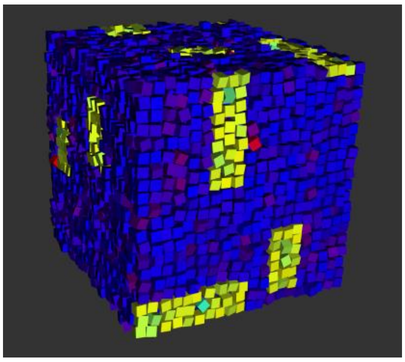
Complex colloidal crystals: Formation, inhomogeneities and defects



July 11th to 14th, 2022

CECAM-AT

TU Wien, Austria

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1. Description

It might be tempting to regard colloidal crystals as one of the simplest states of matter – in the cartoon picture of a colloidal crystal, a set of classically interacting spheres are beautifully arranged on an ordered lattice and simply fluctuate around their respective sites. Reality, however, is in no way bound to this idealized picture, and colloidal crystals exhibit an impressively rich variety of intriguing structural and dynamical phenomena, depending on the constituent particles and the crystallization conditions.

Structurally, for example, (hard) polyhedra with different shapes [1 - 4], asymmetrically charged particles [5, 6], and polydisperse mixtures of hard or charged spheres [7, 8] all can be persuaded to form a stunning variety of different crystal lattices as their microscopic features are tuned. This includes not only traditional crystals, but also plastic crystals [4], substitutional and interstitial solid solutions [9, 10], and “cluster crystals” [11, 12], where multiple particles occupy the same lattice site. Additionally, several of these have even been shown to form quasicrystals: exotic aperiodic structures with symmetries forbidden by the traditional rules of crystallography [13 - 20]. These structures can be further tuned by causing them to self-assemble in complex environments, e.g. under the influence of confinement or at a curved interface [21 - 23]. To further complicate this picture, all equilibrium crystals have a finite defect concentration of point defects, and while in some cases defects in colloidal crystals are fairly standard and localized, such as in hard spheres [24, 25], colloidal crystal have also been found to exhibit extremely high defect concentrations – to the point where one in twenty lattice sites is empty [26], or in colloid-polymer crystals, small polymers accumulate at interstitial sites [27].

On the dynamical side, despite their solid nature, some colloidal crystals permit almost fluid-like self-diffusion, due to e.g. high concentrations of exotically structured defects [26, 28], or the presence of highly mobile interstitial dopants [9, 29, 30]. Alternatively, mobility in crystals can be achieved via the fluctuations of grain boundaries [31- 33], the inclusion of active dopants [34 - 36], or perturbations from external fields. Intriguingly, topologically constrained transport, known from electronic transport, can be obtained using magnetic fields [37 - 39]. Varying quenching speeds, polydispersity and other parameters, the competition between mechanisms leading to crystallization versus vitrification can be followed in situ [7,40-46].

Underlying the rich crystal behaviour of colloidal systems is the important role played by entropy in systems where the interactions are weak compared to the thermal energy scale, implying that colloidal phases are highly susceptible to deformations and defects. Unravelling the physics of these complex colloidal crystals requires a coordinated research effort combining experiments, simulations, and theory. This workshop will act as a platform to bring together scientists from all interested communities, and will give them the opportunity to exchange ideas, knowledge, and methodologies. The main aims of this workshop include:

- To compare experimental, numerical and theoretical insights into the rules that govern the stability of exotic colloidal crystals, with a focus on the emergence of quasicrystals, defect-rich phases, and cluster crystals.
- To compare experimental, numerical and theoretical insights into the crystallization process of complex colloidal building blocks, with both a focus on how crystallization competes with glass formation and how grain boundaries evolve.

- To discuss from experimental, numerical and theoretical perspectives the interplay between geometry, defects, and complex colloidal interactions, with a focus on topological phenomena in crystals caused e.g. by curved interfaces, confinement, or edge states.

Key References

[1] F. Kümmel, P. Shabestari, C. Lozano, G. Volpe, C. Bechinger, Soft Matter, 11, 6187-6191 (2015)

[2] M. Oettel, S. Görig, A. Härtel, H. Löwen, M. Radu, T. Schilling, Phys. Rev. E, 82, (2010)

[3] F. Smallenburg, L. Filion, M. Marechal, M. Dijkstra, Proceedings of the National Academy of Sciences, 109, 17886-17890 (2012)

[4] M. Mortazavifar, M. Oettel, J. Phys.: Condens. Matter, 28, 244018 (2016)

[5] B. van der Meer, R. van Damme, M. Dijkstra, F. Smallenburg, L. Filion, Phys. Rev. Lett., 121, (2018)

[6] J. Tauber, R. Higler, J. Sprakel, Proc Natl Acad Sci USA, 113, 13660-13665 (2016)

[7] M. Girard, S. Wang, J. Du, A. Das, Z. Huang, V. Dravid, B. Lee, C. Mirkin, M. Olvera de la Cruz, Science, 364, 1174-1178 (2019)

[8] T. Skinner, D. Aarts, R. Dullens, Phys. Rev. Lett., 105, (2010)

[9] F. Lavergne, D. Aarts, R. Dullens, Phys. Rev. X, 7, (2017)

[10] S. Ganguly, J. Horbach, Phys. Rev. E, 98, (2018)

[11] B. van der Meer, M. Dijkstra, L. Filion, Soft Matter, 12, 5630-5635 (2016)

[12] K. Dietrich, G. Volpe, M. Sulaiman, D. Renggli, I. Buttinoni, L. Isa, Phys. Rev. Lett., 120, (2018)

[13] S. Pronk, D. Frenkel, J. Phys. Chem. B, 105, 6722-6727 (2001)

[14] J. Loehr, M. Loenne, A. Ernst, D. de las Heras, T. Fischer, Nat Commun, 7, (2016)

[15] J. Loehr, D. de las Heras, A. Jarosz, M. Urbaniak, F. Stobiecki, A. Tomita, R. Huhnstock, I. Koch, A. Ehresmann, D. Holzinger, T. Fischer, Commun Phys, 1, (2018)

[16] S. Deutschländer, P. Dillmann, G. Maret, P. Keim, Proc Natl Acad Sci USA, 112, 6925-6930 (2015)

[17] J. Kurzidim, D. Coslovich, G. Kahl, Phys. Rev. Lett., 103 (2009)

[18] Z. Zheng, F. Wang, Y. Han, Phys. Rev. Lett., 107, (2011)

[19] Z. Zheng, R. Ni, F. Wang, M. Dijkstra, Y. Wang, Y. Han, Nat Commun, 5 (2014)

[20] J. Lutsko, Sci. Adv., 5, (2019)

[21] S. Schütter, J. Roller, A. Kick, J. Meijer, A. Zumbusch, Soft Matter, 13, 8240-8249 (2017)

[22] S. Golde, T. Palberg, H. Schöpe, Nature Phys, 12, 712-717 (2016)

[23] A. Kuhnhold, H. Meyer, G. Amati, P. Pelagejcev, T. Schilling, Phys. Rev. E, 100, (2019)

[24] D. Talapin, E. Shevchenko, M. Bodnarchuk, X. Ye, J. Chen, C. Murray, Nature, 461, 964-967 (2009)

[25] J. Meijer, F. Hagemans, L. Rossi, D. Byelov, S. Castillo, A. Snigirev, I. Snigireva, A. Philipse, A. Petukhov, Langmuir, 28, 7631-7638 (2012)

[26] M. Dijkstra, Entropy-Driven Phase Transitions in Colloids: From spheres to anisotropic particles, 2014

[27] P. Damasceno, M. Engel, S. Glotzer, Science, 337, 453-457 (2012)

[28] E. Bianchi, C. Likos, G. Kahl, ACS Nano, 7, 4657-4667 (2013)

[29] E. Bianchi, C. Likos, G. Kahl, Nano Lett., 14, 3412-3418 (2014)

[30] P. Bommineni, N. Varela-Rosales, M. Klement, M. Engel, Phys. Rev. Lett., 122,(2019)

[31] B. Cabane, J. Li, F. Artzner, R. Botet, C. Labbez, G. Bareigts, M. Sztucki, L. Goehring, Phys. Rev. Lett., 116, (2016)

[32] L. Filion, M. Hermes, R. Ni, E. Vermolen, A. Kuijk, C. Christova, J. Stiefelhagen, T. Vissers, A. van Blaaderen, M. Dijkstra, Phys. Rev. Lett., 107, (2011)

[33] T. Ras, M. Szafarczyk, M. Fuchs, Colloid Polym Sci, 298, 803-818 (2020)

[34] B. Mladek, D. Gottwald, G. Kahl, M. Neumann, C. Likos, Phys. Rev. Lett., 96, (2006)

[35] J. Häring, C. Walz, G. Szamel, M. Fuchs, Phys. Rev. B, 92, (2015)

