Colloidal Phase Transitions under Microgravity

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NASA Advanced Colloids Experiments (ACE) Program on ISS:

ACE-T11: Colloids of spherical particles, Sept 2020 - July 2021

ACE-TR Ellipsoids: Colloids of ellipsoidal particles, Sept 2020 - Feb 2021

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ISSR&D CONFERENCE **ID: 118**

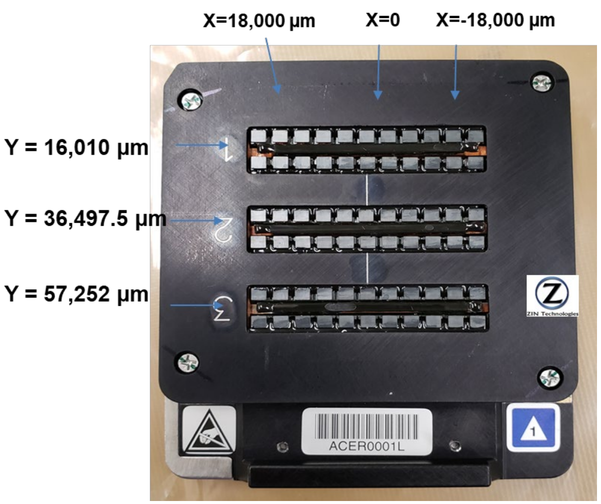
Why study colloids in microgravity?

- On Earth gravitational sedimentation confirms most colloidal systems to 2D with numerous artifacts due to floors and boundaries.
- Long-duration microgravity on the ISS literally adds another dimension to the problem making available 3D motion, 3D processing, 3D fundamental science, and 3D materials and devices

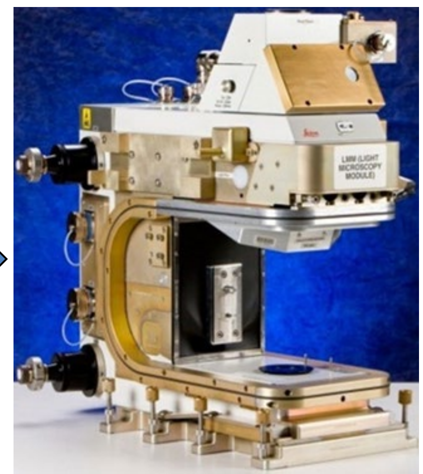
ISS INSTRUMENTATION

ISS Light Microscopy Module (LMM)

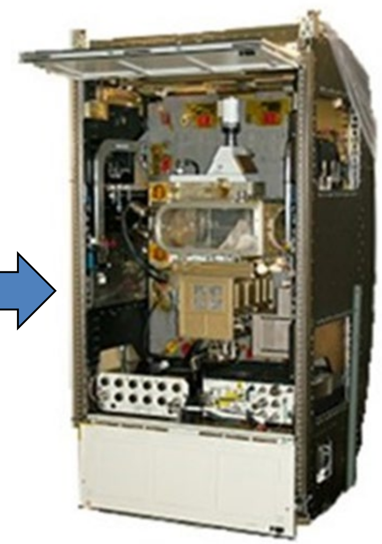
Approach: Use high-resolution confocal (3D) microscopy in long-duration microgravity on the ISS to investigate the colloidal structure at the particle level.



Sample Module



3D Microscope



Fluids Integrated Rack

ACE-T11:

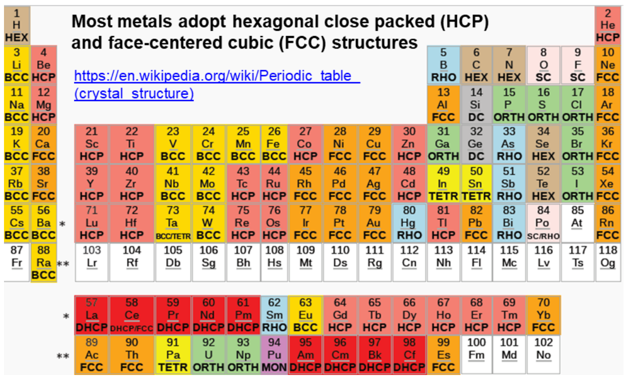
3 colloids of micro-spheres

ACE-TR:

