Phase transitions in colloids under microgravity

Qian Lei*, Boris Khusid, Lou Kondic

New Jersey Institute of Technology, Newark, NJ

Andrew D. Hollingsworth, Paul Chaikin

New York University, New York, NY

William V. Meyer

Universities Space Research Association at the NASA GRC, Cleveland, OH

Alton J. Reich

Streamline Automation LLC, Huntsville, AL

NASA Advanced Colloids Experiment (ACE) Program

NSF CBET grants 1832260 and 1832291, NASA grants 80NSSC19K1655
NASA Glenn Engineering and Research Support (GEARS) Contract: 80GRC020D0003
***Supported by NSF CBET grant 1832260**

MAR21-2021-B07.00004 APS MAR Meeting (virtual), March 15, 2021

MOTIVATION

- Despite the fundamental and technological importance, understanding of colloidal crystallization is still elusive. The origins and even existence of glass transition in concentrated colloids remain poorly understood and hotly debated.

- Experiments ACE-T11 on spherical colloidal particles and ACE-TR-Ellipsoids on ellipsoidal particles apply high-resolution confocal microscopy to capture the colloid formation process over time.

- Understanding of how a disordered arrangement of particles self-organizes into a three-dimensional ordered structure at the particle level is vital for a variety of industries from 3D printing to photonics and electronics and to chemicals and pharmaceuticals.

MEASUREMENT APPROACH

LMM Implementation Philosophy

Philosophy: Maximize the scientific results by utilizing the existing LMM capabilities. Develop small sample modules and image them within the LMM

Light Microscopy Module (LMM)

Payload specific and multi-user hardware customizes the FIR in a unique laboratory configuration to perform research effectively.

Payload Specific Hardware

- Sample Cell with universal Sample Tray
- Specific Diagnostics
- Specific Imaging
- Fluid Containment

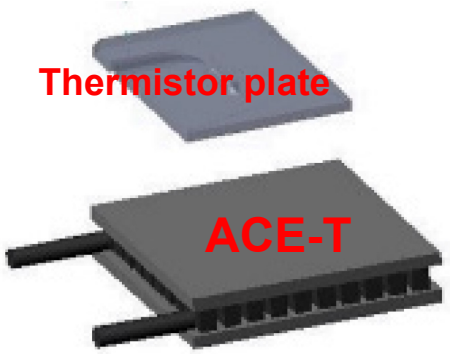
Multi-Use Payload Apparatus

- Test Specific Module
- Infrastructure that uniquely meets the needs of PI experiments
- Unique Diagnostics
- Specialized Imaging
- Fluid Containment

FCF Fluids Integrated Rack

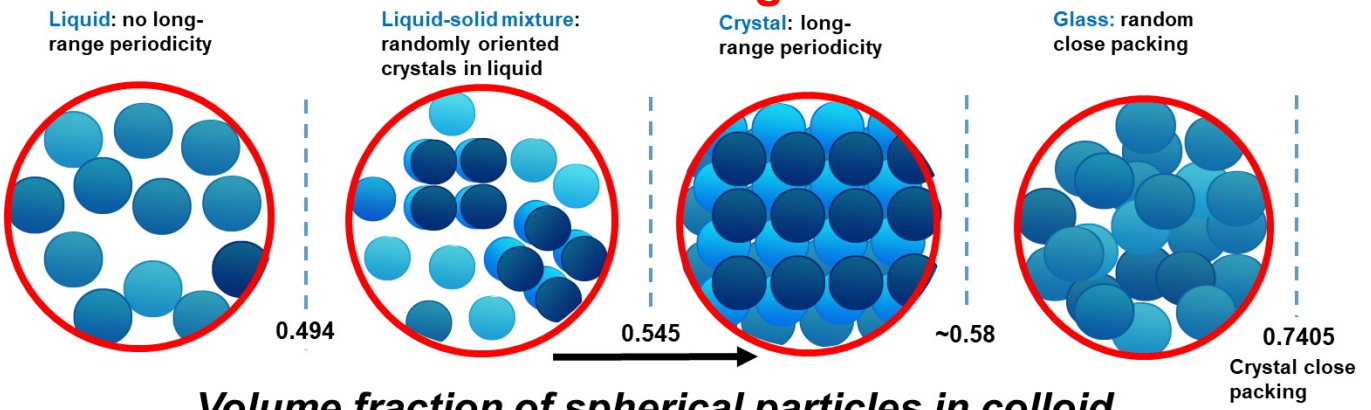
- Power Supply
- Avionics/Control
- Common Illumination
- PI Integration Optics Bench
- Imaging and Frame Capture
- Diagnostics
- Environmental Control
- Data Processing/Storage
- Light Containment
- Active Rack Isolation System

