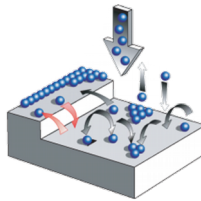# *Non-Equilibrium Dynamics and Structure Formation*

SoftComp Workshop, April 3-5 2017
Schloss Hohentübingen, Tübingen

organized by:

Martin Oettel
Universität Tübingen

Tanja Schilling
Université du Luxembourg / Universität Freiburg

Frank Schreiber
Universität Tübingen

EBERHARD KARLS
UNIVERSITÄT
TÜBINGEN

Institut für Angewandte Physik

1 Introduction

The goal of this SoftComp topical workshop, which is co-funded by the SoftComp Network, is to bring together researchers from inside and outside the network to establish a clear view of the state of the art of this hot topic and formulate a strategy to bring different strands of research together. We are also dedicated to promoting strong interactions and exchange between experiments, computer simulations and theory, with a great potential for synergy effects, and to promoting women researchers and young scientist.

The format consists of invited talks as well as a limited number of oral presentations of "hot topics" and poster presentations. Young academics are especially encouraged to participate and present their work.

Scope of the Workshop

Many forms of soft condensed matter in equilibrium have been the subject of research for many years. While there are of course still many open questions, our understanding of the general principles and concepts of how to address equilibrium properties, both in terms of theory as well as experiments, has reached a certain level of maturity. Non-equilibrium properties are far less well understood, probably also due to the additional challenges associated with their theoretical description as well as their experimental study. Moreover, the broad spectrum of length and time scales characteristic of soft matter is potentially further expanded if dynamic and kinetic issues are being considered. On the other hand, it is undisputable that non-equilibrium effects and phenomena play a key role in many areas, inter alia in for growth and structure formation, which also implies a significant technological relevance. These issues have recently moved this hot topic more and more into focus of research. Importantly, there are rather different angles from which the topic is addressed, including from the theoretical side,

- rate equation approaches
- perturbation ideas for small deviations from equilibrium
- targeted simulations vs. analytical theory
- non-equilibrium work relations and fluctuation theorems

as well as from the experimental side,

- different strategies to characterize structure formation and dynamics

- imaging techniques for morphological features
- non-invasive scattering techniques for and properly averaged information

each of which sensitive to different windows of time and length scales.

In addition to these general points characterizing the field, also recent and more specific developments such as structure formation in colloidal systems or bio-related systems shall be considered, including the very vibrant field of active soft matter featuring many aspects of structure formation. An important issue will be on conceptual understanding and the comparison between (relatively established) "atomic" systems for growth and (far less established) structure formation in more complex molecular or colloidal systems, i.e. the identification of analogies and differences of system-specific and system-independent features.

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Password: ln1ln4

eduroam is also available!

2 Program

Day 1 – Monday, Apr 3

Session I
- 13:00-14:00 – Registration
- 14:00-14:15 – Welcome and Introduction
- 14:15-15:00 – **Joachim Krug**
  A bird's eye view on nonequilbrium surface morphologies

15:00-15:30 – Coffee Break

Session II
- 15:30-16.15 – **Peter G. Vekilov**
  Metastable mesoscopic phases in protein aggregates
- 16:15-16:45 – **Fajun Zhang**
  Two-step nucleation in protein crystallization
- 16:45-17:15 – **Uwe Thiele**
  Crystallisation fronts and (travelling) localised states in (active) phase-field crystal models
- 17:15-17:45 – **Walter Tewes**
  Propagating Dewetting Fronts in Passive and Active Geometries

17:45-19:00 – Discussion with beer and wine (Turmzimmer)

Day 2 – Tuesday, Apr 4

Session III
- 9:00-9:45 – **Sabine Klapp**
  Non-equilibrium self-assembly and growth of anisotropic particles at surfaces
- 9:45-10:05 – **Miriam Klopotek**
  Monolayers of hard rods: A soft matter model for non-equilibrium growth
- 10:10-10:30 – **Svetlana Gurevich**
  Dip-coating with prestructured substrates: transfer of simple liquids and Langmuir-Blodgett monolayers

10:30-11:00 – Coffee Break

Session IV
- 11:00-11:45 – **Stefan Kowarik**
  Real-time experiments, nucleation theory and the issue of molecular attempt frequencies
- 11:45-12:05 – **Alexander Hinderhofer**
  Anomalous Roughness Evolution of Organic Mixed Films
- 12:10-12:30 – **Florian von Wrochem**
  CMOS compatible, ultra-robust metal-terpyridine thin-films

12:30-13:45 – Lunch

13:45   – **Workshop Photo**

Session V
- 14:00-14:45 – **Andreas Heuer**
  Structure formation of molecules on substrates after deposition
- 14:45-15:30 – **Thomas Speck**
  Phase transformation and nucleation in driven systems