6 colloids of micro-ellipsoids

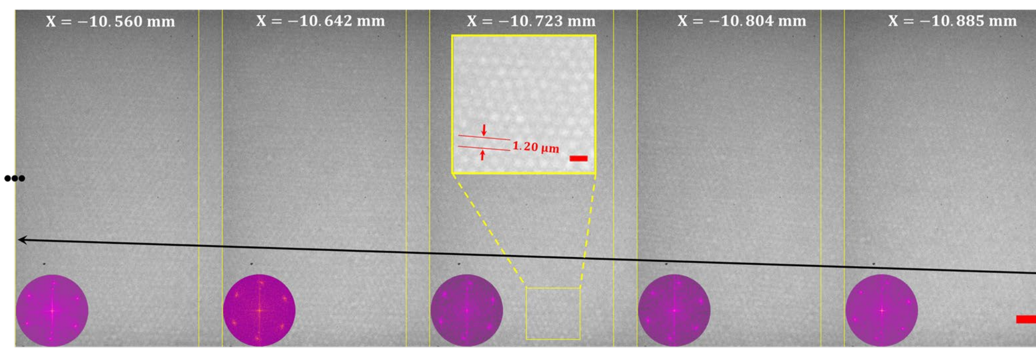
HARD-SPHERE COLLOIDS

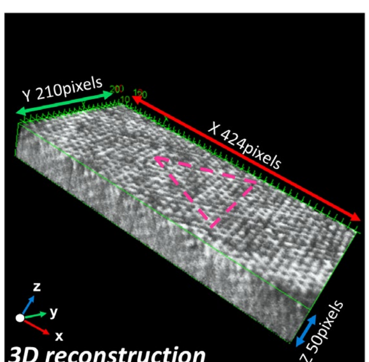
- Hard spheres is the basic model of particles in the statistical physics of fluids and solids. Defined as impenetrable spheres that cannot overlap in space, they mimic the extremely strong repulsion that atoms and spherical molecules experience at very close distances. Hard-sphere colloid provides a theoretical model for the structure and dynamics of simple liquids and enables direct measurements of the motion of individual particles with confocal microscopy.

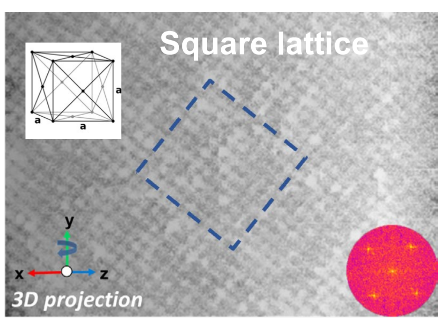


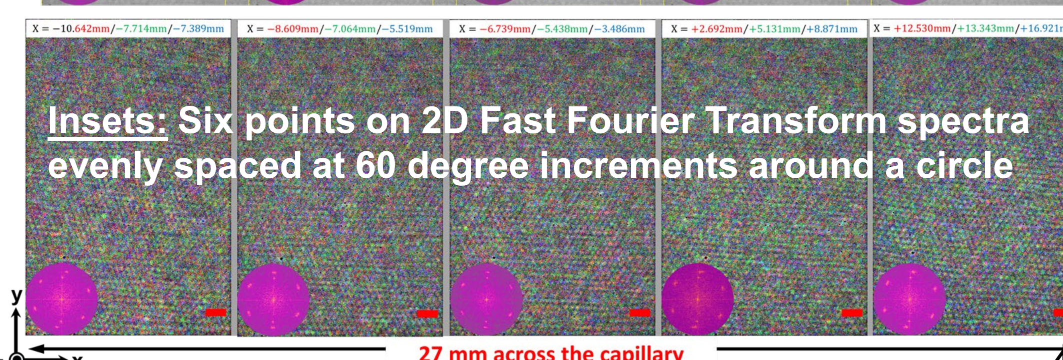
Up until now predictions of molecular dynamics simulations that the crystallization transition of hard-sphere colloid would occur from the liquid state to the **face centered cubic (FCC) crystal** had not been confirmed. These 3D crystals do not form on Earth because of sedimentation and gravitational jamming.