[36] J. Meijer, V. Meester, F. Hagemans, H. Lekkerkerker, A. Philipse, A. Petukhov, Langmuir, 35, 4946-4955 (2019)

[37] X. Ye, J. Chen, M. Eric Irrgang, M. Engel, A. Dong, S. Glotzer, C. Murray, Nature Mater, 16, 214-219 (2016)

[38] M. Zu, P. Tan, N. Xu, Nat Commun, 8, (2017)

[39] H. Pattabhiraman, A. Gantapara, M. Dijkstra, J. Chem. Phys., 143, 164905 (2015)

[40] D. Ratliff, A. Archer, P. Subramanian, A. Rucklidge, Phys. Rev. Lett., 123, (2019)

[41] P. Subramanian, A. Archer, E. Knobloch, A. Rucklidge, New J. Phys., 20, 122002 (2018)

[42] C. Achim, M. Schmiedeberg, H. Löwen, Phys. Rev. Lett., 112, 255501 (2014)

[43] T. Dotera, T. Oshiro, P. Ziherl, Nature, 506, 208-211 (2014)

[44] B. de Nijs, S. Dussi, F. Smallenburg, J. Meeldijk, D. Groenendijk, L. Filion, A. Imhof, A. van Blaaderen, M. Dijkstra, Nature Mater, 14, 56-60 (2014)

[45] J. Wang, C. Mbah, T. Przybilla, S. Englisch, E. Spiecker, M. Engel, N. Vogel, ACS Nano, 13, 9005-9015 (2019)

[46] W. Irvine, M. Bowick, P. Chaikin, Nature Mater, 11, 948-951 (2012)

2. Program

Day 1 – Monday, July 11th 2022

12:00 to 13:00	**Registration & Reception**
13:00 to 13:30	**Introduction to the workshop by the organizers**

Session: Crystallization vs. vitrification

Chair: Gerhard Kahl

13:30 to 14:15	**Stefan Egelhaaf**
	Crystallization - with a little help from seeds and shear
14:15 to 15:00	**Marjolein Dijkstra**
	Birth of binary colloidal crystals in bulk and spherical confinement
15:00 to 15:30	**Coffee break**
15:30 to 16:15	**Michael Engel**
	Bifurcation of colloidal crystallization in a sphere
16:15 to 17:00	**Hans Joachim Schöpe**
	Spatio-temporal fluctuations in- and out-of-equilibrium
17:00 to 17:45	**Tanja Schilling**
	Hard sphere crystal nucleation rates: reconciliation of simulation and experiment

Day 2 – Tuesday, July 12th 2022

08:15 to 09:00 – Registration

Session: Complex structures of anisotropic particles

Chair: **Stefan Egelhaaf**

09:00 to 09:45	**Andreas Zumbusch**
	Structural dynamics of ellipsoidal colloid suspensions
09:45 to 10:30	**Peter Keim**
	2D crystals of squares
10:30 to 11:00	**Coffee break**
11:00 to 11:45	**Daniel de las Heras**
	Colloidal transport in twisted magnetic patterns
11:45 to 12:30	**Thomas Fischer**
	Topological control of colloids on magnetic patterns
12:30 to 14:00	**Lunch**

Chair: **Matthias Fuchs**

14:00 to 14:30	**Emanuela Bianchi**
	How to assemble a hybrid crystal-liquid phase for photonic applications

Poster Session

14:30 to 15:30	**Lightning talks**
15:30 to 16:00	**Coffee break**
16:00 to 18:00	**Poster session**
19:00 to 22:00	**Dinner**

Day 3 – Wednesday, July 13th 2022

Session: Crystal defects

Chair: **Laura Filion**

09:00 to 09:45	**Roel Dullens**
	Formation and shrinkage of grain boundary loops in two - dimensional colloidal crystals
09:45 to 10:30	**Berend van der Meer**
	Uncovering the microscopic dynamics of colloidal grain boundaries
10:30 to 11:00	**Coffee break**
11:00 to 11:45	**Janne-Mieke Meijer**
	Self-assembly of cubic colloids: crystals, dynamics and defects
11:45 to 12:30	**Jürgen Horbach**
	Quasistatic yielding in three-dimensional crystals: a slip-plane condensation transition
12:30 to 14:00	**Lunch**

Session: Soft quasicrystals

Chair: **Michael Engel**

14:00 to 14:45	**Eva Noya**
	Making icosahedral quasicrystals with patchy particles
14:45 to 15:30	**Michael Schmiedeberg**
	Growth of soft quasicrystals
15:30 to 16:00	**Coffee break**
16:00 to 16:45	**Primoz Ziherl**
	Metallic-mean hexagonal quasicrystalline patterns
16:45 to 17:30	**Frank Smallenburg**
	Quasicrystals self-assembly in hard spheres on a plane
17:30 to 18:15	**Xin Cao**
	Colloidal crystals in studies of nanotribology

Day 4 – Thursday, July 14th 2022

Session: Surfaces, interfaces and boundaries

Chair: **Martin Oettel**

09:00 to 09:45	**James Lutsko**
	Using classical DFT to describe solids, fluid-solid interfaces and crystallization
09:45 to 10:30	**Daniela Kraft**
	Dumbbell impurities in 2d crystals of repulsive colloidal spheres induce particle-bound dislocations
10:30 to 11:00	**Coffee break**
11:00 to 11:45	**Alfons van Blaaderen**
	Self-assembly in spherical confinement: hierarchical structuring at multiple length scales
11:45 to 12:30	**Ivo Buttinoni**
	Driven and active colloidal crystals at fluid interfaces
12:30 to 13:30	Conference summary and closing remarks

1. Abstracts – Talks

July 11th, 2022 (Monday)

13:30 to 14:15

Crystallization - with a little help from seeds and shear

Stefan Egelhaaf

Heinrich Heine University, Germany

Hard-sphere colloids crystallize at large volume fractions. Using confocal microscopy, we follow crystallization in the presence of spherical seeds or moderate oscillatory shear.

In the presence of a seed, heterogeneous nucleation occurs. However, since the spherical seed is not perfectly compatible with the favoured crystal structure, elastic distortions develop and, on reaching a critical size, the crystallite detaches from the seed. The detached and relaxed crystallite continues to grow, except close to the seed, which now prevents crystallization. Hence, the crystallization seeds facilitate crystallization only during initial growth and then act as impurities [1].

In addition, shear-induced crystallization in binary suspensions of hard spheres has been investigated. For small size ratios, substitutionally disordered crystals are formed. With increasing size disparity, the volume fraction in substitutionally disordered crystals decreases while in amorphous regions it slightly increases; equal volume fractions are found for a size ratio of 1.18. Beyond this size ratio, shear-induced crystallisation is observed to cease, consistent with the empirical Hume-Rothery rule for metallic alloys.

[1] E. Allahyarov, K. Sandomirski, S. Egelhaaf, H. Löwen, Nat. Commun., 6, 7110 (2015)
[2] P. Maßhoff, I. Elsner, M. Escobedo-Sánchez, J. Segovia-Gutiérrez, A. Pamvouxoglou, S. Egelhaaf, J. Phys. Mater., 3, 035004 (2020)

14:15 to 15:00

Birth of binary colloidal crystals in bulk and spherical confinement

Marjolein Dijkstra

Utrecht University, Netherlands

15:00 to 15:30 Coffee break

15:30 to 16:15

Bifurcation of colloidal crystallization in a sphere

Michael Engel

Friedrich-Alexander-Universität Erlangen-Nürnberg, Germany

Bifurcations in kinetic pathways are critical points that decide the evolution of a system. An example is crystallization, in which the thermodynamically stable polymorph may not form due to kinetic hindrance. Competition between crystallization pathways is crucial at the nanoscale where interfaces and finite size effects produce unique multiply-twinned nanoparticles. Here, we use confined self-assembly to investigate the interplay of thermodynamics and kinetics in the formation of colloidal clusters under spherical confinement. We characterize various types of colloidal clusters and compare their thermodynamic stability. Hard-sphere simulations reveal a bifurcation at an early stage of the crystallization process. Fluid pre-ordering helps shaping the nucleus. Our results are assisted by an improved, high-precision free energy calculation method for partially ordered particle systems.

16:15 to 17:00

Spatio-temporal fluctuations in and out of equilibrium

Hans Joachim Schöpe

University of Tübingen, Germany

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. 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.

17:00 to 17:45

Hard sphere crystal nucleation rates: reconciliation of simulation and experiment

Tanja Schilling, Wilkin Wöhler

University of Freiburg, Germany

Over the past two decades, a large number of studies addressed the topic of crystal nucleation in suspensions of hard spheres. The shared result of all these efforts is that, at low super-saturations, experimentally observed nucleation rates and numerically computed ones differ by more than ten orders of magnitude. In order to compare simulated rate densities to experimentally measured ones, we propose an interpretation of the experimental data as a combination of nucleation and crystal growth processes (rather than purely the nucleation process). In particular, we show that crystalline domains form by a successive twinning process. We argue that experimentalists might have mistaken the scattering signals from these domains as signals from independent nuclei. This interpretation may resolve the long standing dispute about the differing rates.