15:30-16:00 – Coffee Break

Session VI
- 16:00-16:45 – **Olivier Pierre-Louis**
  Solid-state wetting and dewetting
- 16:45-17:05 – **DomenicoTruzzolillo**
  Nonequilibrium Interfacial Tension in Simple and Complex Fluids
- 17:10-17:30 – **Paolo Malgaretti**
  A simple entropy-based model that captures polymer translocation across varying section channels
- 17:30-18:15 – **Tobias Kraus**
  Order and disorder in nanoparticle superstructures

18:45   – **Conference dinner at Ludwigs**

Session VII
- 9:00-9:45 – **Marjolein Dijkstra**
  Unjamming glasses, removing grain boundaries with active colloids, and mechanical and chemical equilibrium in mixtures of active and passive colloids
- 9:45-10:05 – **Hans-Joachim Schöpe**
  Heterogeneous nucleation and growth in colloidal model systems
- 10:10-10:30 – **Josh T Berryman**
  An Amyloid Aggregation Process with Unexpected Dependence on Sequence, Concentration and pH

10:30-11:00 – Coffee Break

Session VIII
- 11:00-11:45 – **Marisol Ripoll**
  Self-assembly of thermophoretic active colloids
- 11:45-12:30 – **Walter Zimmermann**
  Stripe and traveling wave patterns in finite systems
- 12:30-12:45 – Closing remarks

3 Abstracts

A bird's eye view on nonequilbrium surface morphologies

Joachim Krug[1]
[1] Institut für Theoretische Physik, Universität zu Köln

Nonequilibrium processes at surfaces give rise to a variety of structures that range from regular arrays of mounds, pits or ripples to self-affine fractals with nontrivial scaling properties. The quantitative description of these phenomena requires theoretical approaches that link molecular to mesoscopic and macroscopic scales. The talk will review some of the concepts that have been developed mainly in the context of homoepitaxial crystal growth, with a focus on recent developments and the applicability to soft-matter systems.

Metastable mesoscopic phases in protein aggregates

Peter Vekilov[1]
[1] Department of Chemical and Biomolecular Engineering, University of Houston

Protein-rich clusters of steady submicron size and narrow size distribution exist in protein solutions in apparent violation of the classical laws of phase equilibrium. Even though they contain a minor fraction of the total protein, recent evidence indicates that they may serve as essential precursors for the nucleation of ordered solids such as crystals, sickle-cell hemoglobin polymers, and amyloid fibrils. The cluster formation mechanism remains elusive. We use proteins that differ in their structural and chemical properties and explore the response of cluster populations to the electrostatic forces, which govern numerous biophysical phenomena, including crystallization and fibrillization. We tune the strength of intermolecular electrostatic forces by varying the solution ionic strength I and pH and find that despite the weaker repulsion at higher I and pH, the cluster size remains constant. Cluster responses to the presence of urea and ethanol demonstrate that cluster formation is controlled by hydrophobic interactions between the peptide backbones, exposed to the solvent after partial protein unfolding that may lead to transient protein oligomers. These findings reveal that the mechanism of the mesoscopic clusters is fundamentally different from those underlying the two main classes of ordered protein solid phases, crystals and amyloid fibrils, and partial unfolding of the protein chain may play a significant role.

Two-step nucleation in protein crystallization

Fajun Zhang[1], B. Sohmen[1], M. Mikorski[1], R. Schweins[2],F. Schreiber[1]

[1] Institut für Angewandte Physik, Universität Tübingen
[2] ILL, Grenoble, France

Classical nucleation theory (CNT) established about 90 years ago has been very successful in many research fields and continues to be the most commonly used theory in describing the nucleation process. One of the most important assumptions of CNT is that the nucleation process is described by one reaction coordinate and all order parameters proceed simultaneously. Recent studies in experiments, computer simulations and theory have revealed nonclassical features of the early stage of nucleation. In particular, the decoupling of order parameters involved during a fluid-to-solid transition leads to the so-called two-step nucleation mechanism, in which a metastable intermediate phase (MIP) exist between the initial supersaturated solution and the final crystals. Depending on the exact free energy landscapes, the MIPs can be a high density liquid phase, mesoscopic clusters, or a pre-ordered state. In this talk, I will first discuss the various scenarios of the nonclassical nucleation pathways in colloid and protein solutions based on the established phase diagram. I will then show one example from our recent study of a two-step crystallization in bovine β-lactoglobulin solutions in the presence of tri-valent salts.

Crystallisation fronts and (travelling) localised states in (active) phase-field crystal models

Uwe Thiele[1]
[1] Institut für Theoretische Physik, Universität Münster

The conserved Swift-Hohenberg equation [or Phase-Field-Crystal (PFC) model] provides a simple microscopic continuum description of the thermodynamic transition from a fluid to a crystalline state [1]. First, we present the bifurcation structure of localised states which are found in the coexistence region between the homogeneous and structured phases [2]. Second, we analyse crystallisation fronts and show that 1d front speeds may be obtained via a marginal stability criterion [3]. The relation of quench depth, front speed, created disorder and subsequent aging is briefly discussed. The thermodynamic PFC model can be combined with the Toner-Tu theory for self-propelled particles to obtain a simple model for crystallisation (swarm formation) in active systems [4]. Within the resulting active PFC model, resting and traveling crystals can be identified. In the linear regime, we give analytical expressions

for the transitions from the liquid state to both types of crystals [5]. In the nonlinear regime, again we describe a variety of localized clusters besides spatially extended crystals. Numerical continuation is applied to follow resting and traveling localized states while varying the activity and the mean concentration. Finally, we provide a semi-analytical criterion for the onset of motion in the nonlinear regime, that corresponds to a drift pitchfork bifurcation [6].