- **In microgravity, colloid formed a large FCC crystal of capillary size (27mm x 1.5mm x 0.15mm)**



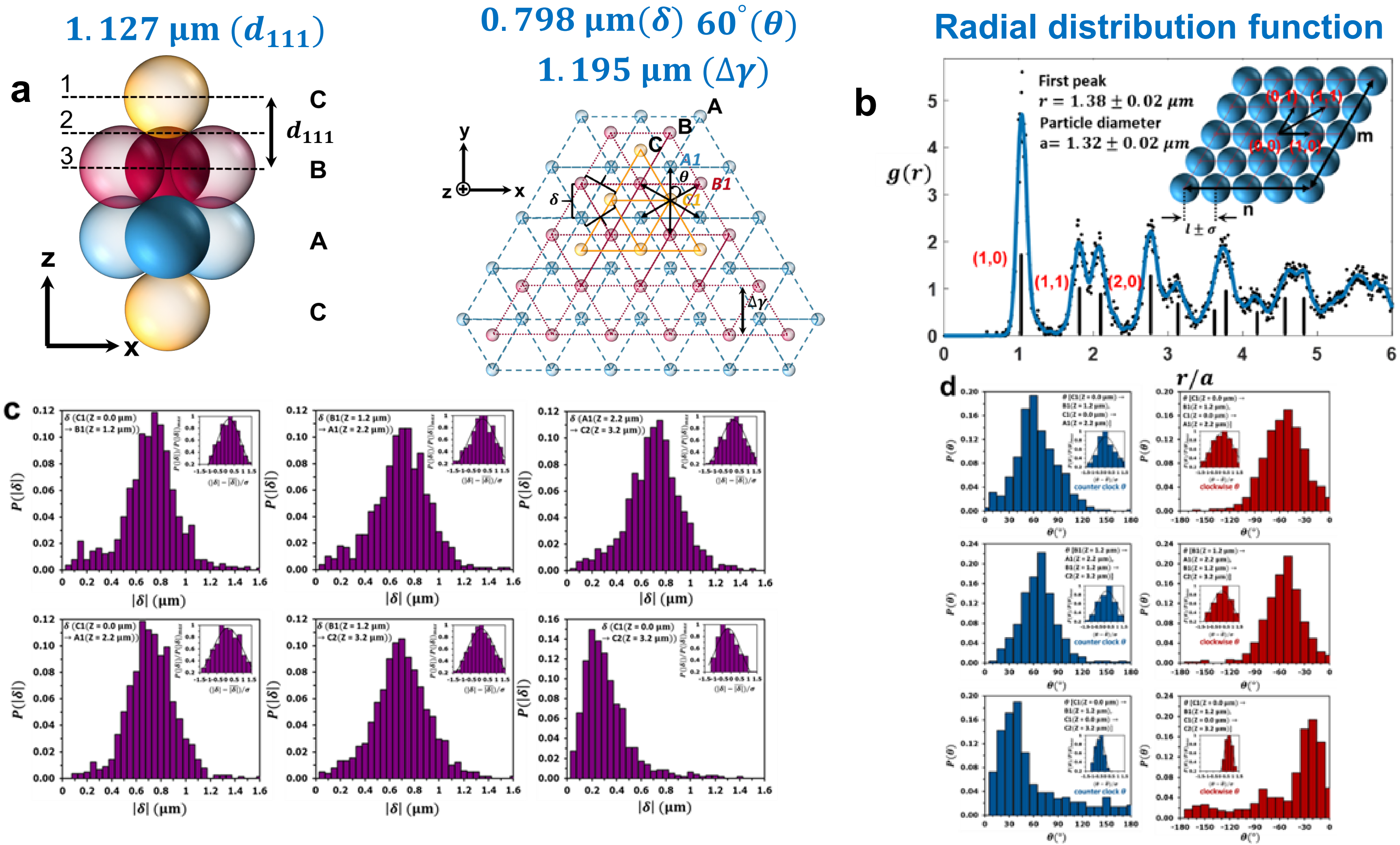






QUANTIFICATION OF FCC CRYSTAL

a and **b**, **Characteristics measured to identify the FCC crystal structure**

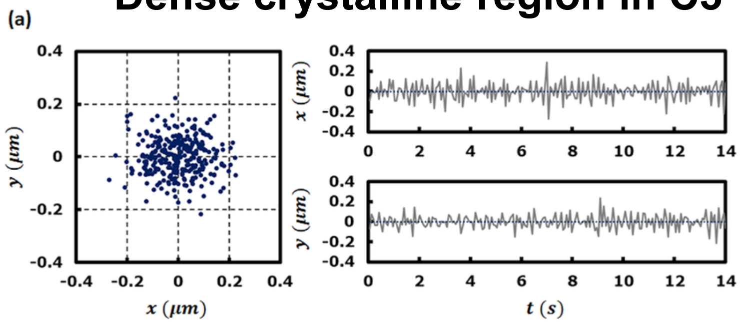


c and **d**, Histograms of separation $|\delta|, \Delta\gamma$ and angles θ between projections of the particle centers in four consecutive stacking layers computed for 3 stacks in different locations with 24 images in every stack. Inset shows Gaussian distribution for the histogram top part.

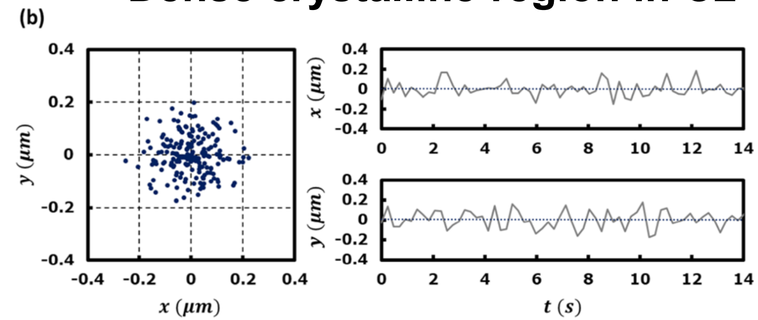