July 12th 2022 (Tuesday)

09:00 to 09:45

Structural dynamics of ellipsoidal colloid suspensions

Andreas Zumbusch, Jörg Roller, Markus Voggenreiter, John Geiger, Marcel Rudolf, Aleena Laganapan, Matthias Fuchs

University of Konstanz, Germany

During the last two decades, real space imaging experiments based on white light or fluorescence microscopy yielded many important insights into the structural dynamics of spherical colloidal particle suspensions. Only recently, these experiments were extended to cover also ellipsoidal particles in 2D. The presence of translational and rotational degrees of freedom in such systems leads to numerous new phenomena. In this presentation, we will present results of experiments on dense suspensions of prolate ellipsoidal particles in 3D. As is the case for similar 2D systems, we find two separate glass transitions for translation and rotation. This leads to the emergence of a ‘liquid glass’, i.e. a state in which the particles are still mobile enough to change their position while their orientation is frozen. A detailed analysis of our data shows that the liquid glass state is accompanied by the emergence of nematic precursors. These are extended clusters of similarly oriented particles which are intersected by particles with random orientations. This is in stark contrast to the 2D case which is coined by the presence of pseudonematic domains. As an outlook we will discuss how the structural dynamics of the liquid glass state might further be investigated using active microrheology.

[1] J. Roller, A. Laganapan, J. Meijer, M. Fuchs, A. Zumbusch, Proc. Natl. Acad. Sci. U.S.A., **118**, (2021)

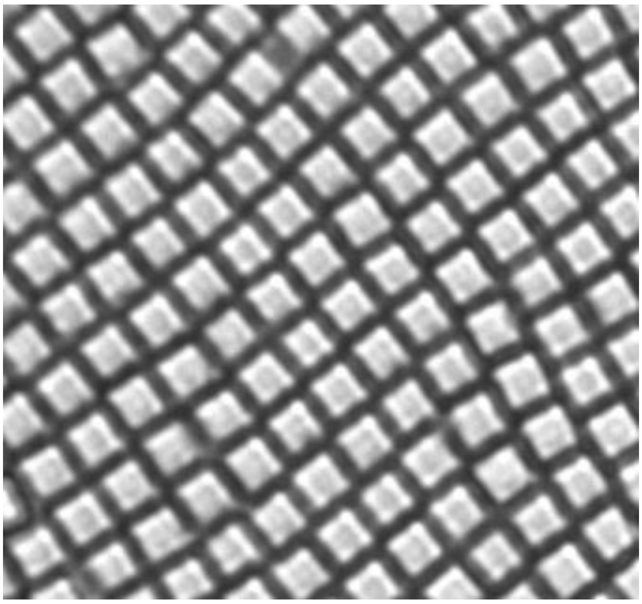
09:45 to 10:30

2D crystals of squares

Peter Keim

University of Konstanz, Germany

Squares of about 5 x 5 µm^2 with rather sharp edges were manufactured from a photo-resist using a 3D nanoprinter (nanoscribe GT). Particles in aqueous solution are sedimented by gravity to a thin cover slide where they form a monolayer. The curvature of the cover slide can be adjusted from convex to concave applying pressure to the solution, which allows to vary the area-density of the monolayer 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. Using a four-folded bond-order correlation function, we try to resolve the tetratic phase with quasi long range orientational order but short rang translational one. Simultaneously the body orientation of individual squares will be correlated with the local bond order or the ensemble.



10:30 to 11:00 **Coffee break**

11:00 to 11:45

Colloidal transport in twisted magnetic patterns

Daniel de las Heras, Nico C. X. Stuhlmüller, Thomas M. Fischer

University of Bayreuth, Germany

Recent studies have demonstrated that the motion of colloidal particles above periodic magnetic patterns and driven by a time-dependent uniform external field is topologically protected [1,2]. The colloidal trajectories exhibit clear analogies with the behaviour of electrons in topological insulators [3].

Motivated by recent studies in twisted graphene layers, we analyse with computer simulations the motion of colloidal particles located between two parallel magnetic patterns [4]. The patterns are periodic lattices of regions with positive and negative magnetization normal to the pattern. Both patterns are twisted by a given angle and shifted by half a unit-cell. An external drift force, which is constant in space and time, and small in amplitude compared to the magnetic forces, causes the particles to move along flat channels that emerge in the magnetic potential.

Due to the twist between the patterns, super-cells that are large compared to the periodicity of the underlying pattern develop. At the so-called magic twist-angles the super-cells are periodic, while they form a quasi-crystal for other angles. Only at magic angles, macroscopic transport is possible, whereas at non-magic angles the transport ceases after a few super-cells.

[1] J. Loehr, M. Loenne, A. Ernst, D. de las Heras, T. Fischer, Nat. Commun., **7**, 11745 (2016)
[2] D. Heras, J. Loehr, M. Loenne, T. Fischer, New J. Phys., **18**, 105009 (2016)
[3] J. Loehr, D. de las Heras, A. Jarosz, M. Urbaniak, F. Stobiecki, A. Tomita, R. Huhnstock, I. Koch, A. Ehresmann, D. Holzinger, T. Fischer, Commun. Phys., **1**, 4 (2018)
[4] N. Stuhlmüller, T. Fischer, D. de las Heras, Commun. Phys., **5**, 48 (2022)

11:45 to 12:30

Topological control of colloids on magnetic patterns

Thomas Fischer

University of Bayreuth, Germany

Topology plays a crucial role in electronic systems protecting the transport of charge and spin against dissipative scattering, which in topologically trivial systems usually destroys the transport. In topologically nontrivial electronic systems a whole zoo of new kind of particles, originally postulated in high energy physics, with unusual transport properties could be found experimentally in medium energy solid state physics. The field is large enough to entertain the solid-state community for more than a decade. Here we show with experimental and theoretical examples [1-6] that similar, albeit not identical, behaviour can be found in soft matter systems, where driven magnetic colloids carrying a magnetic moment replace the electrons, and periodic magnetic patterns replace the background solid state.

[1] A. Ernst, A. M. E. B. Rossi, and T.M. Fischer, Adiabatic and irreversible classical discrete time crystals, 10.48550/ARXIV.2203.04063.
[2] N. C. X. Stuhlmüller, T.M. Fischer, and D. de las Heras, Communications Physics **5**, 48 (2022).
[3] M. Mirzaee-Kakhki, A. Ernst, D. de las Heras, M. Urbaniak, F. Stobiecki, J. Gördes, M. Reginka, A .Ehresmann, and T.M. Fischer, Nature Communications **11**, 4670 (2020).
[4] J.Loehr, D. de las Heras, M. Loenne, J. Bugase, A. Jarosz, M. Urbaniak, F. Stobiecki, A. Tomita, R. Huhnstock, I. Koch, A. Ehresmann, D. Holzinger, and T.M. Fischer, Soft Matter **13**, 5044 (2017).
[5] J. Loehr, D. de las Heras, A. Jarosz, M. Urbaniak, F. Stobiecki, A. Tomita, R. Huhnstock, I. Koch, A. Ehresmann, D. Holzinger, and T.M. Fischer, Communications Physics **1**, 4 (2018).
[6] J. Loehr, M. Loenne, A. Ernst, D. de las Heras, and T.M. Fischer, Nature Communications **7**, 11745 (2016).

12:30 to 14:00 Lunch

14:00 to 14:30

How to assemble a hybrid crystal-liquid phase for photonic applications

Emanuela Bianchi

Technische Universität Wien, Austria

Hybrid Crystal-Liquid (CL) phases -- where a crystal is enriched by mobile particles that stabilize the whole structure -- have been recently observed in very diverse systems ranging from mixtures of complementary DNA-functionalized nanoparticles [1] to elemental solids under high pressures [2]. We focus on a layered crystal where delocalized inter-layer particles move freely between the planes [3], a lattice that we target because of the photonic properties that lamellar crystals usually have [4]. This novel structure has been observed to spontaneously form out of the fluid phase under temperature annealing for a system of heterogeneously charged particles [5]. We propose here a minimal particle model to search for the key requirements of the inter-particle potential that are needed to self-assemble the CL structure [6].

