CPP 47: Heterogeneous Nucleation and Microstructure Formation (related to SYMF, jointly with MM)

Time: Friday 10:30–12:45 Location: ZEU 160

CPP 47.1 Fri 10:30 ZEU 160

Nanoparticle solution casting kinetics on colloidal templates - an in-situ microbeam GISAXS and ellipsometry study — •Stephan V. Roth[1], Adeline Buffet[1], Mottakin Abul Kashem[1], Gunthard Benecke[1,2], Rainer Gehrke[1], Gerd Herzog[1], Volker Koerstgens[3], Jan Perlich[1], Monika Rawolle[3], Andre Rothkirch[1], Matthias Schwartzkopf[1], and Peter Mueller-Buschbaum[3] — [1]Deutsches Elektronen-Synchrotron (DESY), Notkestr. 85, D-22607 Hamburg, Germany — [2]MPIKG Golm, Abt. Biomat., Wissenschaftspark Potsdam-Golm, D-14424 Potsdam, Germany — [3]TU München, Physik Department, LS Funkt. Mat., James-Franck-Str. 1, 85748 Garching, Germany

Nanoparticle solution casting is a very efficient process to produce large scale nanostructured layers [1] which are applied in optical or sensor technology [2]. We investigated the self-assembly process of gold nanoparticles during solution casting on a colloidal polymeric template. The template was installed by spin-coating and further nanostructured by toluene treatment prior to solution casting. In order to follow the different stages of self-assembly during evaporation of the solvent, we combined imaging ellipsometry [3] and microbeam grazing incidence small-angle x-ray scattering (μGISAXS) at the beamline MiNaXS (P03) of DESY, Hamburg. Thereby we are able to image the structure formation of the gold nanoparticle layer and its influence on the colloidal template. [1] Roth et al., Appl. Phys. Lett. 91, 091915 (2007)& Langmuir, 26, 1496 (2010) [2] Xia et al., Adv. Mat. 12, 693 (2000) [3] Koerstgens et al., Anal. Bioanal. Chem. 396, 139 (2010)

CPP 47.2 Fri 10:45 ZEU 160

Microstructures of a binary mixture of charged colloidal spheres with eutectic phase behaviour — •Thomas Palberg, Nina J. Lorenz, and Izidor Lorencak — Johannes Gutenberg Universität Mainz, Institut f. Physik, Staudingerweg 7, 55128 mainz

Using Microscopy and static light scattering we determined the phase behavior and the occurring microstructures in a binary mixture of charged colloidal spheres with a size ratio of $\Gamma = 0.57$. The eutectic point is observed at a molar fraction of small particles $x_S = 0.8$ and total number densities of about $(30-40)\mu m^{-3}$, which is one, resp. two orders of magnitudes larger than the freezing densities of the pure small (S) resp. large (L) particles. In the present system density and composition fluctuations needed for (heterogeneous) nucleation of coexisting S- and L-crystals are coupled to differential sedimentation. Under fluid-solid coexistence conditions we observe the majority component of small particles to form faceted crystals (at $x_S = 0.9-0.97$) in the bulk of the samples, while the minority component of large particles forms a network of rough, bottom wall based, hemispherical cap crystals. No faceting is observable for other mixing ratios and for the pure component systems at coexistence conditions. At number densities well above the eutectic density a correlated solidification scenario is observed, in which both species form a mosaic with intersection facets of columnar crystals growing from the container walls. Similarities and differences to morphologies observed for metallic eutectics are discussed.