ACE-T11 SPHERES

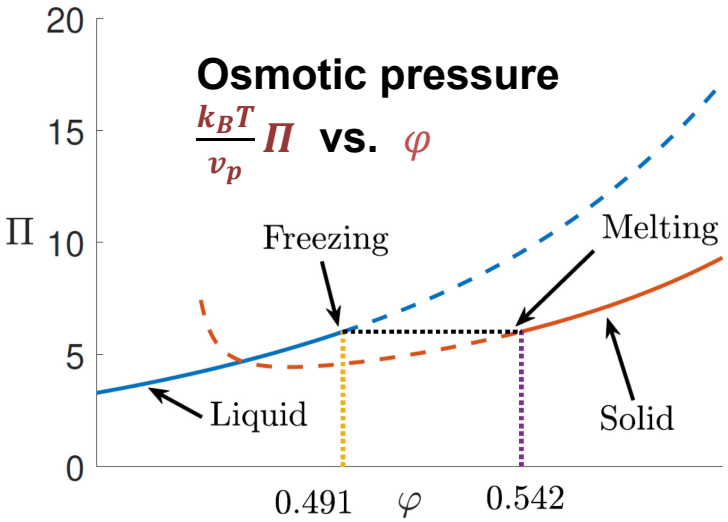


ACE-T Cell, ZIN-Technologies, OH

Observations at 1g



Volume fraction of spherical particles in colloid



MD simulations Speedy 1997, 1998

MD simulations & Experiments

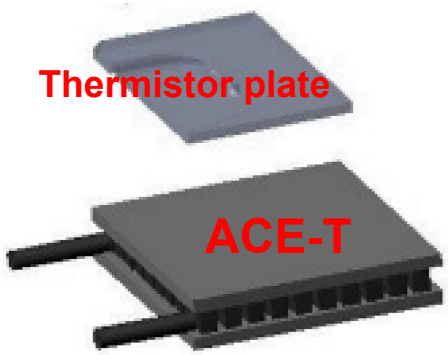
Liquid: $\varphi < \varphi_{fr} \approx 0.49$

Crystalline up to maximum packing:
$\varphi_m \approx 0.54 < \varphi < \varphi_0 = \sqrt{2}\pi/6 = 0.74$