[1] M. Girard, S. Wang, J.S. Du, A. Das, Z. Huang, V.Y. Dravid, B. Lee, C.A. Mirkin, M. Olvera de la Cruz, Science, **364**, 1174- 1178 (2019)
[2] V.N. Robinson, H. Zong, G.J. Ackland, G. Woolman, A. Hermann, PNAS, **116**, 10297-10302 (2019)
[3] S. Ferrari, E. Bianchi, G. Kahl, Nanoscale, **9**, 1956 (2017)
[4] J.-C.P. Gabriel, F. Camerel, B.J. Lemaire, H. Desvaux, P. Davidson, P. Batail, Nature, **413**, 504?508 (2001)
[5] E. Bianchi, P.D.J. van Oostrum, C. Likos, G. Kahl, Current Opinion in Colloid & Interface Science, **30**, 18 (2017)
[6] D. Jin, S. Ferrari, F. Benedetti and E. Bianchi, in preparation

14:30 to 15:30	**Lightning talks**
15:30 to 16:00	**Coffee break**
16:00 to 18:00	**Poster session**
19:00 to 22:00	**Dinner**

July 13th 2022 (Wednesday)

09:00 to 09:45

Formation and shrinkage of grain boundary loops in two- dimensional colloidal crystals

Roel Dullens

University of Oxford, UK

Understanding the dynamics of grain boundaries in polycrystalline metals and alloys is crucial to enable tuning their mechanical properties. From an experimental point of view, grain boundaries in colloidal crystals are convenient model systems since imaging their dynamics requires only simple optical microscopy and they can be manipulated using optical tweezers. The formation and kinetics of grain boundaries are closely related to the topological constraints imposed on their complex dislocation structure. As such, loop-shaped grain boundaries are unique structures to establish such a link because their overall topological "charge" is zero due to their null net Burgers vector.

Here, we study the formation and shrinkage of such grain boundary loops by creating them on demand via a local rotational deformation of a two-dimensional colloidal crystal using an optical vortex. In particular, we observe that a grain boundary loop only forms if the product of its radius and misorientation exceeds a critical value. In this case, the deformation is plastic and the grain boundary loop spontaneously shrinks at a rate that solely depends on this product while otherwise, the deformation is elastically restored. We show that this elastic-to-plastic crossover is a direct consequence of the unique dislocation structure of grain boundary loops. Our results thus reveal a new general limit on the formation of grain boundary loops in two-dimensional crystals and elucidate the central role of defects in both the onset of plasticity and the kinetics of grain boundaries.

09:45 to 10:30

Uncovering the microscopic dynamics of colloidal grain boundaries

Berend van der Meer[1], Mathieu Baltussen[1], Arran Curran[1], Marjolein Dijkstra[2], Roel Dullens[1]

[1]Radboud University, Netherlands

[2]Utrecht University, Netherlands

Grain boundary (GB) migration induces significant changes in structure in polycrystalline solids and has a pronounced effect on the macroscopic properties of these materials [1-3]. On the atomic scale, GB migration is governed by the atomistic mechanism via which constituent particles are displaced between lattice sites of neighbouring grains. Despite many efforts to characterize such atomistic mechanism for GB motion, a comprehensive understanding of the migration mechanism in terms of the dynamics of atoms and dislocations remains largely unclear. Crucially, previous work has identified that how atoms are displaced from one grain to another is highly related to the (re)arrangement of the atoms inside the GB [4, 5]. Namely, GBs are not simply amorphous structures, and have been variously described in terms of combinations of different structural units [6], coincidence site lattices [7], and dislocations [8]. Yet, predicting how the GB structure translates into its motion provides a considerable challenge owing to the many microscopic degrees of freedom at play.

We investigate this link between particle, defect and GB motion experimentally by using holographic optical tweezers [9] to create loop-shaped GBs in a two-dimensional colloidal model system. We reveal a clear connection between the dynamics of particles and dislocations where local block rotations observed in the particle dynamics correspond to the reaction and glide of dislocations along the paths of maximum misfit. Hence, the paths taken by the dislocations correspond to regions of largest particle displacement. Moreover, we identify the particle dynamics to be guided by a set of equivalence points between both lattices that act as points of transition as particles displace between grains. Using this geometric underpinning, we establish a framework that predicts the microscopic dynamics of particles and dislocations during GB motion in our two-dimensional crystals.

[1] JM Howe, Interfaces in materials. (John Wiley & Sons, 1997)
[2] EO Hall, Proceedings of the Physical Society, Section B 64 (9), 747 (1951)
[3] NJ Petch, Journal of the Iron and Steel Institute **174**, 25 (1953).
[4] J Wei et al., Nature Materials **20 (7)** , 951 (2021).
[5] Z Hao et al., Acta Materialia **54 (3)**, 623 (2006).
[6] AP Sutton et al., Philosophical Transactions of the Royal Society of London. Series A **309** (1506), 1-36(1983).
[7] DG Brandon, Acta Metallurgica 14 (11) 1479 (1966).
[8] WT Read et al., Physical Review **78 (3)**, 275 (1950).
[9] FA Lavergne et al., PNAS **115** (27), 6922 (2018).

10:30 to 11:00 Coffee break

11:00 to 11:45

Self-assembly of cubic colloids: crystals, dynamics and defects

Janne-Mieke Meijer

Eindhoven University of Technology, Netherlands

Colloids with a cubic shape can be prepared on the nano and micron scale and have become of interest for self-assembly of superstructures, with applications in photonics, surface coatings, and solar cells. During the self-assembly processes a distinct coupling appears to occur between the cubic shape, the interparticle interactions and the applied forces. However, the relationship between the cubic colloid properties and the order/disorder in the final self-assembled superstructures remains unclear.

In this talk, I will address our experimental work on how cubic colloids with rounded corners, i.e. superballs [1], and their interparticle interactions influence their self-assembly in- and out-of-equilibrium. Using a combination of small angle x-ray scattering, (optical) microscopy and quantitative image analysis, we studied the cubic superstructures formed by different self-assembly methods. For hard body interactions between the cubes, we observed a rich phase behaviour, including a plastic crystal, dense rhombic packings and solid-solid phase transitions [1], which we found to be partly in agreement with theory and simulations [2]. When employing solvent evaporation to drive self-assembly, we find that strong immersion capillary forces and solvent flow, lead to distinct coupling between the cubic shape and these forces. We observed co-existence of the densest packings and switching due to sliding of the cubes as well as unique defect structures and the absence of vacancies [3,4]. In addition, we explored external pressure to organize the cubes in monolayers and find that parallel to the direction of the applied force increased order arises but perpendicular to the force the ordered is lost [5]. Finally, I will discuss our most recent results on 2D monolayers in equilibrium, where plastic crystal and rhombic phases with distinct diffusing defects are observed. Our experimental results reveal that finite colloidal cube shape details couple with the self-assembly process and lead to complex superstructure symmetries.

[1] J. Meijer, A. Pal, S. Ouhajji, H. Lekkerkerker, A. Philipse, A. Petukhov, Nat. Commun., **8**, 14352 (2017)
[2] R. Ni, A. Gantapara, J. de Graaf, R. van Roij, M. Dijkstra, Soft Matter, **8**, 8826 (2012)

[3] J. Meijer, F. Hagemans, L. Rossi, D. Byelov, S. Castillo, A. Snigirev, I. Snigireva, A. Philipse, A. Petukhov, Langmuir, **28**, 7631-7638 (2012)
[4] J. Meijer, V. Meester, F. Hagemans, H. Lekkerkerker, A. Philipse, A. Petukhov, Langmuir, **35**, 4946-4955 (2019)
[5] D. Napel, J. Meijer, A. Petukhov, Applied Sciences, **11**, 5117 (2021)

11:45 to 12:30

Quasistatic yielding in three-dimensional crystals: a slip-plane condensation transition

Jürgen Horbach[1], Parswa Nath[2], Peter Sollich[3], Surajit Sengupta[2]

[1]Heinrich Heine University Düsseldorf, Germany

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[3]University of Göttingen, Germany

When a crystalline solid is slowly deformed with a constant deformation rate, plastic flow initiates only at a limiting deformation called the yield point. In this talk, the yielding of a Lennard-Jones fcc crystal with periodic boundary conditions is studied using particle-based simulation techniques. We show that in the quasi-static limit the yielding can be described as a slip-plane condensation transition. This is a novel type of first-order phase transition that occurs in the limit of very large but finite linear dimension of the system. It is associated with a jump of an intensive thermodynamic variable. For finite deformation rates, we also present a theory for the deformation rate dependence of the yield strain which is in excellent agreement with the simulation data.