TRACKING OF PARTICLE MOTION

Entropy-driven crystallization

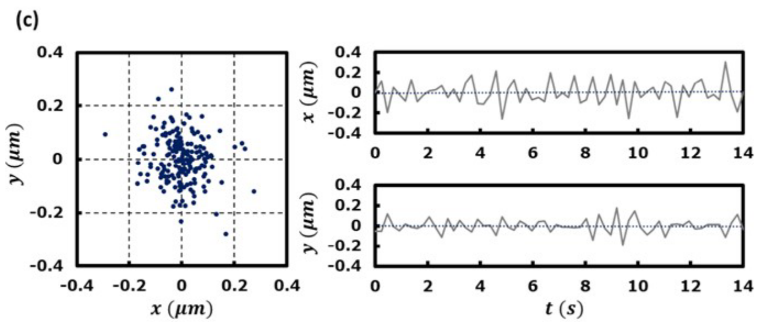
Dense crystalline region in C3



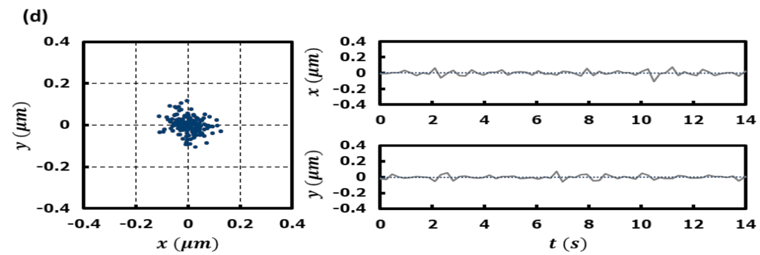
Dense crystalline region in C2



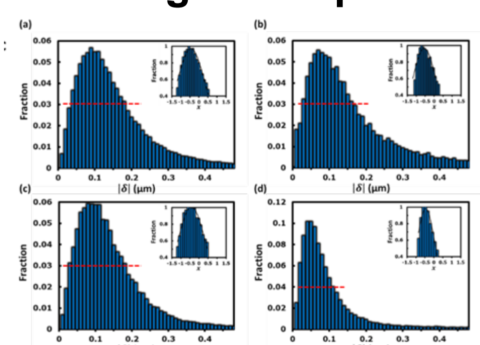
Low density non-crystalline region in C3



Dense non-crystalline region in C2



Histogram of particle displacements



	mean (μm)	STD (μm)	threshold fraction for the inset	Gaussian fitting curve R^2 for the inset
(a) Crystalline in C3	0.15	0.09	0.03	0.95
(b) Crystalline in C2	0.14	0.10	0.03	0.90
(c) Low density non-crystalline in C3	0.13	0.09	0.03	0.94
(d) Non-crystalline in C2	0.08	0.07	0.04	0.93

