particles has grown to include phases like smectics and cholesterics.

An important parameter in the formation of these phases is the particle shape anisotropy. By considering unusual particle shapes new phases can be discovered. Recently, Dussi and Dijkstra[2] showed the formation of stable chiral nematic phase in twisted polyhedral particles using only entropic interactions.

We used Monte Carlo NVT simulations to produce phase diagrams of a different class of twisted particles - convex triangular and rectangular prisms. The results show a host of previously undiscovered phases arising from the shape anisotropy parameters like twist angle and aspect ratios.

[1] Onsager, L., Ann. N. Y. Acad. Sci., 51, 627 (1949).

[2] Dussi, S., Dijkstra, M., Nat Commun. 7, 11175 (2016).

CPP 24.9 Tue 12:15 MOL 213

Optimizing the Structure of Acene Clusters — •PHILIPP EL-SÄSSER and TANJA SCHILLING — Institute of Physics, University of Freiburg, Germany

In the production of organic solar cells, neutral acene cluster beams are used to create thin films. The molecules in these clusters can be arranged in various ways. Most types of acene molecules are quasi two dimensional with one long axis - they prefer stacked or herring-bone structures. Additionally, the positions of the molecules in a cluster may vary. Thus, the exact way of how they arrange has a strong influence on the overall energy of the cluster.

We have investigated the structures of anthracene, tetracene, and pentacene clusters with up to 30 molecules. In order to find the configurations at the global minimum of the potential energy surface with respect to the positions of the atoms, we applied the Basin-Hopping Monte-Carlo (BH) algorithm to atoms described by the polymerconsistent force field - interface force field (PCFF-IFF). We studied for these cluster structures the relative stability between different sizes of clusters, as well as the accessibility of the global minimum at different temperatures.

CPP 24.10 Tue 12:30 MOL 213 Ergodicity breaking in overpacked colloidal hard spheres — •HANS JOACHIM SCHÖPE — Universität Tübingen, Institut für angewandte Physik, Auf der Morgenstelle 10, 72076 Tübingen

The ergodic hypothesis is an essential prerequisite for the applicability of statistical mechanics in thermodynamic equilibrium. Direct experimental evidence of the validity of the ergodic hypothesis is extremely rare. Furthermore, the question arises to what extent - if at all - ergodicity exists in non-equilibrium. We have realized a novel dynamic light scattering experiment, which makes it possible to determine the probability distribution of relaxation-times in colloidal suspensions. We present here a systematic study of the relaxation-time distribution in colloidal hard spheres at the transition from equilibrium to non-equilibrium. In thermodynamic equilibrium, we can impressively confirm the ergodic hypothesis and show that the fluctuations are of a Gaussian nature. Out of equilibrium, we can detect non-Gaussian behavior, which increases rapidly with increasing undercooling (overpacking). The ergodic hypothesis is no longer fulfilled in non-equilibrium. Furthermore, we observe that the metastable fluid ages in the induction stage, the non-Gaussian fluctuations increase in the time before crystallization sets in. To what extent these fluctuations cause crystallization must be clarified in the future.

CPP 24.11 Tue 12:45 MOL 213 Coacervates from a polyelectrolye and a small polyion: preparation, phase behavior and theoretical modeling — LUCY CORIA-ORIUNDO¹, EUGENIA APUZZO², SANTIAGO HERRERA¹, MARCELO CEOLÍN², GABRIEL DEBAIS¹, FERNANDO BATTAGLINI¹, and •MARIO TAGLIAZUCCHI¹ — ¹Universidad de Buenos Aires, Bs.As., Argentina — ²Universidad Nacional de la Plata, Bs.As., Argentina.

A mixture of oppositely charged polyelectrolytes, under the proper experimental conditions, can undergo liquid-liquid phase separation. The resulting polymer-rich phase is usually known as polyelectrolyte coacervate. This work reports liquid coacervates composed of a small polyanion (ferricyanide) and branched poly(ethyleneimine) (BPEI), a polycation. The phase diagram of the system was measured as a function of the concentration of both components at fixed pH = 6 and a concentration of added NaCl of 0.5 M. The salt resistance of the coacervate was studied and it was found that the coacervate is stable up to [NaCl] = 1.35 M. The phase diagram and salt-resistance experiments were modeled with a statistical-thermodynamics formalism that models the association of the oppositely charged species as a pseudochemical-equilibrium. The model fits very well the experimental data and was used to analyze the differences between polymer-polymer and polymer-small ion coacervates. Finally, the diffusion coefficient of ferricyanide within the coacervated (measured with cyclic voltammetry) was shown to increase 10 times when the concentration of added NaCl was increased from 0 to 1.2 M.