12:30 to 14:00 Lunch

14:00 to 14:45

Making icosahedral quasicrystals with patchy particles

Eva Noya[1], Chak Wong[2], Daniel Tracey[2], Pablo Llombart[1], Jonathan Doye[2]

[1]Consejo Superior de Investigaciones Científicas, Spain

[2]University of Oxford, United Kingdom

Icosahedral quasicrystals (IQC) are materials that display long-range order but are not periodic in any direction of space. IQC were the first experimentally discovered quasicrystals in the ground-breaking work of Shechtman *et al*. [1]. Since then, many icosahedral quasicrystals were found in many metallic alloys [2], but, so far, they have not been ever observed in any other type of materials. By contrast, axial quasicrystals have been identified in several soft-matter systems [3,4]. In this talk we will show that, building in our previous work [5,6], model patchy colloids can be designed to assemble into icosahedral quasicrystals. These model systems might be experimentally realized using DNA origami particles [7]. Our rational design strategy leads to model systems that robustly assemble in simulations into target IQC

through directional bonding [8]. The key design feature is the geometry of the particles that favors the propagation of an icosahedral network of bonds, even when many particles are not fully bonded.

[1] D. Shechtman, I. Blech, D. Gratias, J. Cahn, Phys. Rev. Lett., **53**, 1951-1953 (1984)
[2] H. Takakura, C. Gómez, A. Yamamoto, M. De Boissieu, A. Tsai, Nature. Mater., **6**, 58-63 (2006)
[3] X. Zeng, G. Ungar, Y. Liu, V. Percec, A. Dulcey, J. Hobbs, Nature, **428**, 157-160 (2004)
[4] T. Dotera, Isr. J. Chem., **51**, 1197-1205 (2011)
[5] D. Tracey, E. Noya, J. Doye, J. Chem. Phys., **151**, 224506 (2019)
[6] D. Tracey, E. Noya, J. Doye, J. Chem. Phys., **154**, 194505 (2021)
[7] Y. Wang, L. Dai, Z. Ding, M. Ji, J. Liu, H. Xing, X. Liu, Y. Ke, C. Fan, P. Wang, Y. Tian, Nat. Commun., **12**, 3011 (2021)
[8] E. Noya, C. Wong, P. Llombart, J. Doye, Nature, **596**, 367-371 (2021)

14:45 to 15:30

Growth of soft quasicrystals

Michael Schmiedeberg[1], Cristian V. Achim[2], Hartmut Löwen[3], Stefan Wolf[1], Michael Engel[1], Anja Gemeinhardt[1], Miriam Martinsons[1]

[1]Friedrich-Alexander-Universität Erlangen-Nürnberg, Germany

[2]Aalto University, Finland

[3]Heinrich-Heine-Universität Düsseldorf, Germany

We present and discuss different approaches to grow quasicrystals. First, we consider a phase field crystal model [1] as well as Brownian Dynamics simulations [2] for structures that are stabilized by an interaction potential with two incommensurate lengths scales. We explore the growth processes of two-dimensional colloidal quasicrystals and observe two different growth modes. Close to the triple point a perfect, defect-free quasicrystal can grow. However, far away from the triple pint the growth is dominated by phasonic flips which are incorporated as local defects into the grown structure [1,2]. The later structure corresponds to a dislocation-free random-tiling-like quasicrystal. A similar growth has been reported for experiments [3].

Second, we study a systems where the quasicrystalline ordering is due to interactions that possess preferred binding angles as for patchy colloids [2]. Preferred binding angles support the creation of chain-like structures that reach out of the surface and lead to the incorporation of dislocations [2].

Finally, in a toy model for the growth of quasicrystals 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. Phasonic rearrangements or fluctuations might be important to facilitate the formation of almost perfect quasicrystals [1-3] but they are not indispensable.

[1] C. Achim, M. Schmiedeberg, H. Löwen, Phys. Rev. Lett., **112**, 255501 (2014)
[2] A. Gemeinhardt, M. Martinsons, M. Schmiedeberg, Eur. Phys. J. E, **41**, 126 (2018)
[3] K. Nagao, T. Inuzuka, K. Nishimoto, K. Edagawa, Phys. Rev. Lett., **115**, 075501 (2015)

15:30 to 16:00 Coffee break

16:00 to 16:45

Metallic-mean hexagonal quasicrystalline patterns

Primoz Ziherl

University of Ljubljana and Jozef Stefan Institute, Slovenia

The most striking feature of conventional quasicrystals is their non-traditional symmetry characterized by icosahedral, dodecagonal, decagonal, or octagonal axes. The symmetry and the aperiodicity of these materials stem from an irrational ratio of two or more length scales controlling their structure, the best-known examples being the Penrose and the Ammann–Beenker tiling as two-dimensional models related to the golden and the silver mean, respectively. For a long time, these two means were the only metallic means known to be related to quasicrystals. We propose a self-similar bronze-mean hexagonal pattern [1], and we use numerical simulations to demonstrate that its disordered variant can be materialized in colloidal particles with a core–shell architecture [2]. By varying the geometry of the pattern we furthermore generate a sequence of structures, which provide an alternative interpretation of quasicrystalline approximants observed in some metal–silicon alloys. We also construct generalizations of the bronze-mean pattern that consist of increasingly larger periodic domains, which can be viewed as aperiodic approximants of a periodic crystal [3]. Consisting of small and large triangles and rectangles, these tilings are based on metallic means of multiples of 3. Together with the non-metallic-mean three-tile hexagonal tilings, they provide a comprehensive theoretical framework for the complex structures seen, e.g., in some binary nanoparticles, oxide films, and intermetallic alloys.

16:45 to 17:30

Quasicrystals self-assembly in hard spheres on a plane

Frank Smallenburg

CNRS, Université Paris-Saclay, France

Over the last few decades, quasicrystals have been observed in a variety of soft-matter systems, including nanoparticles, micelles, and block copolymers. However, realizing the self-assembly of quasicrystals in systems of colloidal particles on the micron-scale remains a challenge. Here, we use computer simulations to demonstrate that an extremely simple two-dimensional model system -- a binary mixture of hard spheres lying on a flat substrate -- is capable of self-assembling into not just one, but two quasicrystal phases: one dodecagonal and one octagonal. The octagonal quasicrystal consists of three different type tiles: two sizes of squares and an isosceles triangle, whose relative concentrations can be tuned by changing the fraction of small particles in the binary mixture. Moreover, these concentrations can be theoretically predicted based on the assumption of a tiling with octagonal symmetry. The eight-fold quasicrystal appears over a significant range of both number ratio and size ratio of the two sphere species, which can be understood from geometrical arguments. Our results strongly suggest that this simple, entropy-driven model is an ideal playground for the studying the behaviour of soft quasicrystals.

17:30 to 18:15

Colloidal crystals in studies of nanotribology

Xin Cao

University of Konstanz, Germany

Colloids are often used as ideal model systems to study many fundamental physics from equilibrium phase transitions to out-of-equilibrium glassy dynamics and far-from-equilibrium collective behaviours. Here we show that colloidal systems are also excellent for the studies of nanoscale friction, which considers two contacting

crystalline surfaces coming into relative motion. We focus mainly on rigid and finite sized contacts which includes the following two topics: 1) Orientational/directional locking and friction anisotropy of rigid colloidal crystalline clusters sliding across periodic and quasiperiodic surfaces. 2) The rotational friction and its interplay with translational friction when rigid colloidal crystal line clusters are rotated and translated simultaneously on periodic surfaces. These works demonstrate that colloidal systems are powerful tools to explore and to predict interesting tribological behaviours relevant for nano manipulations and for the functioning of nano mechanical devices.

July 14th 2022 (Thursday)

09:00 to 09:45

Using classical DFT to describe solids, fluid-solid interfaces and crystallization

James Lutsko

Université Libre de Bruxelles, Belgium

This talk is an overview of the work from our group which is aimed at using modern classical DFT to understand crystallization. While the methods are broadly applicable, our work is targeted towards simple fluids and colloidal systems interacting via two-body potentials such as hard-spheres and Lennard-Jones. I will discuss the quality of the description of the solid phase, the solid-fluid interface (recent, unpublished work) and how, combined with fluctuating hydrodynamics, this provides a theoretical approach to studying crystallization.

[1] J. Lutsko, Phys. Rev. E, **102**, 062137 (2020)
[2] J. Lutsko, C. Schoonen, Phys. Rev. E, **102**, 062136 (2020)
[3] J. Lutsko, The Journal of Chemical Physics, **136**, 034509 (2012)
[4] J. Lutsko, Sci. Adv., **5**, (2019)

09:45 to 10:30

Dumbbell impurities in 2d crystals of repulsive colloidal spheres induce particle-bound dislocations

Daniela Kraft

Leiden University, Netherlands

Impurity-induced defects play a crucial role for the properties of crystals, but little is known about impurities with anisotropic shape. In this talk, I will show how colloidal dumbbells distort and interact with a hexagonal crystal of charged colloidal spheres at a fluid interface. We find that subtle differences in the dumbbell length determine whether it induces a local distortion of the lattice or traps a dislocation, and determine how the dumbbell moves inside the repulsive hexagonal lattice. Our results provide new routes towards controlling material properties through particle-bound dislocations.