	2D Density (μm^{-2}), Mean $\pm$ STD
(a) Crystalline in C3	0.61 ± 0.01
(b) Crystalline in C2	0.67 ± 0.02
(c) Low density non-crystalline in C3	0.52 ± 0.01
(d) Non-crystalline in C2	0.63 ± 0.02

- Particle displacements in dense crystalline regions in are greater than in dense non-crystalline regions in, but smaller than in a low density non-crystalline region!
- The most frequent histogram part is Gaussian distribution for short-range Brownian diffusion of individual particle.
- Tail in the lower histogram part represents long intermittent jumps of particles.

TRANSLATIONAL PERIODICITY

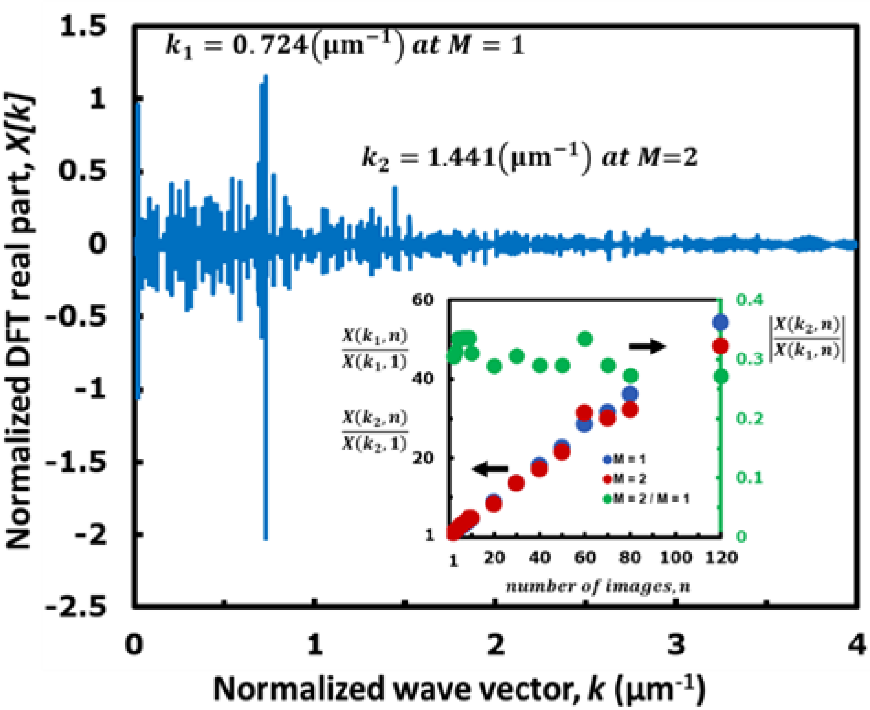
No imperfections in regular geometrical arrangement of particles in FCC crystal

Lattice constant $L = 1.38\mu m$ **Particle diameter** $a = 1.32\mu m$ **Gaussian STD** $\sigma = 0.15\mu m$

1D Fourier Spectrum

$$\langle \hat{X}_M \rangle = X_{aver} L(2N+1)\left[\frac{sin(\pi M a/L)}{\pi M}\right] e^{-2(\pi M \sigma/L)^2}$$

$$\left|\langle \hat{X}_2 \rangle / \langle \hat{X}_1 \rangle\right| \approx 0.49$$