Coexisting liquid & crystalline phases:
$\varphi_m > \varphi > \varphi_{fr}$

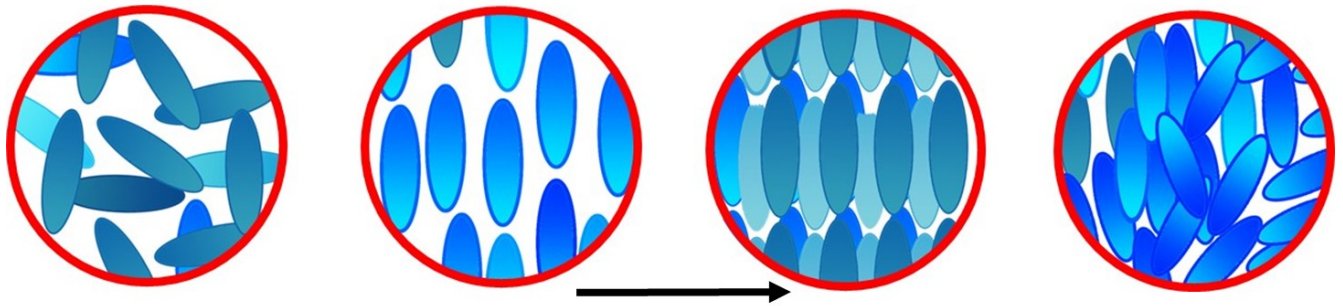
Reviews: *Gasser 2004, Palberg 2014*

ACE-TR ELLIPSOIDS



ACE-T Cell, ZIN-Technologies, OH

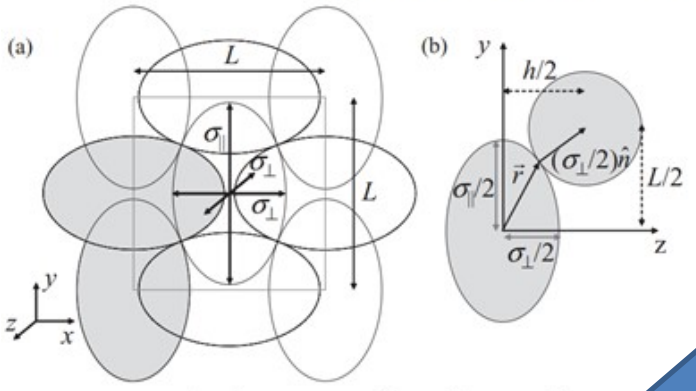
Observations at 1g



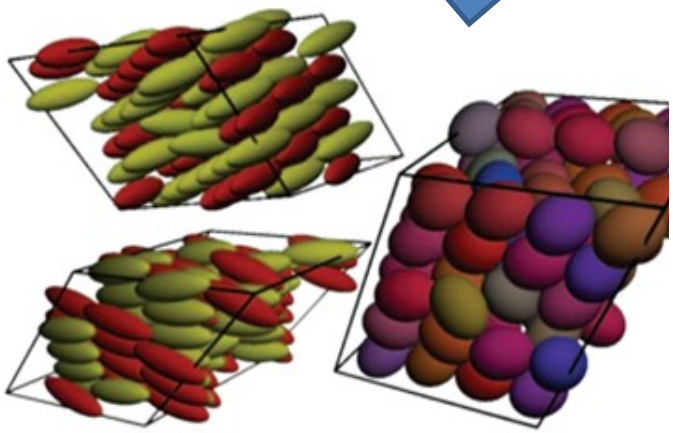
Volume fraction of elliptical particles in colloid

PHASE DIAGRAM HARD ELLIPSOIDS

Bautista-Carbajal, Moncho-Jordá, and Odriozola
J. Chem. Phys. **138**, 064501 (2013)