10:30 to 11:00 Coffee break

11:00 to 11:45

Self-assembly in spherical confinement: hierarchical structuring at multiple length scales

Alfons van Blaaderen

University of Utrecht, Netherlands

Drying emulsion droplets of particles is a powerful methodology to create structured nanomaterials, supraparticles (SPs) [1-8], by self-assembly (SA) as also the final structures can still be in the colloidal domain and thus take part in further/additional SA. Interestingly, we have shown that both for single sized [1] and binary [2] dispersions of colloids interacting with a hard interaction potential SA inside a spherical boundary results in equilibrium phases with *icosahedral* symmetry and are thus different from bulk phases up until SPs composed of >100.000 particles! Deviations from a spherical shape can be used to induce directional attachment of the particles inside the SPs [3]. Next to many possible applications in fields like catalysis, sensing [4-5], lighting [6-8], structural colors, (nano)photonics [5-7] (including lasing [8]), quantum matter [3] and sustainability research in general, supraparticles are leading to new insights into fundamental questions in condensed matter science as well.

When the dispersion droplets are not dried fully, but e.g. coated with a (meso)porous silica layer the resulting supraparticles have particles inside their shells with remaining Brownian motion [9-10] and can thus be manipulated with external (e.g. electric) fields [9-10]. This allows for dynamical tuning of e.g. the scattering and/or interactions of these dispersion SPs.

[1] de Nijs, et al., Dijkstra, AvB, Nature Materials 14, 56 (2015).
[2] Wang, et al. Dijkstra AvB, Nature Physics 17, 128–134 (2021).
[3] Wang, et al., AvB, Nature Comm., 8, 2228 (2018).
[4] Besseling, et al, Dijkstra, AvB, J. of Phys: Cond. Mat., 27, 194109 (2015)
[5], Van der Hoeven, et al. AvB, Adv. Functional Mat., accepted (2022).
[6] Vanmaekelbergh, et al., AvB, ACS Nano, 9, 3942 (2015).
[7] Montanarella et al., AvB, ACS Nano, 11 (9), 9136-9142 (2017).
[8] AvB, , Stöferle, Vanmaekelbergh, ACS Nano, 12 (12), 12788-12794 (2018).
[9] Welling, et al. AvB, ACS Nano 15, 11137(2021).
[10] Watanabe, et al. AvB, Nagao, JCIS 566, 202–210 (2020).

11:45 to 12:30

Driven and active colloidal crystals at fluid interfaces

Ivo Buttinoni[1], Roel Dullens[1], Lucio Isa[2]

[1]Heinrich-Heine University, Germany

[2]ETH Zürich, Switzerland

Colloidal monolayers are usually prepared either by letting microparticles sink onto a bottom substrate or by spreading them at flat liquid interfaces. If charged colloids are used, the second method can lead to the formation of very loosely-packed lattices which are therefore easy to deform.

By spreading monodisperse particles, we prepare two-dimensional hexagonal crystals with tuneable lattice constant and study how they deform if local stresses are applied. More specifically, we look at the mechanical and structural response when shear flows are applied by driving a magnetic shearing probe or when local deformation are imposed by optically driving single colloidal particles. The latter situation can be also achieved by adding particles that are able to move autonomously in the fluid.

Our results highlight that the colloidal lattices behave as isotropic elastic materials when the applied deformations are (much) smaller than the lattice spacing and show pronounced plasticity otherwise. Remarkably, the work required to displace particles through the crystal agrees well with the dynamics of self-propelling colloids, provided that an effective force is considered

12:30 to 13:30

Conference summary and closing remarks

2. Posters

Active crystallization from power functional theory

Sophie Hermann, Matthias Schmidt

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We investigate the full phase behaviour of three-dimensional active Brownian particles, including active gas, liquid, and crystalline phases [1,2]. We develop a power functional approximation [3] for the non-equilibrium equation of state and validate it against recent simulation results [4,5]. Phase separation emerges from equality of non-equilibrium state functions in the coexistence phases. Fluid-fluid motility-induced phase separation ends at a critical point and becomes metastable above a gas-liquid-solid triple point, in agreement with simulation data [4,5]. The behaviour of the system can be rationalized by the presence of the mean swim speed as a further relevant degree of freedom, besides the density.

[1] S. Hermann, P. Krinninger, D. de las Heras, M. Schmidt, Phys. Rev. E, 100, 052604 (2019)
[2] S. Hermann and M. Schmidt, Active crystallization from power functional theory (to be published).
[3] M. Schmidt, Rev. Mod. Phys., 94, 015007 (2022)
[4] F. Turci, N. Wilding, Phys. Rev. Lett., 126, 038002 (2021)
[5] A. Omar, K. Klymko, T. GrandPre, P. Geissler, Phys. Rev. Lett., 126, 188002 (2021)

Continuum mechanics of non-ideal crystals: derivation starting from microscopic density fluctuations

Saswati Ganguly

[1]University of Konstanz, Germany

Continuum elasticity theory, widely used to describe the reversible mechanical response in solids, cannot be derived from a first principle microscopic description as soon as defects are introduced in a crystalline solid. This ends up requiring a general hydrodynamic description of defect-rich solids derived from microscopic particle dynamics using Mori-Zwanzig projection formalism [1,2]. The crystalline phase at a finite temperature has a non-zero concentration of vacancies and interstitials. These local-defects influence the reversible mechanical response as well as irreversible transport processes. Starting from the classical many-body Hamiltonian, we derive [2] the laws of continuum mechanics governing the elastic response, defect diffusion and heat transport in non-ideal crystals. The theory finds partial validation[3] through successfully predicting the isothermal elastic properties of cluster-crystal phases known to mimic DNA-based dendritic nanostructures. In ensembles with constant temperature or entropy, the framework presents reversible mechanical properties from equilibrium correlations and transport coefficients as Green-Kubo formulae. The formulation prescribes ways to measure these quantities in atomistic simulations and particle tracking colloidal experiments.

[1] C. Walz and M. Fuchs, Phys. Rev. B 81, 134110 (2010)
[2] F. Miserez, S. Ganguly, R. Haussmann and M. Fuchs, arXiv:2205.08405 (2022)
[3] S. Ganguly, G. P. Shrivastav, S.-C. Lin, J. Häring, R. Haussmann, G. Kahl, M. Oettel and M. Fuchs, J. Chem. Phys. 064501, 156, (2022)

Defects and nucleation in charged colloids

Marjolein de Jager, Laura Filion, Rinske M. Alkemade

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Suspensions of charged colloids are among the most fundamental systems in colloidal science. Due to their straightforward realization in the lab and impressive agreement with computational and theoretical studies, these systems have been instrumental in enhancing our understanding of both atomic and colloidal crystals. Nonetheless, despite the century-long investigation into charged colloids, their behaviour can still surprise us.

In this poster I will discuss two well-kept secrets of charged colloids related to crystal nucleation and point defects. With respect to nucleation, I will show that even highly screened charge interactions can clearly change the nucleation free-energy barriers of (almost) hard-spheres, bringing into question to what extent well-screened charged particles can ever be considered to behave like hard spheres in the context of crystal nucleation. Additionally, I will discuss how defects manifest in both face-centred cubic (FCC) and body-centred cubic (BCC) crystals of charged colloids. I will show that point defects are predicted to occur in surprisingly high concentrations in the BCC crystal, and that the interstitials in the BCC crystal form exotic, one-dimensional, long-ranged deformations called crowdions [1].

[1] R. Alkemade, M. de Jager, B. van der Meer, F. Smallenburg, L. Filion, J. Chem. Phys., **154**, 164905 (2021)

Inverse design of self-assembling soft and porous materials via free energy-based and deep learning-based evolution strategies

Alberto Pérez de Alba Ortíz, Chaohong Wang, Marjolein Dijkstra

Utrecht University, Netherlands

Self-assembly provides a promising route to synthesize nanomaterials with desired chemical and physical properties. While precise control of such ordering and aggregation processes remains a daunting task, computational inverse design can guide and accelerate their realization. Here, we apply evolutionary computation—i.e., covariance matrix adaptation evolution strategy—to reverse-engineer the formation of various target phases. Our typical design parameters include temperature, volume, pressure, terms of the interaction potential and molar fractions. Our target phases include FCC, HCP and diamond, as well as porous phases with square and hexagonal pores, and zeolitic materials. We successfully stabilize a target phase by optimizing a fitness across few generations of Monte Carlo or Molecular Dynamics sampling. Employing a machine-learned fitness based on a convolutional neural network classifier of diffraction patterns, we flexibly target different phases without previous knowledge about order parameters. Alternatively, using a free energy-based fitness along an environment similarity order parameter, we accelerate sampling and control the stability of a target phase. By including multiple order parameters, we design minimum free-energy pathways between different metastable phases. Our design pipeline provides a robust tool to control not only stable phases, but also transition pathways and barriers in complex soft materials.