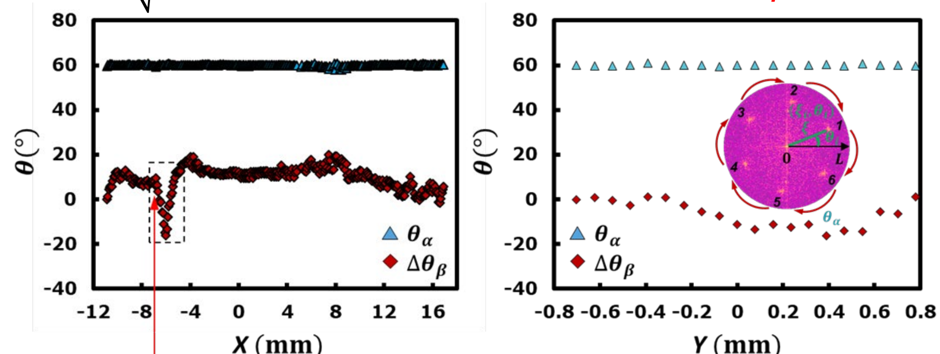
2D Fourier Spectra

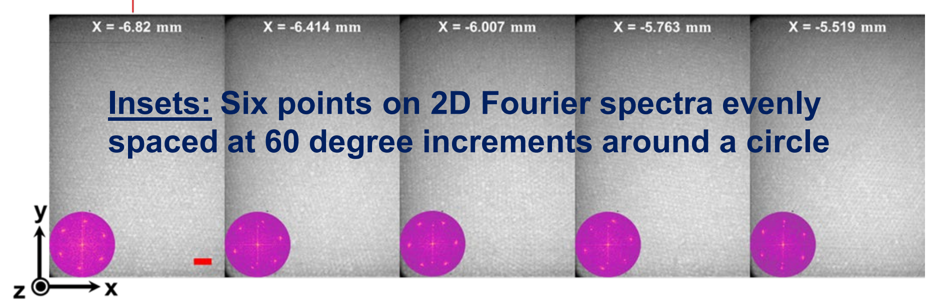
$$\langle \hat{X}_{M_1,M_2} \rangle = \pi a^2 somb(\pi F\, a/L) e^{-2(\pi F \sigma/L)^2} X_{aver}(2N_1+1)(2N_2+1)$$

Lattice nodes $$Lk_{M_1,M_2} = \left(M_1 - \frac{1}{2}M_2\right)e_x + \frac{\sqrt{3}}{2}M_2 e_y$$

$$F = \sqrt{M_1^2 - M_1 M_2 + M_2^2}$$

$$\theta_i = \theta_\alpha i + \Delta\theta_\beta(length)$$





Insets: Six points on 2D Fourier spectra evenly spaced at 60 degree increments around a circle

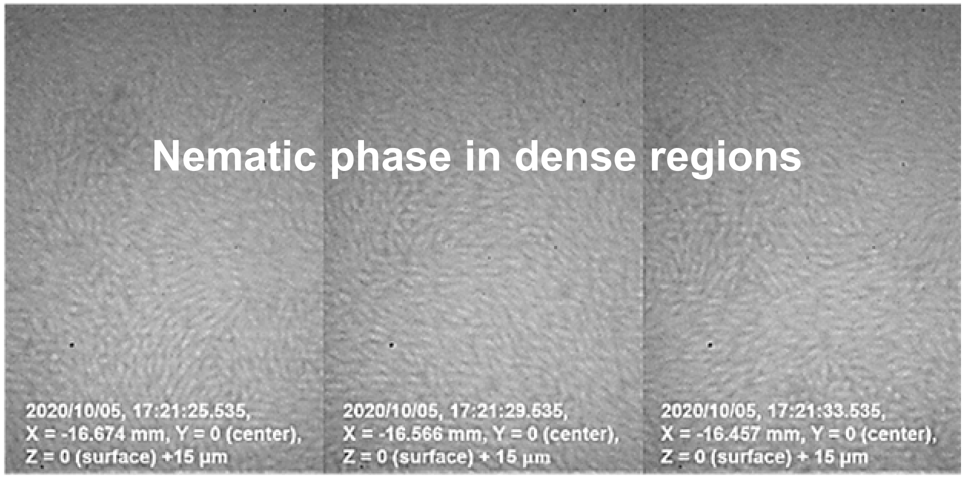
342 images in X direction; 20 images in Y directions.