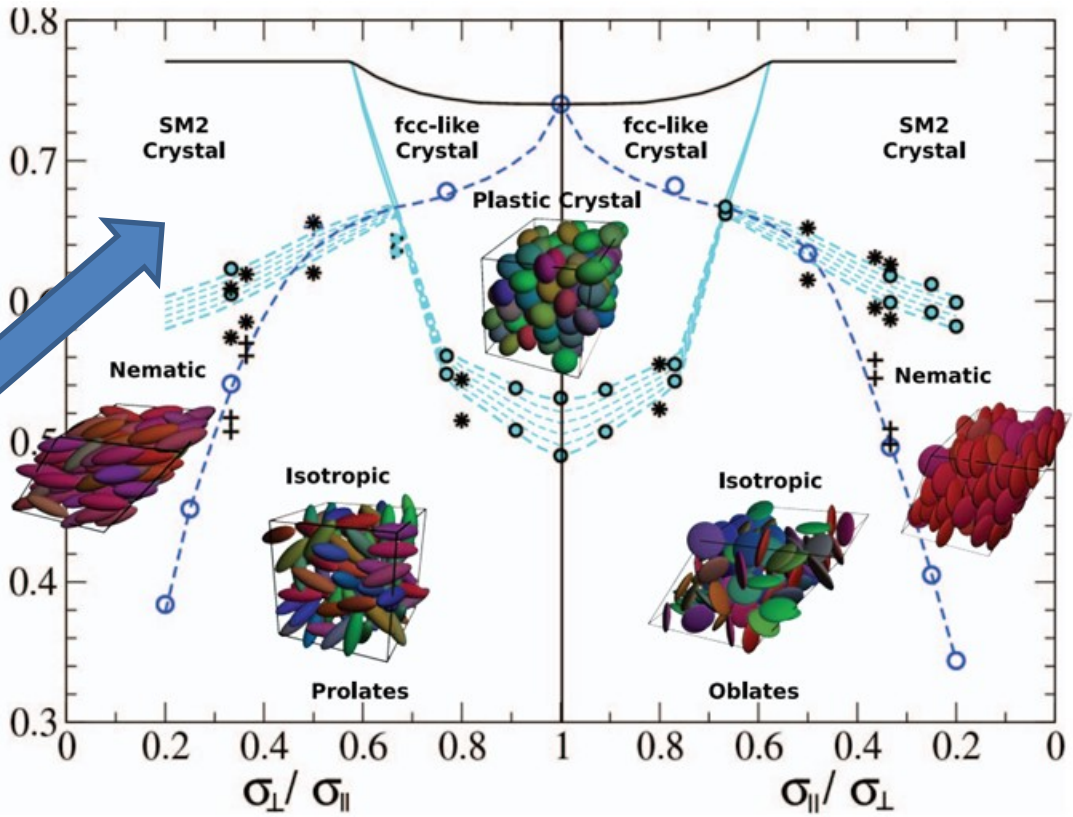
prolate sm2 structur



Prolate SM2 Crystal

Prolate fcc-like Crystal

MD simulations



Odriozola
J. Chem. Phys. **136**, 134505 (2012)

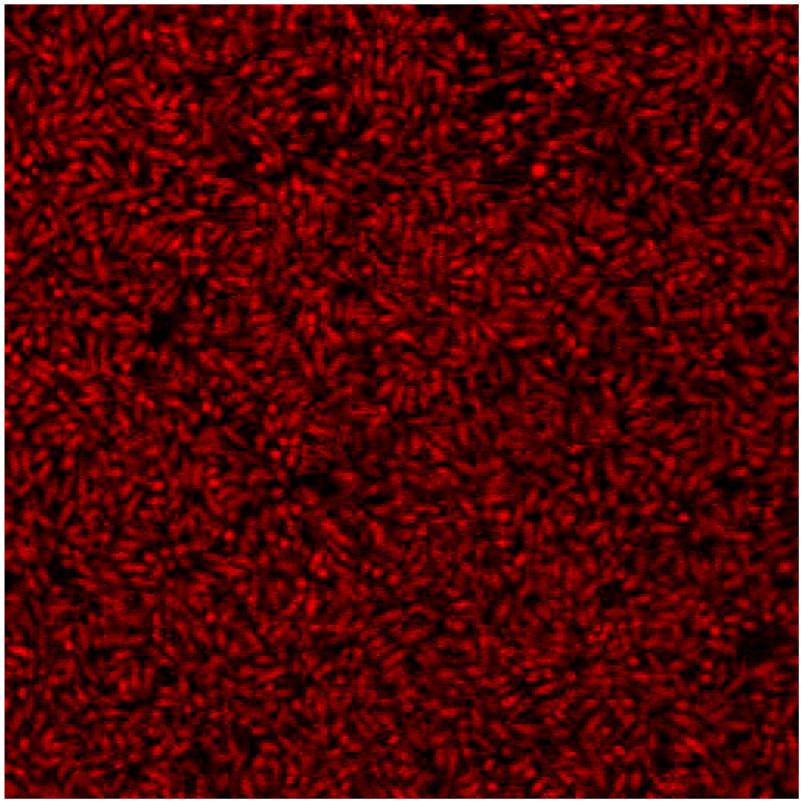
SYNTHESIS STRATEGY

Andrew D. Hollingsworth, NYU

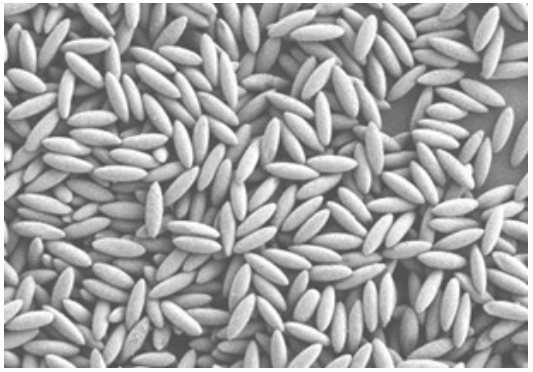
ELLIPSOIDAL COLLOIDS

- 1 to 2 micron dia. PHS-stabilized PMMA spheres
- flurorescent labeling
- disperse PMMA spheres in PDMS prepolymer
- cast/cure PDMS film
- thermomechanical stretching / T soak / quench
- harvest ellipsoids
- photocrosslinking
- post-processing