Surface-tension induced crystal polymorphism

Cédric Schoonen, James Lutsko

Université Libre de Bruxelles (ULB), Belgium

Classical Density Functional Theory is used to determine the fluid-solid surface tensions for low-index faces of crystals of hard-sphere and Lennard-Jones particles. The calculations make use of the recently introduced explicitly stable Fundamental Measure Theory model for hard-spheres and we show that this gives state-of-the-art accuracy compared to simulation. For the Lennard-Jones system, results are presented for both solid-liquid and solid-vapor interfaces and in both cases the FCC results compare favorable with existing results from the literature. We find that the BCC crystal has significantly lower solid-liquid surface tension than the FCC structure. For the solid-vapor interface, our results indicate that the BCC phase is unstable with respect to transition to the HCP structure, in agreement with various zero-temperature results in the literature. We use our results to construct a capillary model for the free energy of solid clusters and show that for crystallization from the melt, small BCC clusters have lower free energy than FCC clusters, but with a crossover to favoring the bulk FCC phase for clusters in the range of 10-50 molecules. These results allow us to make sense of the large and somewhat contradictory literature concerning FCC-BCC polymorphism in such systems and thereby demonstrates a new theoretical tool for understanding crystallization.

Towards real-space analysis of supraparticles with icosahedral symmetry

Roy Hoitink, Ruizhi Yang, Alfons van Blaaderen

[1]Utrecht University, Netherlands

A lot of research has been conducted on studying how particles pack together when compressed in a slowly drying emulsion droplet, both in simulations and experiments. It is therefore widely known that such a system of particles equilibrates into structures that differ much from the bulk FCC-like, but rather form structures with icosahedral symmetry [1]. Computer simulations have the benefit of directly obtaining particle coordinates, in experiments this is much less trivial, but nevertheless not less important to see if it resembles predicted theory. Most experimental research up to now is not able to give detailed information about the internal structures of these supraparticles and therefore also about the pathway towards them [2,3]. Only few have obtained coordinates by electron tomography [4], but we are not aware of any other papers that employ real-space techniques for obtaining full 3D structural information. By using silica nanoparticles with a fluorescent core, we are able to form supraparticles with icosahedral symmetry at reasonable timescales and also (almost) able to obtain the real-space coordinates of the final structures using laser scanning confocal and stimulated emission depletion (STED) microscopy. These results are a first step in direction of revealing the experimental pathway towards icosahedral supraparticles.

[1] B. de Nijs, S. Dussi, F. Smallenburg, J. Meeldijk, D. Groenendijk, L. Filion, A. Imhof, A. van Blaaderen, M. Dijkstra, Nature. Mater., 14, 56-60 (2014)
[2] J. Wang, C. Mbah, T. Przybilla, B. Apeleo Zubiri, E. Spiecker, M. Engel, N. Vogel, Nat. Commun., **9**, 5259 (2018)
[3] R. Ohnuki, N. Kunimoto, Y. Takeoka, S. Yoshioka, Part. &. Part. Syst. Charact., **39**, 2100257 (2022)
[4] D. Wang, T. Dasgupta, E. van der Wee, D. Zanaga, T. Altantzis, Y. Wu, G. Coli, C. Murray, S. Bals, M. Dijkstra, A. van Blaaderen, Nat. Phys., **17**, 128-134 (2020)

Understanding of clustering dynamics and phase separation in a mixture of active and passive colloids

Tanmay Biswas, Snigdha Thakur

TU Wien, Austria

Self-propulsion has important roles in some biological and chemical activities. The aim is to study the formation of self-propelling clusters. Here we take two types of colloids, active and passive, which are initially oriented randomly. Particles are active in the sense that they take part in a chemical reaction to produce product particles. The passive/inert particles are kept in the surround to sense the environment created by the active sphere. This self-assembly can then form active clusters. This study is to explore the cluster kinetics in a suspension of active and passive colloids. Our focus is to highlight the effect of activity on the clustering phenomena in a fluidic environment.

[1]P. Chuphal, I. Venugopal, S. Thakur, Bull. Mater. Sci., 43, 183 (2020)
[2]E. Westphal, S. Singh, C. Huang, G. Gompper, R. Winkler, Computer Physics Communications, 185, 495-503 (2014)
[3]K. Binder, D. Stauffer, Phys. Rev. Lett., 33, 1006-1009 (1974)
[4]T. Yu, P. Chuphal, S. Thakur, S. Reigh, D. Singh, P. Fischer, Chem. Commun., 54, 11933-11936 (2018)

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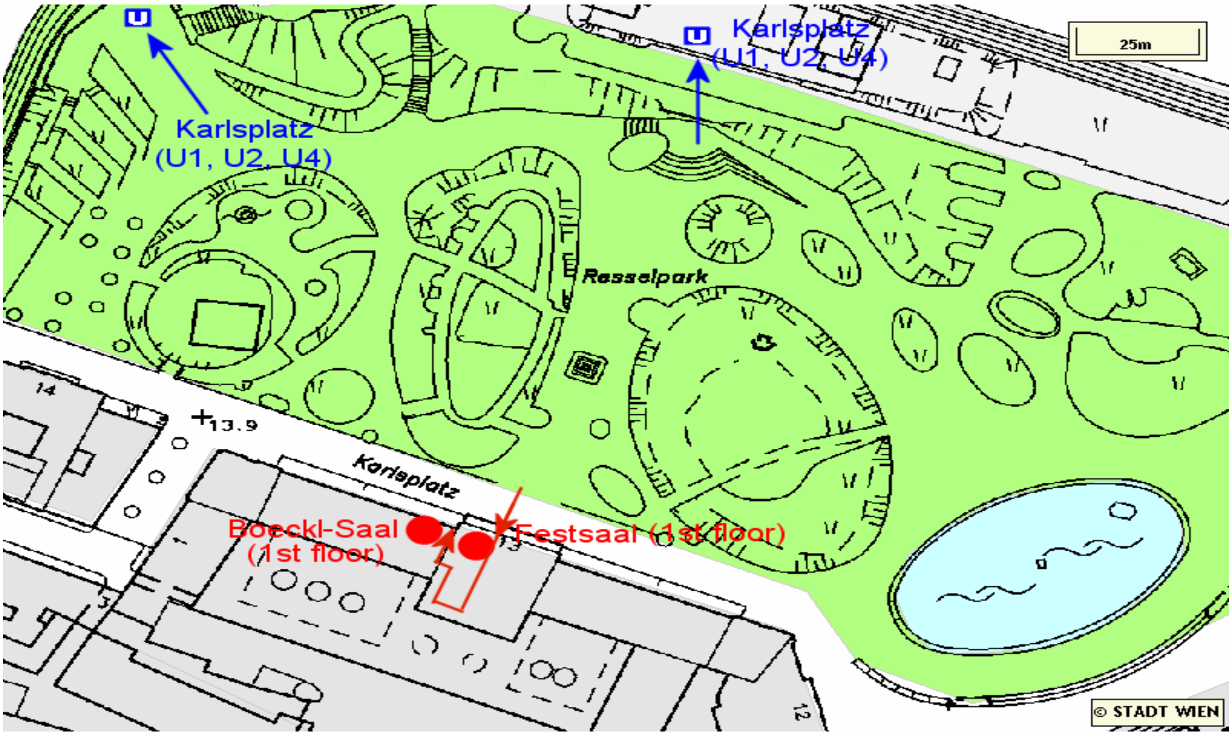
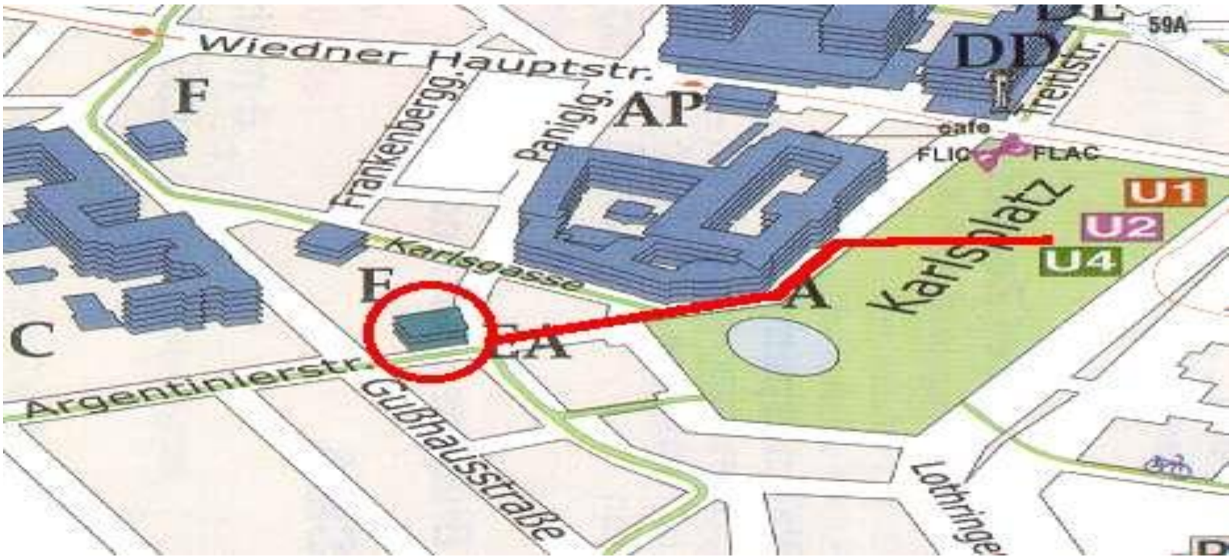
Maps- Location

Site of the Workshop

Adress: Karlsplatz 13, 1040 *Wien*, 1.OG. Raumcode: AA 01 62.