[1] H. Emmerich, H. Löwen, R. Wittkowski, T. Gruhn, G. Toth, G. Tegze, and L. Granasy, *Adv. Phys.* **61**, 665 (2012).
[2] U. Thiele, A. J. Archer, M. J. Robbins, H. Gomez, and E. Knobloch, *Phys. Rev. E.* **87**, 042915 (2013).
[3] A. J. Archer, M. J. Robbins, U. Thiele, and E. Knobloch, *Phys. Rev. E* **86**, 031603 (2012).
[4] A. M. Menzel and H. Löwen, *Phys. Rev. Lett.* **110**, 055702 (2013).
[5] A. I. Chervanyov, H. Gomez and U. Thiele, *EPL* **115**, 68001 (2016).
[6] L. Ophaus, S. V. Gurevich and U. Thiele, in preparation (2016).

Propagating Dewetting Fronts in Passive and Active Geometries

Walter Tewes[1]
[1] Institut für Theoretische Physik, Universität Münster

We study the dynamics and the patterns obtained through the propagation of dewetting fronts into unstable thin liquid films both on resting noninclined substrates and in a dip-coating geometry in 1D and 2D. For the rather simple case of resting substrates in 1D, we study the transition from steadily propagating dewetting fronts with simple periodic droplet pinch-off events for very small film heights to the case of the propagation of a dewetting rim for larger film heights. It is shown that for a small film height, the front propagation speed is well described by a marginal stability analysis. In the transition region to the case of the dewetting rim, secondary oscillations occur during the pinch-off events, leading to more complex periodic deposition patterns. In order to control the deposition patterns and suppress transversal instabilities of the 1D dewetting fronts, we furthermore study the dewetting in the meniscus region in a dip-coating geometry. We show that in this case, well controlled stripe-like depositions can be obtained in 2D through the meniscus-directed instability.

Non-equilibrium self-assembly and growth of anisotropic particles at surfaces

Sabine Klapp[1]

[1] Institut für Theoretische Physik, Technische Universität Berlin

Anisotropic pair interactions of steric, chemical or electrostatic origin are known to strongly influence a system's equilibrium phase behavior and self-assembly. In the present talk I will discuss recent results for the impact of anisotropy on a typical non-equilibrium situation, that is, surface growth at constant flux. Understanding anisotropic surface growth is an emerging, yet very challenging field which is so far still in its infancy. The first part of my talk concerns the growth of a monolayer of rod-shaped particles with quadrupolar interactions at a patterned surface. This model is inspired by a technologically important hybrid material system of p-sexiphenyl (6P) molecules at a zinc oxide (1010) surface. Using kinetic Monte Carlo (kMC) simulations of a coarse-grained model we explore the sub-monolayer growth. Due to the competition of the various anisotropic interactions, on one side, and that between internal time scales, on the other side, the resulting state diagram reveals a wealth of features including lying and standing phases, re-entrant growth and a critical adsorption rate. The latter can be explained in terms of a rate equation approach. In the second part I will discuss kMC results for more general models of anisotropic growth involving spherical units with anisotropy in terms of either the interparticle interactions or the diffusion along the surface.

Monolayers of hard rods: A soft matter model for non–equilibrium growth

Miriam Klopotek[1], H. Hansen–Goos[2], M. Dixit[3], T. Schilling[3], F. Schreiber[1], and M. Oettel[1]

[1] Institut für Angewandte Physik, Universität Tübingen
[2] Institut für Theoretische Physik, Universität Tübingen
[3] Physics and Materials Sciences Research Unit, Université du Luxembourg

We model purely hard rods at the vicinity of a substrate both in a lattice model, where position and orientation of rods are restricted, and in a continuum model with hard spherocylinders, where both position and orientation are continuous variables. In a systematic, two–part study combining analytic theory and simulation we have determined the properties of these monolayers in equilibrium [*J. Chem. Phys.* **145**, 074902 (2016)], as well as in non–equilibrium under conditions of