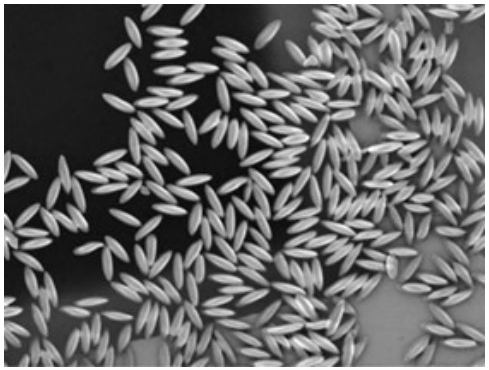
ELLIPSOIDAL COLLOIDS



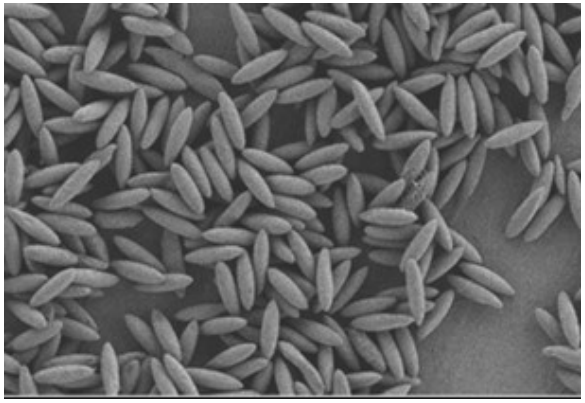
Leica SP5 confocal
63 X glycerol, 543 nm HeNe



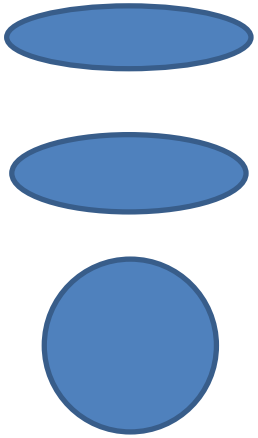
CSF-02-139A pulls 5 6b 7 8 9
$e = 0.8 \quad \alpha \approx 3.0$



CSF-02-139B pull 1
$e = 1.0 \quad \alpha \approx 3.7$

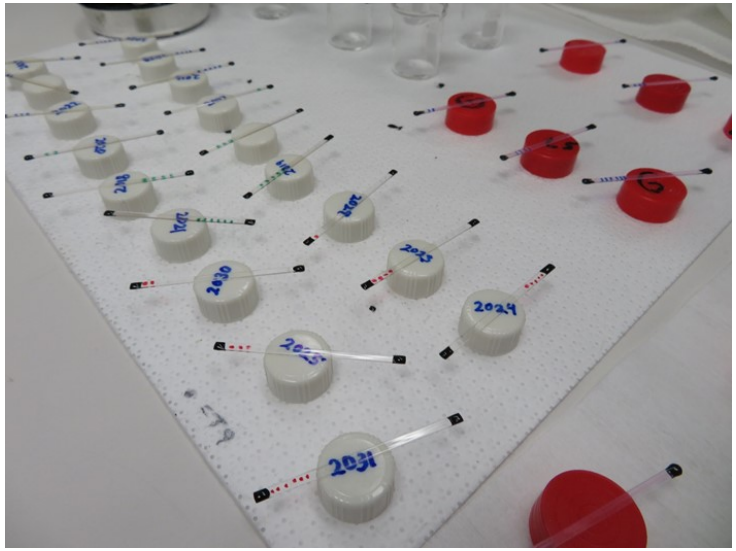


CSF-02-139C pull 10
$e = 1.0 \quad \alpha \approx 3.8$



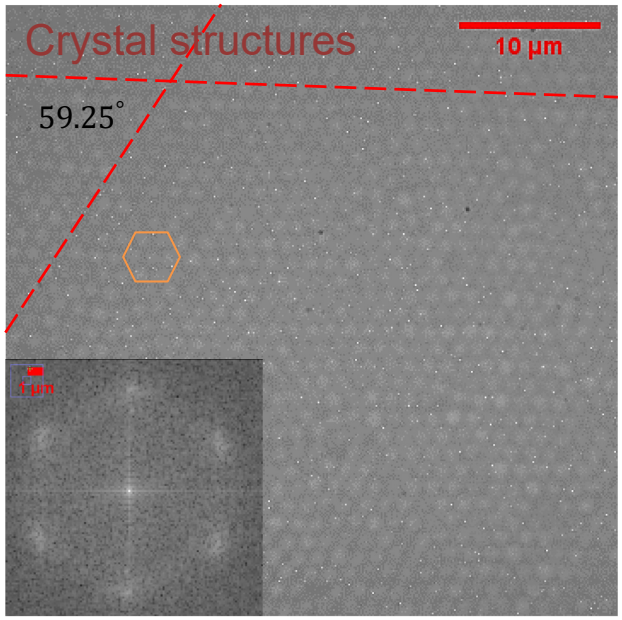
ACE-TR ELLIPSOIDS SAMPLE FILL JAN 2, 2020