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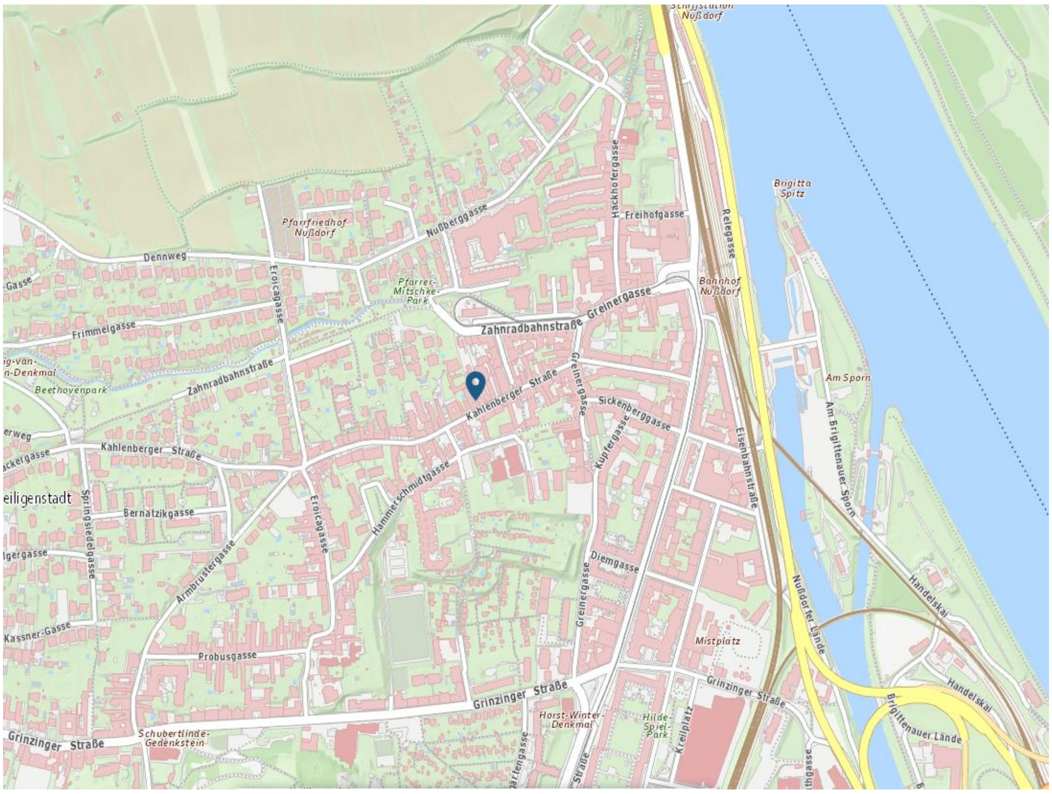
Karlsplatz 13, 1040 Wien, 1.OG. Raumcode: AA 01 62.

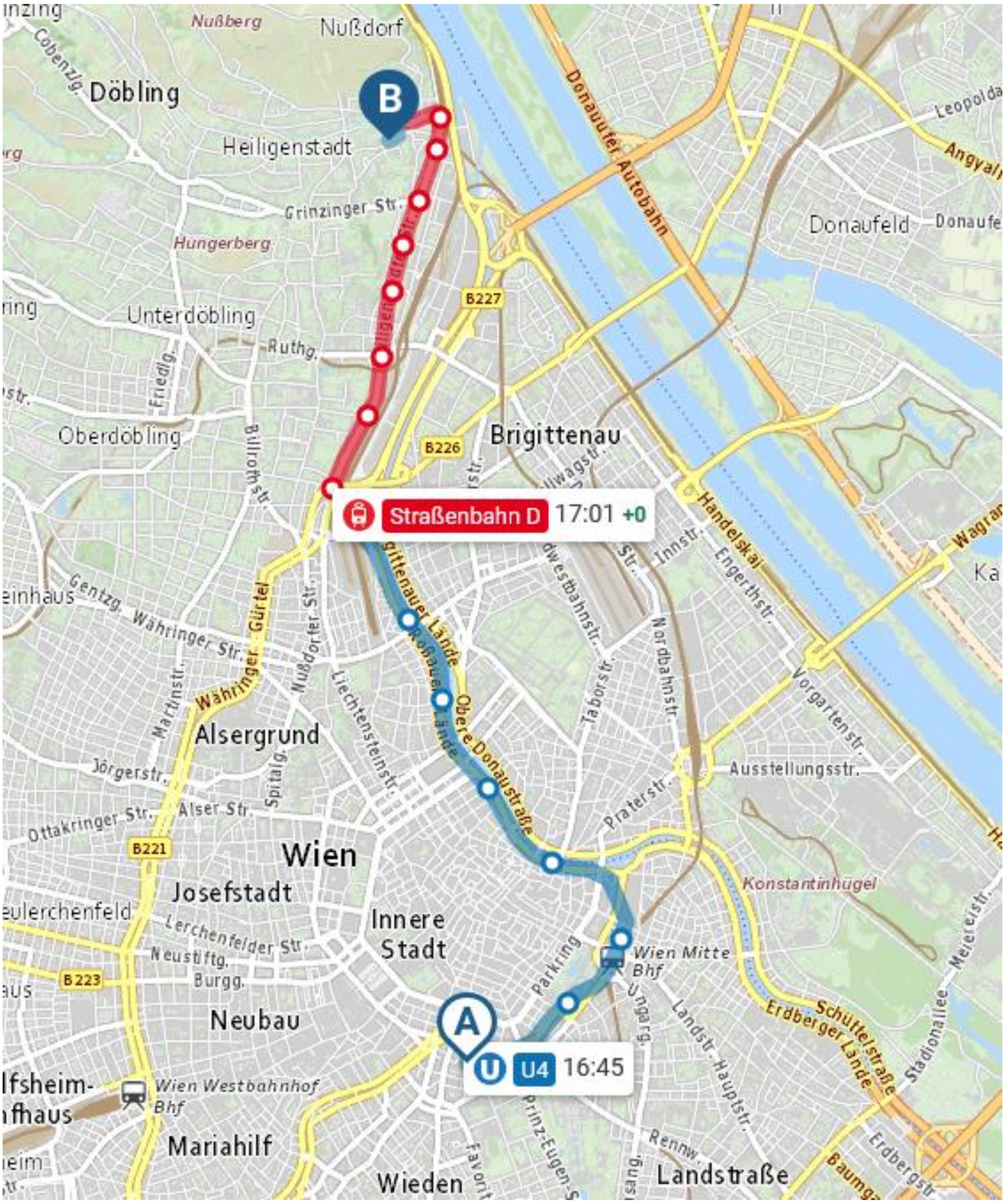
Additional Information

Restaurant (Dinner, July 12th, 2022)

Address: Kahlenberger Str. 22, 1190 Wien

Heuriger
SCHÜBEL-Auer
seit 1711




B
A
Straßenbahn D 17:01 +0
U U4 16:45
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Nußdorf
Döbling
Heiligenstadt
Grinzinger Str.
Hungerberg
Unterdöbling
Ruthg.
Oberdöbling
Billrothstr.
Brigittenau
Donauufer Autobahn
Donaufeld
B227
B226
Handelskai
Engerthstr.
Nordbahnstr.
Taborstr.
Gürtel
Währinger Str.
Währinger
Nußdorfer Str.
Liechtensteinstr.
Alsergrund
Jörgerstr.
Alser Str.
Ottakringer Str.
B221
Wien
Josefstadt
Innere Stadt
Lerchenfelder Str.
Neustiftg.
Burgg.
B223
Neubau
Wien Westbahnhof Bhf
Mariahilf
Wieden
Wien Mitte Bhf
Parkring
Ungarg.
Landstr. Hauptstr.
Landstraße
Rennw.
Prinz-Eugen-S
Obere Donaustraße
Rossauer Lände
Brigittenauer Lände
Praterstr.
Ausstellungsstr.
Vorgartenstr.
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