HARD ELLIPSOIDS

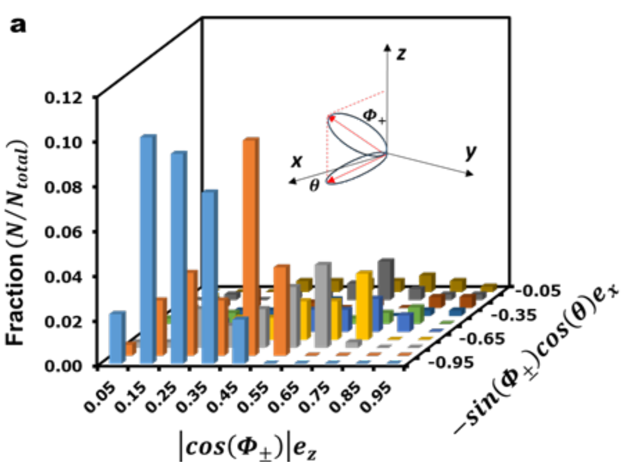
Colloid of hard ellipsoids provides a theoretical model for **quasi-universal structure and dynamics of liquid crystals**. Gravitational settling of particles on Earth does not allow testing phase transitions predicted by molecular dynamics simulations.

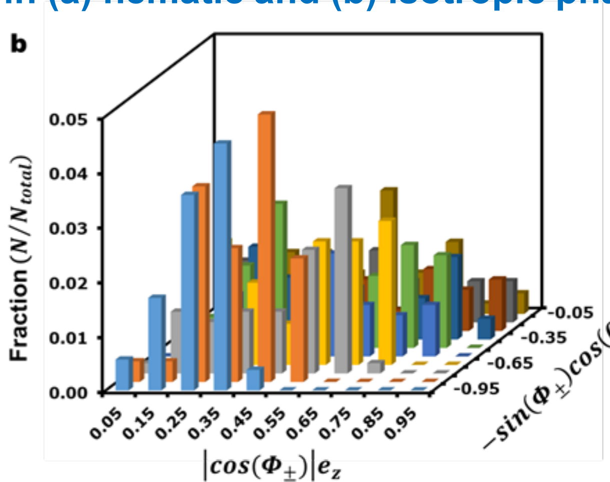
Ellipsoidal particles with the aspect ratio ~4

Domains of dense nematic phase (rafts of aligned particles) and isotropic phase in low-density regions