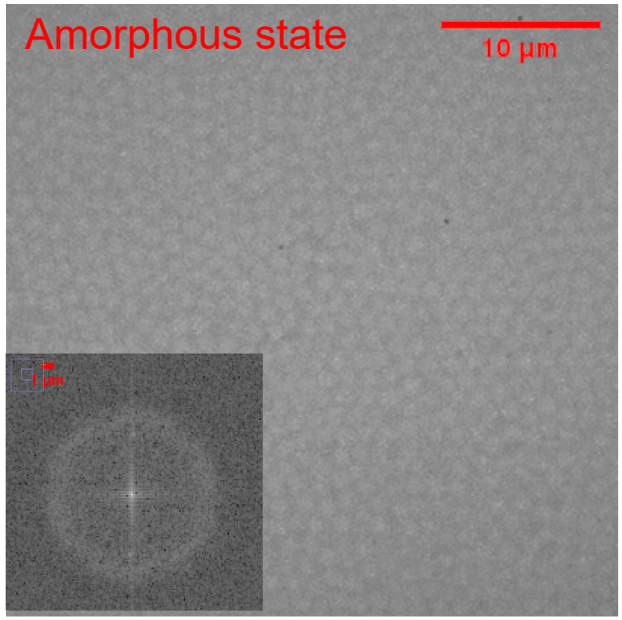
ZIN-Technologies



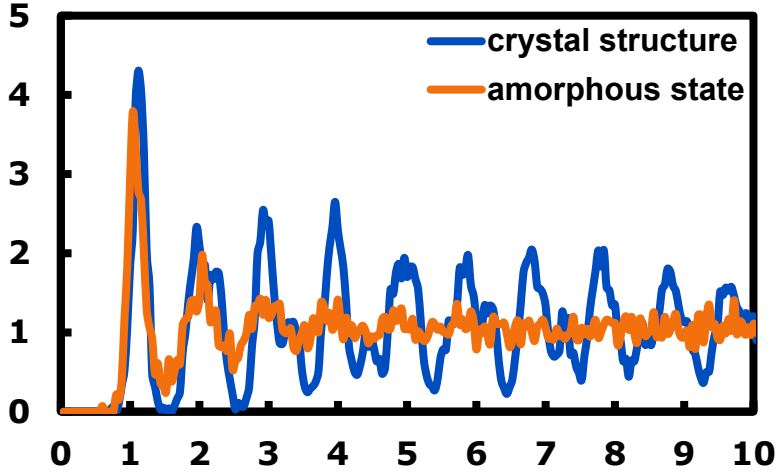
SPHERES

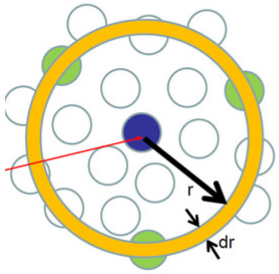
Comparison of crystalline and non-crystalline structure ACE-T11



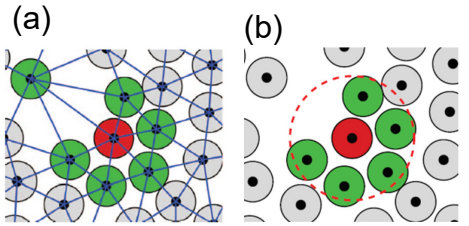


Radial Distribution Function g(r/D)