monotonic growth by means of random deposition [*J. Chem. Phys* **146**, 084903 (2017)]. Special attention is given to the orientational ordering of the rods: our generic model showcases the "standing–up" transition found in molecular thin film growth, e.g. with organic molecules, which are rod–like in shape, both in experiment [1] and in recent simulations [2]. The transition from 'lying' to 'standing' is thermodynamically continuous with hard rods both on the lattice and in the continuum. It remains so for a strong, orientation–dependent attractive substrate; however, this introduces spatio–temporal correlations during non–equilibrium growth: the self–assembly becomes sensitive to the diffusion coefficient of the rods at the substrate. Details of monolayer growth are relevant for the cases of Frank–van–der–Merwe (layer–by–layer) and Stranski–Krastanov (islands–on–layers) growth: in ongoing simulations of multi–layer growth, we recover the standing–up transition in the monolayer regime before further layers self–assemble. Slow monolayer growth is characterized by dynamic observables approaching a quasi–equilibrium master curve. Both models—lattice and continuum—in fact reveal qualitatively the same ensemble behavior during highly non–equilibrium growth, differing only via their respective equation–of–states. We discuss how mapping dynamic quantities between models means matching microscopic and macroscopic time–scales non–trivially. Our equilibrium studies employ Monte Carlo (MC) simulations in the Grand Canonical ensemble, classical lattice Density Functional Theory (DFT) for lattice systems [3], and continuum MC simulations; in non–equilibrium, kinetic Monte Carlo, dynamic DFT on the lattice, and dynamic MC for Brownian dynamics in the continuum were employed.

[1] S. Kowarik et al., *Phys. Rev. Lett.* **96**, 125504 (2006).
[2] N. Kleppmann and S. H. L. Klapp, *Phys. Rev. B.* **94**, 241404 (2016).
[3] L. Lafuente and J. A. Cuesta, *Phys. Rev. Lett.* **93**, 130603 (2004).

Dip-coating with prestructured substrates: transfer of simple liquids and Langmuir-Blodgett monolayers

Svetlana Gurevich[1], Markus Wilczek[1], Uwe Thiele[1]
[1] Institut für Theoretische Physik, Universität Münster

When a plate is withdrawn from a liquid bath, either a static meniscus forms in the transition region between the bath and the substrate or a liquid film of finite thickness is transferred onto the moving substrate. If the substrate has a prestructure consisting of e.g., stripes of different wettabilities, the meniscus can show a complex dynamic behavior. Here we study the dynamics of a dragged meniscus occurring for striped prestructures with different orientations using a thin film model.

Numerical continuation is used to obtain steady free surface profiles and corresponding bifurcation diagrams in the case of substrates with different homogeneous wettabilities, whereas direct numerical simulations are employed in the case of the various striped substrates.

Real-time experiments, nucleation theory and the issue of molecular attempt frequencies

Stefan Kowarik[1]
[1] Institut für Physik, Humboldt Universität zu Berlin

We present our recent results on real-time observation of nucleation and multilayer thin film growth. In particular we employ X-ray reflectivity (XRR), small angle scattering (GISAXS) and atomic force microscopy (AFM) techniques for following nucleation processes, island densities and roughness in situ and in real-time. We demonstrate the accuracy of the fast, in situ GISAXS technique by benchmarking it against often AFM technique. For the molecules PTCDI-C8, 6P and 6PF2 we determine the scaling constants, energy parameters and attempt frequencies needed for a description of film growth with nucleation theory. Interestingly, we find very large attempt frequencies in the 10E20 range. Further, the attempt frequency changes by 7 orders of magnitude for a slight change of the 6P molecule through fluorination. Both these findings indicate that the attempt frequency cannot necessarily be interpreted as a timescale, but other effects such entropic contributions enter as well. As an outlook we will demonstrate how experimental in situ methods and theoretic understanding can be used to implement novel control strategies for molecular self-assembly and growth. We demonstrate that the non-standard control schemes of modulated substrate temperature, chemical tuning, and laser control work for molecular growth.

Anomalous Roughness Evolution in Organic Mixed Film Growth

Alexander Hinderhofer[1], A. Gerlach[1], M. Oettel[1], F. Schreiber[1]
[1] Institut für Angewandte Physik, Universität Tübingen

The surface morphology and roughness of thin films and crystals depend on competing mechanisms, which either roughen or smooth the film surface during growth. Important roughening mechanisms, are kinetic roughening based on shot noise and roughening due to mound

growth, which is facilitated by reduced interlayer transport, often associated with a step edge. While these issues have been well studied for growth of simple atomic species, comparatively little is known about organic systems. In crystalline organic thin film growth high step edge barriers of ~0.7 eV have been reported. Theoretical models show that by neglecting the step edge barrier roughening would be strongly reduced. These findings suggest that mound growth, induced by high step edge barriers, often dominates the roughness evolution for crystalline organic thin films. The situation is more complex for growth of organic-organic mixtures, which are important for many device applications like organic photovoltaic cells or organic light emitting diodes. We use in situ x-ray reflectivity and complementary atomic force microscopy to monitor crystallinity and roughness evolution during growth of organic binary mixtures of several compounds, i.e. pentacene (PEN), perfluoropentacene (PFP), diindenoperylene (DIP) fullerene (C60). A general trend of reduced roughness in the mixed films compared to the pure materials is observed. We will discuss this roughness evolution in relationship to the in-plane crystallinity of the thin films and will show that the growth behavior can be rationalized by a, compared to homoepitaxy, lowered step edge barrier for lower in-plane crystallinity. [1,2]