CPP 47.3 Fri 11:00 ZEU 160

Influence of Pattern Height on the Dewetting of Polymer Thin Films on Topographically Patterned Substrates — •Rabibrata Mukherjee — Departmnet of Chemical Engineering, IIT Kharagpur, Pin 721302, India

Controlled dewetting of thin polymer films on physically patterned complex surfaces produces variety of ordered meso-scale structures like arrays of ordered droplets, membranes etc. In contrast to dewetting of a thin polymer film on a defect free surface, which occurs spontaneous amplification of capillary surface waves, on a topographically patterned substrate dewetting is engendered by a rapid rupture of the film along the contours of the substrate pattern. The subsequent morphological evolution is therefore strongly influenced by the geometry of the substrate pattern. We show that the final structure depends not only on a commensuration between the initial film thickness and substrate pattern geometry, but is also influenced significantly by the film preparation condition. For example, when the film is directly spin coated on to the topographically patterned substrate, the resulting morphology is strongly influenced by the height of the features, in addition to their periodicity. In contrast, when a film of uniform thickness is transferred and then dewetted on the patterned substrate, the influence of height variation of the substrate features is rather limited. In the latter case, the initial conformation of the film with respect to the underlying substrate dominates the positioning of the dewetted droplets.

CPP 47.4 Fri 11:15 ZEU 160

Heterogeneous nucleation at a flat wall in a hard sphere colloidal model system — •Markus Franke und Hans Joachim Schöpe — Johannes Gutenberg-Universität Mainz, Institut für Physik, Staudinger Weg 7, 55099 Mainz

The hard sphere system is the simplest model system studying crystallization. Experimentally it can be realized using steric stabilized colloids. We studied the kinetics of homogeneous and wall induced heterogeneous nucleation in colloidal hard spheres as function of super saturation. Using light scattering and microscopy we can determine the time trace of the absolute crystal volume, the crystal size as well as the nucleation rate density. We show that homogeneous nucleation is mediated by precursors, while wall induced crystallization is induced by a layering at the cell wall leading to a different crystallization kinetics of wall and bulk crystals.

CPP 47.5 Fri 11:30 ZEU 160

Heterogeneous nucleation in colloids with spherical interaction — •Hans Joachim Schöpe — Johannes Gutenberg-Universität Mainz, Institut für Physik, Staudinger Weg 7, 55099 Mainz

Understanding the process that drives a metastable liquid or fluid to the crystal state is still a challenging issue for condensed matter physics. One promising approach to the problem is offered by the study of colloidal suspensions of particles with spherical interaction. Identifying the colloidal particles as macroatoms close analogies to atomic systems are observed. Colloids offer, compared to true atomic systems, a number of advantages. On one side, the interaction potential in colloidal macroatoms can be finely controlled and tuned by the experimentalists, by engineering the properties of the particles, and/or changing the characteristics of the dispersing medium. On the other side, typical length and time scales of colloidal systems allow time-resolved observations of solidification with easily manageable experimental techniques such as microscopy and light scattering. In particular it is possible to study the competition between homogeneous and heterogeneous nucleation induced by container walls or by seeds. The results of various experiments on colloids with various interaction will be presented together with their analysis within the framework of classical theory.

CPP 47.6 Fri 11:45 ZEU 160

Nucleation in a colloidal hard sphere system using density functional theory — •Andreas Härtel[1], Martin Oettel[2], and Hartmut Löwen[1] — [1]Institut für Theoretische Physik II: Weiche Materie, Heinrich-Heine-Universität Düsseldorf, Germany — [2]Institut für Physik, Johannes-Gutenberg-Universität Mainz, Germany

The fluid-solid interface in a system of hard spheres is studied by density functional theory. For this purpose we employ a free minimization [1] of the White Bear II fundamental measure density functional [2] and perform a comparative study using Monte Carlo simulations [3]. Both the bulk freezing transition and the structure and thermodynamics of the hard sphere fluid-solid interface are discussed and compared to simulation data.

[1] R. Ohnesorge et al, Phys. Rev. E 50, 4801 (1994).

[2] H. Hansen-Goos and R. Roth, J. Phys.: Condens. Matter 18, 8413 (2006).

[3] M. Oettel et al, Phys. Rev. E 82, 051404 (2010).