Orientation of ellipsoids in (a) nematic and (b) isotropic phases



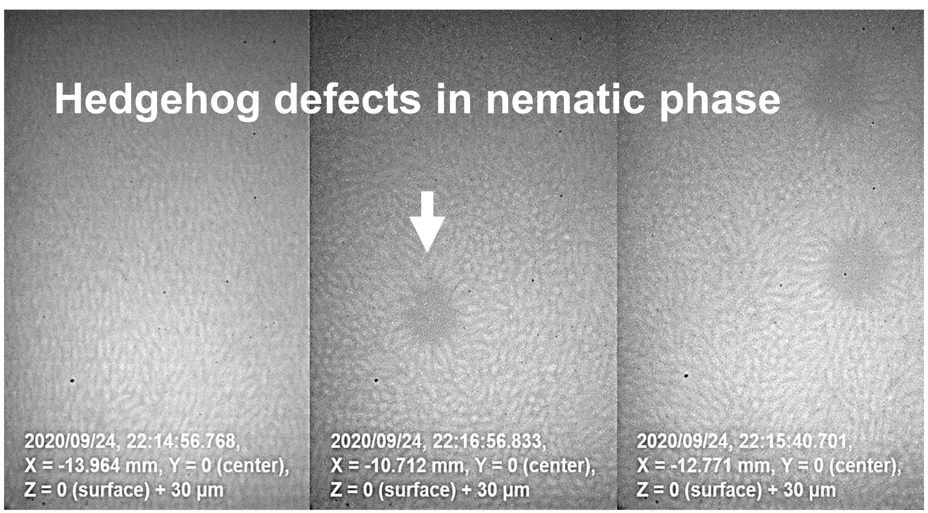


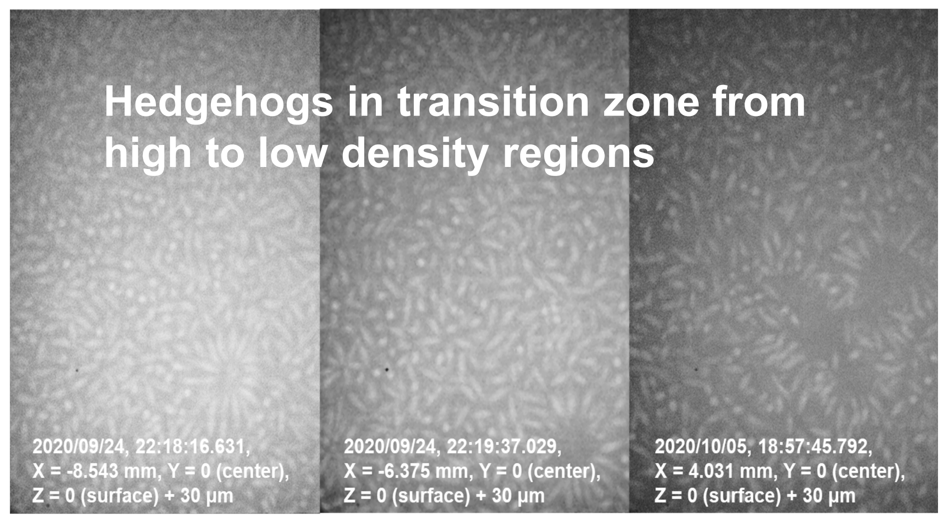
(a) High-density region:
X = -17.000 mm, Y = 0 (center),
Z = 0 (surface) + 30 μm
(b) Low-density region:
X = -8.453 mm, Y = 0 (center),
Z = 0 (surface) + 30 μm

ACE-TR ELLIPSOIDS

- Discovery of a similarity between the behavior of colloidal ellipsoids and liquid crystals, along with their structural defects, is an important finding.
- Micrometer-sized colloidal particles used in microgravity are huge in size compared to molecules in liquid crystals which are too small to be seen with an optical microscope. ISS measurements of the arrangement and motion of individual particles in the colloidal nematic and isotropic phases and around structural defects provide a unique opportunity for testing theories for liquid crystals.

Nematic domains and associated "hedgehog" defects





Colloidal structures formed in microgravity in X-Y plane parallel to cover slip; 50 $\mu m \times$ 70 μm images

COLLOIDAL PHOTONIC CRYSTALS

- An exciting aspect of the microgravity experiments on colloids in the ISS is the realization that in microgravity we can build large colloidal photonic materials with an ordered arrangement of particles that survive reentry to Earth.

- A photonic crystal is the optical analogy to an atomic lattice, in which the refractive index repeats periodically in on the scale of the light wavelength. The particle sizes, shapes and properties in colloidal photonic materials can be varied based on the wavelength of light we wish to control.

- Due to the unique properties, photonic materials operating at infrared wavelengths are expected to be crucial for applications in remote sensing, fiber-optic communication, chemical analysis, biomedical diagnostics, optical computing, security and defense.

- Fabrication of large-scale colloidal materials on Earth remains a challenge as the particle assembly is strongly influenced by gravity effects, such as particle sedimentation, convection and jamming.

- The following invention describes fabrication of large-scale colloidal photonic materials in a Low-Earth orbit and returning them to Earth:

M. Murphy (Nanoracks, LLC), Q. Lei (NJIT), B. Khusid (NJIT), A.D. Hollingsworth (NYU), P.M. Chaikin (NYU), W.V. Meyer (USRA). Patent Application # 63/365,667; 06/01/2022.

Summary of invention: A suspension of desirable colloidal particles transferred to a sample container and delivered to Low-Earth orbit (LEO). A long-duration microgravity environment in a LEO enables fabrication of colloidal crystals in non-buoyancy-matched suspensions without the detrimental effects of gravity.

CONCLUSIONS

Up until now the simplest, most common, most fundamental Crystallization Transition, Liquid – FCC Crystal, had not been confirmed!

- **Molecular Dynamics (MD) simulations now confirmed: Large FCC crystals observed in *μg***
- **Large nematic domains and associated "hedgehog" defects observed in *μg***
- **MD simulations of equilibrium confirmed to work only without gravitational effects**
- **MD simulations can be used for modeling equilibrium in materials processing in *μg***
- **Submitted a patent application for method & apparatus for fabrication in *μg* of large-scale colloidal photonic materials in a Low-Earth orbit**

Build for the Future†

- Non-equilibrium processing and dynamic materials/devices in *μg*
- Proceed to understand effects of 1g in materials growth and design
- Manufacture in *μg* better materials for ceramics, paints, photonics, pastes for Earth applications and space exploration

† *See Grand Challenges in Soft Matter Science: Prospects for Microgravity Research* - NASA/CP-20205010493

Thank you for your attention!

Any questions and comments

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