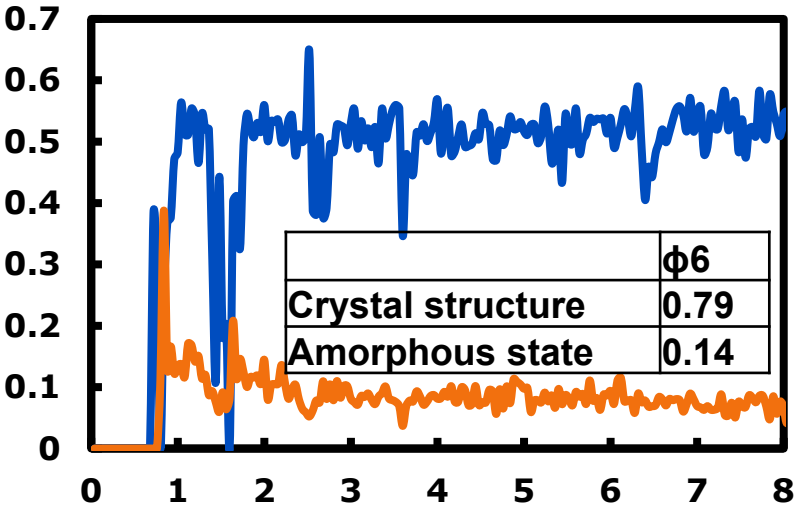




$$g(r) = \rho(r)/\rho$$



6-fold bond-orientational correlation function $g_6(r/D)$



$g_m(r) = \langle \varphi_m(0)\varphi_m(r) \rangle$ When N_b = 4,5,6,7, only center particles with nearest m = 4, 5, 6 ,7 neighbors are used to compute $g_m(r)$ and φ_m,

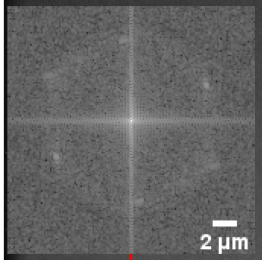
$$\varphi_m = \left| \frac{1}{N} \sum_N \left(\frac{1}{N_b} \sum_{k=1}^{N_b} \exp(im\theta_k) \right) \right|$$

Where N_b is the number of nearest neighbors(NN) and θ_k is the angle made by the nearest m neighbor bond vector with an arbitrary fixed reference X-axis. (a) Delaunay definition of NN: the Delaunay neighbors of the red sphere are highlighted in green. (b) NN definition with cutoff radius r_c. Here (b) used

SPHERES

Coexistence of crystalline and non-crystalline structures

ROIs: fixed 32um*32um



1 Crystal structure

Interval 8um

0um

2

3

4

h

Order – disorder transition

$A_{particle} = 1.13(um^2)$

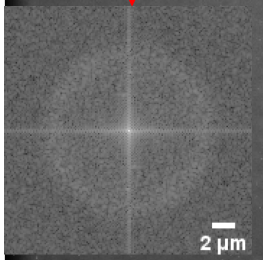
5 Amorphous state

Particle diameter (D) = 1.25um

6

48um

7



	h(um)	ρ(um^{-2})	Area%	φ6	ρ(um^{-2})*1.13(um^2)
1	0.00	0.61	35.00	0.38	0.69
2	8.00	0.60	34.53	0.36	0.68
3	16.00	0.60	33.44	0.23	0.68
4	24.00	0.60	32.61	0.13	0.68
5	32.00	0.57	32.08	0.11	0.64
6	40.00	0.62	32.75	0.13	0.70
7	48.00	0.67	32.00	0.10	0.75