CPP 47.7 Fri 12:00 ZEU 160

DDFT calibration and investigation of an anisotropic phase-field crystal model — •Muhammad Ajmal Choudhary[1], Daming Li[1], Heike Emmerich[1], and Löwen Hartmut[2] — [1]Lehrstuhl

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The anisotropic phase field crystal (APFC) model recently proposed by Prieler et al.[J. Phys.:Condens. Matter 21, 464110 (2009)] is derived from microscopic density functional theory for anisotropic particles with fixed orientation. Further its morphology diagram is explored to demonstrate its capacity to model structures beyond those captured by the simplistic phase field crystal (SPFC) equations originally introduced by Elder et al. [Phys. Rev. Lett. 88, 245701 (2002)]. In particular we investigated the influence of anisotropy and undercooling on the process of nucleation and microstructure formation from atomic to the microscale. To that end numerical simulations were performed varying those dimensionless parameters which represent anisotropy and undercooling in our APFC model. The results from these numerical simulations are summarized in terms of a morphology diagram of the stable state phase. These stable phases are also investigated with respect to their kinetics and characteristic morphological features.

CPP 47.8 Fri 12:15 ZEU 160

Homogeneous and heterogeneous nucleation in phase field crystal models studied by the simplified string method

— Rainer Backofen and •Axel Voigt — Institut für Wissenschaftliches Rechnen, TU Dresden, Dresden, Germany

The phase field crystal (PFC) model is an approximation to the classical density functional theory of liquid and solids. In recent years it has been shown that with some modification PFC gives decent descriptions of colloids or metals. We investigate a PFC model for homogeneous and heterogeneous nucleation at unstructured walls. We will show, that in order to include wall interactions the density in the classical PFC model has to be bound from below. This can be achieved by introducing a density dependent mobility or an additional density dependent potential. Thus, the wall interaction maybe tuned from wetting to non-wetting conditions. The nucleation process is analyzed directly by the Simplified String Method and the minimal stable nucleus is defined. We see that the nucleation path deviates from that one assumed in classical nucleation theory. In particular the minimal stable nucleus is not in the bulk state. Thus, the nucleation barrier is decreased.

CPP 47.9 Fri 12:30 ZEU 160

Curvature Dependence of Surface Free Energy of Liquid Drops and Bubbles

— B. J. Block[1], S. K. Das[2], •M. Oettel[1,3], P. Virnau[1], and K. Binder[1] — [1]Institut für Physik, Universität Mainz, Germany — [2]Theoretical Sciences Unit, Jawaharlal Nehru Centre for Advanced Scientific Research, Bangalore, India — [3]Material- und Prozesssimulation, Universität Bayreuth, Germany

We study the excess free energy due to phase coexistence of fluids by Monte Carlo simulations using successive umbrella sampling in finite boxes with periodic boundary conditions, allowing for stable spherical/cylindrical droplets or flat interface configurations [1]. Both the vapor-liquid phase coexistence of a simple Lennard-Jones fluid and the coexistence between A-rich and B-rich phases of a symmetric binary (AB) Lennard-Jones mixture are studied. For the surface tension $\gamma(R)$ of droplets/bubbles in the vapor-liquid case, we find for $R \to \infty$ the leading order (Tolman) correction for droplets has sign opposite to the case of bubbles, consistent with the Tolman length being independent on the sign of curvature. For the symmetric binary mixture the expected non-existence of the Tolman length is confirmed. In all cases and for a range of radii R relevant for nucleation theory, $\gamma(R)$ deviates strongly from $\gamma(\infty)$ which can be accounted for by a term of order $\gamma(\infty)/\gamma(R) - 1 \propto R^{-2}$. Comparison with density functional theory in the vapor-liquid case yields agreement in the overall behavior of $\gamma(R)$ as well as in the sign and magnitude of the Tolman length.

[1] B. J. Block, S. K. Das, M. Oettel, P. Virnau, and K. Binder, J. Chem. Phys. **133**, 154702 (2010).