[1] A. Hinderhofer, A. Gerlach, S. Kowarik, F. Zontone, J. Krug and F. Schreiber, *EPL* **91**, 56002 (2010)
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CMOS compatible, ultra-robust metal-terpyridine thin-films

Florian von Wrochem[1], M. Rampi[2], W. Wenzel[3]
[1] Materials Science Laboratory, Sony Deutschland GmbH, Stuttgart
[2] Dipartimento di Chimica Università di Ferrara
[3] Institute of Nanotechnology, Karlsruhe Institute of Technology

Considerable efforts have been undertaken within the past decades to shift organic-based thin-film devices from basic research to the application level. A major obstacle is given by the thermal deposition of metal electrodes, which remained elusive due to the damage and the electrical shorts experienced by the fragile molecular layers. Here, we show that large area molecular junctions of outstanding electronic properties and robustness can be realized using densely packed molecular wires consisting of FeII-terpyridine complex oligomers. Surprisingly, these oligomer-based devices are stable for over 2 years under regular current-voltage cycling, withstanding a wide range of

temperatures (150-360 K) and applied voltages (3 V). Electrical studies in conjunction with ab-initio calculations reveal that charge transport (i) occurs via electron (hopping) conduction and is limited by the charge injection through a Shottky barrier (0.72 eV), following Richardson-Schottky injection.

Structure formation of molecules on substrates after deposition

Andreas Heuer[1]

[1] Institut für Physikalische Chemie, Universität Münster

Via vacuum deposition of organic molecules, often on pre-patterned substrates, one can achieve to generate structures in and out of equilibrium with high accuracy. After a general introduction I will highlight recent theoretical insight into interesting phenomena, mainly obtained in collaboration with our experimental partners. (1) Phase behavior of 2D layers of adenine derivatives on non-patterned substrates. Here one observes an anomalous non-equilibrium behavior, i.e. by approaching equilibrium conditions upon decreasing the flux, the phase behavior departs stronger from the equilibrium phase. (2) Structure formation on gold stripes. Three types of structure formation are observed when comparing different molecules. The different scenarios can be understood from kinetic Monte Carlo simulations with appropriately chosen model parameters. In particular, one observes specific instabilities which give rise to the formation of bulges. (3) Structure formation on substrates which are pre-patterned with a square grid. Under specific experimental/simulation conditions arrangements of molecular clusters with high regularity can be achieved. The mechanisms of the high regularity are identified.

Phase transformation and nucleation in driven systems

Thomas Speck[1]

[1] Condensed Matter Theory Group, Johannes Gutenberg Universität Mainz

The making of a material necessarily is a non-equilibrium process.Moreover, typically it involves phase transformations, which realistically occur under external stresses. I will discuss numerical results for two steadily driven model systems that undergo a discontinuous first order transition upon changing the volume: repulsive particles in shear flow and active Brownian particles in two dimensions. These model systems are paradigms for two different routes to steady driving: through an external field (here shear flow) or, in the second

case, autonomous units (³particles²) that are constantly converting energy into directed motion.

Solid-state wetting and dewetting
Olivier Pierre-Louis[1]
[1] Institut Lumière Matière, Université Claude Bernard Lyon 1

At the nanoscale, the morphological evolution of solid films and islands under annealing is strongly influenced by wetting properties. Inspired by analogies with recent advances in the wetting behavior of liquids, we explore two situations where solid-state wetting plays a crucial role. In a first part, we discuss the dewetting dynamics of a thin solid film based on 2D Kinetic Monte Carlo (KMC) simulations and analytical models. We focus on the role of the faceting of the dewetting rim, which changes the asymptotic behavior of the dewetting velocity. In addition, we analyze the instability of the dewetting front, which leads to the formation of fingers. In a second part, we will present some results on the wetting statics and dynamics of islands (or nanoparticles) on surface topographical structures with a large aspect ratio, such as pillars or trenches using 3D KMC simulations including elastic effects. We show that elasticity induces novel states such as asymmetric and partially impaled configurations.

Nonequilibrium interfacial tension in simple and complex fluids
Domenico Truzzolillo[1]
[1] Universität Montpellier

Interfacial tension between immiscible phases is a well-known phenomenon, which manifests itself in everyday life, from the shape of droplets and foam bubbles to the capillary rise of sap in plants or the locomotion of insects on a water surface. More than a century ago, Korteweg [1] generalized this notion by arguing that stresses at the interface between two miscible fluids act transiently as an effective, nonequilibrium interfacial tension, before homogenization is eventually reached. In spite of its relevance in fields as diverse as geosciences, polymer physics, multiphase flows, and fluid removal, experiments and theoretical works on the interfacial tension of miscible systems are still scarce, and mostly restricted to molecular fluids. This leaves crucial questions unanswered, concerning the very existence of the effective interfacial tension, its stabilizing or destabilizing character, and its dependence on the fluid's composition and concentration gradients. We

present an extensive set of measurements on miscible complex fluids that demonstrate the existence and the stabilizing character of the effective interfacial tension [2,3], unveil new regimes beyond Korteweg's predictions, and quantify its dependence on the nature of the fluids and the composition gradient at the interface. We introduce a simple yet general model that rationalizes nonequilibrium interfacial stresses to arbitrary mixtures, beyond Korteweg's small gradient regime [3], and show that the model captures remarkably well both our new measurements and literature data on molecular and polymer fluids. Finally, we briefly discuss the relevance of our model to a variety of interface-driven problems, from phase separation to fracture, which are not adequately captured by current approaches based on the assumption of small gradients [3].

