Complex Colloidal Self-Assembly: Towards Designer Materials

19 JAN 2023 – SOFT MATTER & SELF-ASSEMBLY WORKSHOP 4TU.HTM

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Spontaneous Self-Organization

- In nature: spontaneous organization of building blocks occurs
- Clear relationship macroscopic structure and microscopic building blocks







Bottom-up Assembly of Functional Materials

- Many novel building blocks: molecules, nanoparticles, colloids
- Employ spontaneous organization to make new materials



Colloids

- Size: 1-1000 nm
- "Soft, Slow & Seeable"





Glas



Functional Materials of Colloidal Building Blocks

Spontaneous Assembly



Photonic Crystals & Glasses

Image: space of the space of

Biomimicry









Cubes: a new frontier!

Advances in synthesis: cubic shaped (nano)particles have become available.



Rossi et al. (2011) Soft Matter



Zhang et al. (2011) PRL



Zhou *et al.* (2011) PNAS



Self-Assembled Nanomaterials of Cubes



Superball Self-Assembly - Shape Driven

Confocal Microscopy



SINGLE PARTICLE

- ✓ Spatial resolution
- ✓ Dynamics
- ✓ Interactions
- ✓ Self-assembly

Low concentration (full duration: 6 hours)



High concentration (4.3x real time)



Dense Rhombic Crystals

Small Angle X-ray Scattering



LOCAL & BULK

- Microscopic \checkmark
- Macroscopic \checkmark
- **Better statistics** \checkmark





e

Hollow-site

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Solid-Solid Phase Transitions Induced by Shape



Jiao *et al.* Phys Rev E 2009, Batten *et al.* Phys Rev E 2010, Ni *et al.* Soft Matter 2012

Driving Assembly with Solvent Evaporation

Convective Assembly



D = 774 ± 35 nm







 Λ_0 –lattice

 Λ_1 –lattice





J.M. Meijer et al, Langmuir (2012)

Different Structures in Dense Monolayer Structures





D = 774 ± 35 nm



D = 1028 ± 35 nm



D = 1266 ± 27 nm





Single-Particle Characterization of Lattices



Characterization of Lattices for different m

Different corner roundness



Lattice Frequency



J.M. Meijer et al. Langmuir (2019)

Superstructures of Attractive Superballs



Rossi et al. (2011) Soft Matter



 $q = 2R_g / L$



Rossi et al. (2015) Soft Matter

In-situ Control over Attractions

Critical Casimir force



Nguyen et al Nat Comm 2016

TU/e

Stuij, PhD Thesis, UvA, 2020

Cubes + Critical Casimir force

Anisotropic interactions

μm 10 Potential (k_bT) ΔT=0.14K ΔT=0.02K -10

In-situ control of self-assembly process





Formation of Different Superstructures

Square



Hexagonal





Lattice Analysis at Different Attraction Strength

Square



Hexagonal









Shape Controlled Switching Structures





0.2

Δ*T* (°C)

0.4

0.0

1.00

0.00

Summary: Shape, Interactions & Pathway Matters

Cubic particles







Hard body interactions





Long-range Attractions and Shear





Short-range Attractions





Acknowledgements

Van 't Hoff Laboratory **Utrecht University**

- Antara Pal ٠
- Samia Ouhajji
- **Fabian Hagemans** .
- Vera Meester ٠
- Andrei Petukhov ٠
- Albert Philipse ٠
- Henk Lekkerkerker ٠
- Laura Rossi .
- **Dmytro Byelov** ٠
- Jan Hilhorst ٠
- Anke Leferink op Reinink ٠
- Joost Wolters .
- Jos van Rijssel •

Hasylab, DESY

- Ivan Vartanyants
- Anatoly Shabalin
- Alexey Zozulya
- Sergey Lazarev

HHU Dusseldorf

- Astrid Rauh •
- Marco Hildebrandt
- Matthias Karg

SWING, SOLEIL

Javier Perez



Jan Maarten van Doorn

Joris Sprakel

ID06 & DUBBLE

ESRF, Grenoble

Soft Matter, IoP,

Peter Schall

Physics, TU/e

Piet Swinkels

Daphne Sayasilpi

Steffen Lootsma

Chris Kennedy

Kees Storm

Tine Stevens

Max Schelling

Ties van de Laar

Anatoly Snigirev

Guiseppe Portale

Daniel Hermida Merino

University of Amsterdam

Soft Matter and Biological

Irina Snigireva

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DÈŚY



Funding:

Unterstützt von / Supported by



Alexander von Humboldt Stiftung/Foundation

SFB 1214







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Thank you!



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