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[2] D. Truzzolillo, S. Mora, C. Dupas and L. Cipelletti, Off-Equilibrium Surface Tension in Colloidal Suspensions, *Phys. Rev. Lett.* **112**, 128303 (2014).
[3] D. Truzzolillo, S. Mora, C. Dupas and L. Cipelletti, Nonequilibrium Interfacial Tension in Simple and Complex Fluids, *Phys Rev. X* (In Press – 2016).

A simple entropy-based model that captures polymer translocation across varying section channels

Paolo Malgaretti[1]
[1] Max Planck Institute for Intelligent Systems, Stuttgart

We study the translocation of polymers across varying-section channels. Using systematic approximations, we derive a simplified model that reduces the problem of polymer translocation through varying-section channels to that of a point-like particle under the action of an effective potential. Such a model allows us to identify the relevant parameters controlling the polymer dynamics and, in particular, their translocation time. By comparing our analytical results with numerical simulations we show that, under suitable conditions, our model provides reliable predictions of the dynamics of both Gaussian and self-avoiding polymers, in two- and three-dimensional confinements. Moreover, both theoretical predictions, as well as Brownian dynamic results, show a non-monotonous dependence of polymer translocation velocity as a

function of polymer size, a feature that can be exploited for polymer separation.

Order and disorder in nanoparticle superstructures
Tobias Kraus[1]

[1] INM - Leibniz-Institut für Neue Materialien gGmbH, Saarbrücken

We study the behavior of nanoparticles with uniform diameters between 1 and 100 nm that freely move in a liquid. Such particles have metal, semiconductor or insulator cores that are covered with organic molecules. They form superstructures when their concentration increases or when their interaction becomes more attractive. The geometry of the superstructures is technologically relevant: for example, dense crystalline packing may be desirable for an optical coating, while a loose but percolating mesh may lead to transparent electrodes. Here, I will discuss how the structure of the superstructures depends on temperature, core material, molecular structure of the shell, and the confinement of the particles. First, consider the crystalline order of the superstructures. For sufficiently narrow size distribution, a crystal is the minimum free energy configuration of the particles. But if the short-range potential is very attractive, the particles do not reach this minimum and form dense amorphous or loose superstructures. An extreme case is the fractal "flocs" that form when electrostatically repulsive particles are rapidly "quenched" by removing charge as first analyzed by Dave Weitz. I will show assembly for intermediate potentials and discuss the different rolse of long- and short-range interactions: we find that it is possible to rapidly form high-quality supercrystals if the short-ranged "tribology" of the particles allows for it. Confinement changes the superstructures. Assembly in small droplets leads to "supraparticles" that are surprisingly similar to atomic clusters of noble gases or certain metals. Such clusters provide the maximal possible number of nearest neighbors while minimizing "dangling bonds" on their surfaces, and they strongly deviate from crystal sections for small numbers of particles. Their structure is known for the case of uniform particles. We collaborate with Tanja Schilling to predict the structures that will form in mixtures of particles. Simulations and experiments show that binary supercrystals, "core-shell"-structures, and "Janus" arrangements with phase separation are possible. Some of them are minimum-energy configurations, but many seem to form through kinetic pathways that reflect the different stability of the components.

Unjamming glasses, removing grain boundaries with active colloids, and mechanical and chemical equilibrium in mixtures of active and passive colloids

Marjolein Dijkstra[1]
[1] Universität Utrecht

In this talk, we present a simulation study of self-propelled hard spheres, for which we find that with increasing activity the relaxation dynamics can be sped up by orders of magnitude [1]. As a consequence, the glass transition shifts to higher packing fractions upon increasing the activity, allowing the study of sphere packings with fluid-like dynamics close to random close packing. We apply these findings to devitrify glassy systems consisting of mixtures of active and passive hard spheres [2]. We show that the crystallization of hard-sphere glasses can be dramatically promoted by doping the system with small amounts of active particles. Our results suggest a novel way of fabricating crystalline materials from (colloidal) glasses. This is particularly important for materials that get easily kinetically trapped in glassy states, and the crystal nucleation hardly occurs. In addition, we show that grain boundaries can be removed in a polycrystalline material by the addition of active particles [3,4]. Finally, we study fluid-solid phase coexistence of mixtures of active and passive particles and show that the two coexisting phases are in mechanical and chemical equilibrium.

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[2] R. Ni, M.A. Cohen Stuart, M. Dijkstra and P.G. Bolhuis, *Soft Matter* **10**, 6609 (2014).
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[4] B. van der Meer, M. Dijkstra and L. Filion, *Soft Matter* **12**, 5630-5635 (2016).

Heterogeneous nucleation and growth in colloidal model systems

Hans-Joachim Schöpe[1]
[1] Institut für Angewandte Physik, Universität Tübingen

Understanding crystal nucleation and growth is of great interest in condensed matter physics and material science. Especially at low metastabilities crystal nucleation is dominated by heterogeneous nucleation on container walls or impurities and the competition between wall and bulk crystallization determines the microstructure of the

polycrystal. Notwithstanding its relevance heterogeneous nucleation is not well understood. One promising approach to the problem is offered by the study of colloidal suspensions. In colloids the particle interactions can be experimentally adjusted to include short (hard spheres, HS) and long ranged (charged spheres, CS) repulsions. We studied the kinetics of homogeneous and heterogeneous nucleation and further crystal growth in different colloidal model systems with various interaction as function of super saturation. Using light scattering as well as microscopy techniques a comprehensive analysis of the crystallization process was obtained. We analyzed the heterogeneous nucleation process on flat smooth walls as well as on flat structured walls with various lattice spacing. Furthermore we induced heterogeneous nucleation by controlled addition of spherical seeds modifying the crystallization kinetics and the microstructure evolution in close analogy to inoculation in metallic systems.

An Amyloid Aggregation Process with Unexpected Dependence on Sequence, Concentration and pH

Joshua T. Berryman[1], N. Reynolds[1], J. Adamcik[1], R.Mezzenga[1]
[1] Physics and Materials Sciences Research Unit, Université du Luxembourg

We investigate the aggregation kinetics of anisotropic interacting bodies, and describe a regime in which a small shift in the balance of attraction strengths along two of the principal axes of the bodies leads to a dramatic shift in the kinetic pathway taken. Counterintuitive predictions from simulation and theory are confirmed by experiment.

Self-assembly of thermophoretic active particles

Marisol Ripoll[1,2]
[1] Theoretical Soft-Matter and Biophysics, Institute of Complex Systems,
[2] Forschungszentrum Jülich, 52425 Jülich, Germany

Thermophoresis refers to the directed motion of colloidal particles in the presence of a temperature gradient, which can occur towards cold (thermophobic colloids) or warm areas (thermophilic colloids) [1]. Thermophoretic self-propelled motion can be induced in the cases of Janus or dimers colloidal particles, where the colloid surface has an asymmetric heat capacity [2,3]. The thermophoretic properties of the non-heated part produce then a propulsion against or towards the heated part. Equally sized thermophobic microdimers have shown to hydrodynamically behave like pullers, thermophobic microdimers like pushers, and half-coated Janus particles like neutral swimmers [4].

Hydrodynamic simulations of microdimers clearly indicate that these hydrodynamic behaviors can be modified and even reversed for microdimers built with beads of different sizes. Thermophobic microdimers with small heated beads show for example an important lateral attraction, which combined with the thermophoretic repulsion, gives rise to the self-assembly of the microdimers in flattened moving structures with a well-defined orientation and hexagonal order [5].

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[3] Yang, M. and Ripoll, M., *Phys. Rev. E* **84**, 061401, (2011).
[4] Yang, M., Wysocki, A., and Ripoll, M., *Soft Matter* **10**, 6208, (2014).
[5] Wagner, M. and Ripoll, M., preprint, (2017).

4 Participant List

Scientific Organizers

Martin Oettel – *Universität Tübingen, Germany*
Tanja Schilling – *University of Luxembourg/ Universität Freiburg*
Frank Schreiber – *Universität Tübingen, Germany*

Invited Speakers

Marjolein Dijkstra – *Utrecht University, Netherlands*
Andreas Heuer – *Universität Münster, Germany*
Sabine Klapp – *TU Berlin, Germany*
Stefan Kowarik – *HU Berlin, Germany*
Tobias Kraus – *Saarland University, Germany*
Joachim Krug – *Universität zu Köln, Germany*
Olivier Pierre-Louis – *Université Claude Bernard Lyon, France*
Marisol Ripoll – *Forschungszentrum Jülich, Germany*
Peter G. Vekilov – *University of Houston, US*
Walter Zimmermann – *Universität Bayreuth, Germany*

Conference dinner, Tuesday April 4th, Ludwigs

Restaurant Ludwigs
Uhlandstraße 1
72070 Tübingen
Tel: +49 7071 133121
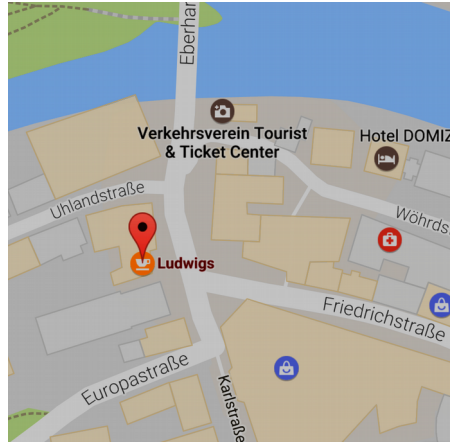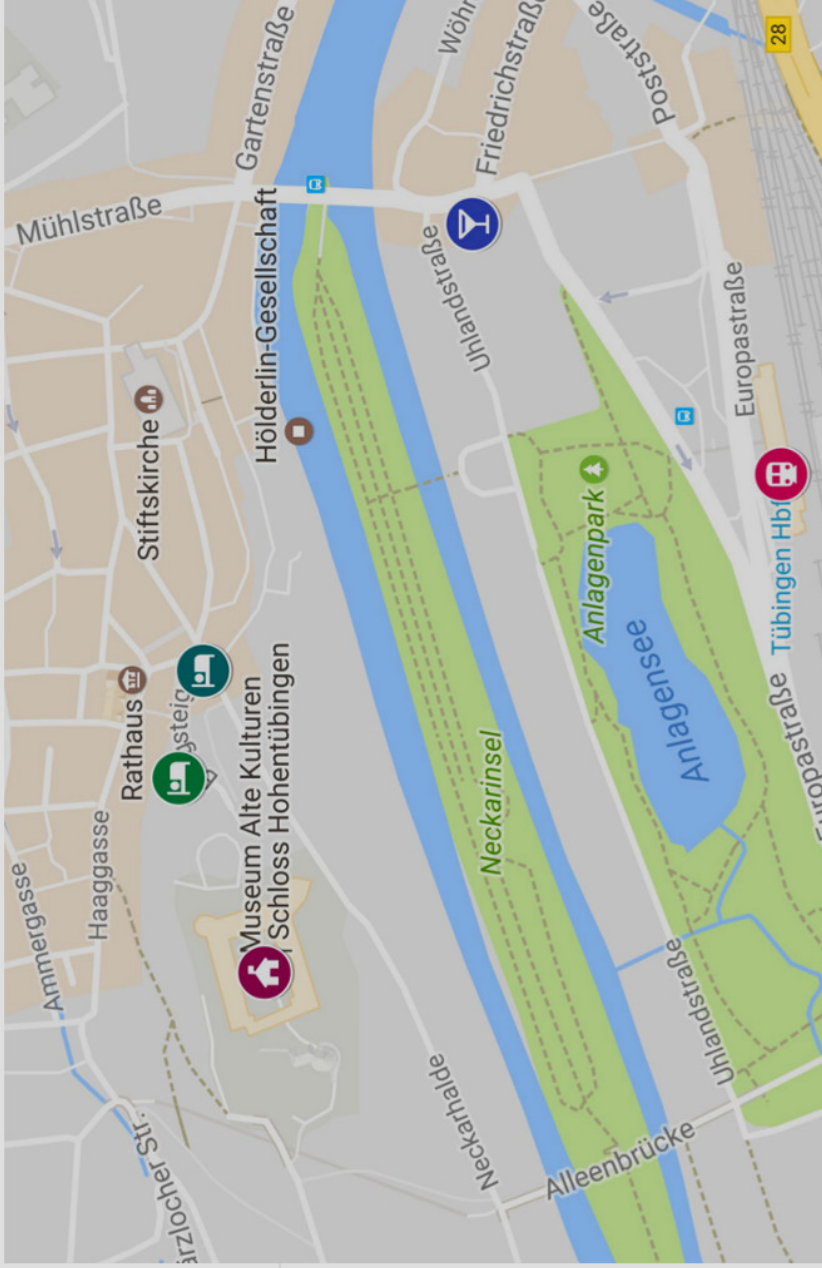

Direction from Schloss

It takes about 10 minutes to walk from Schloss to Ludwigs. You have to go east through the old town and pass the Neckar bridge.
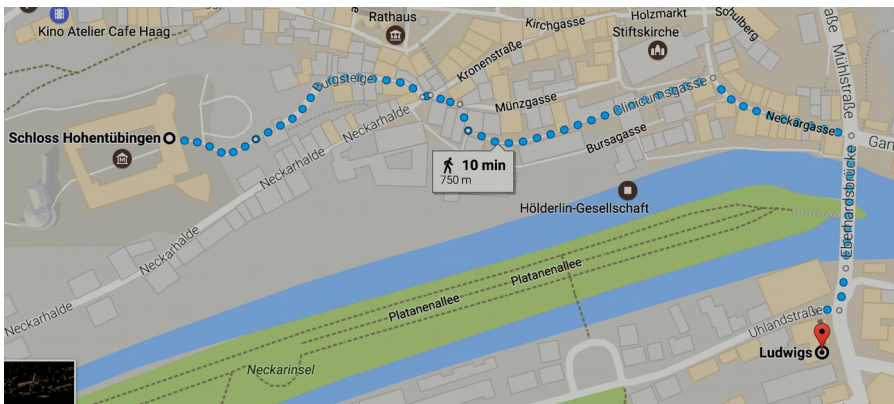
SoftComp - Tuebingen

SoftComp

- Train Station
- Hotel Hospiz
- Hotel Am Schloss
- Hohentübingen Castle
- Ludwigs