7 6 5 4

1

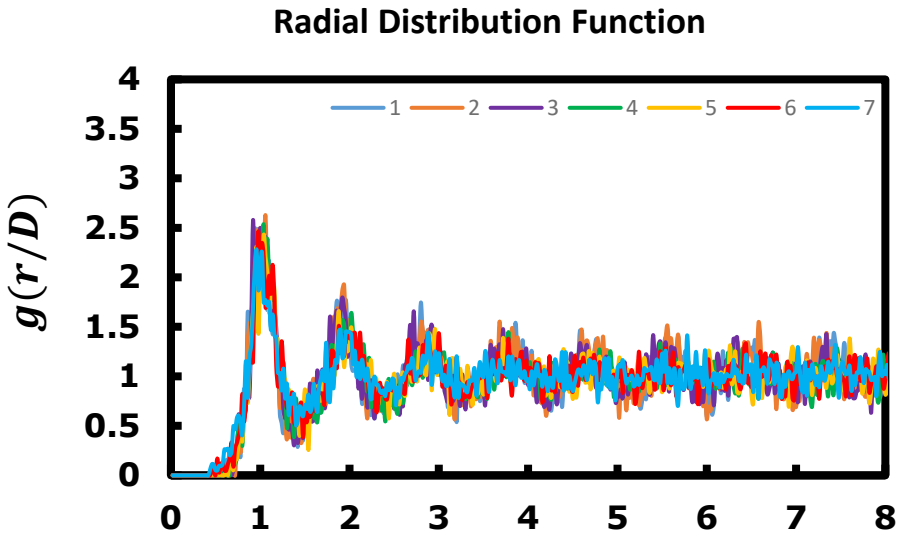
2

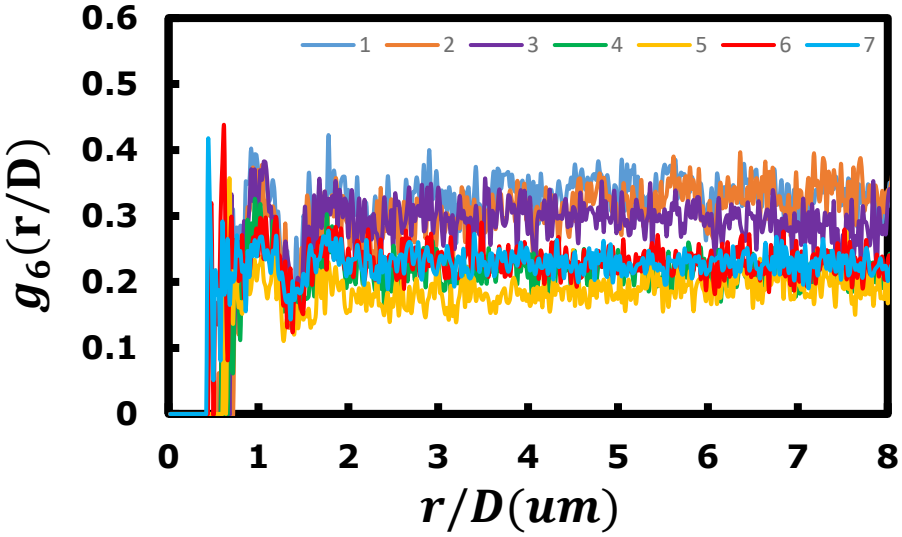
3

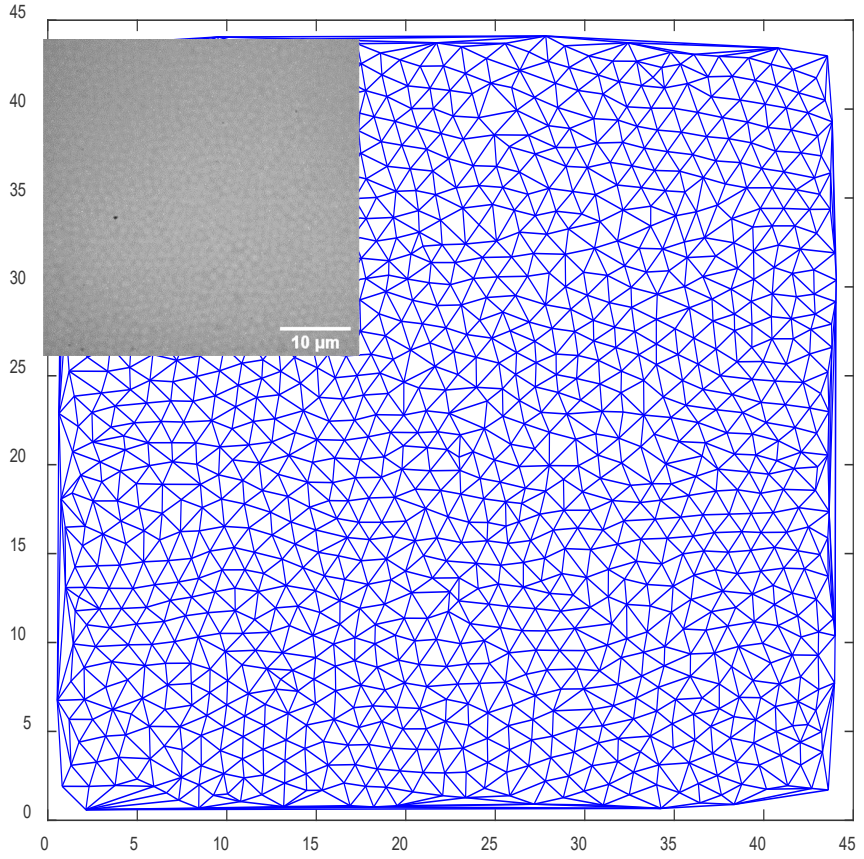
2020/11/22, 21:23:50.501, CnFcl, ACET11, S2017, C3, Y-20, Z0+120, X100, X-12186, P11, E1053922, 00010

SPHERES

Coexistence of crystalline and non-crystalline structures





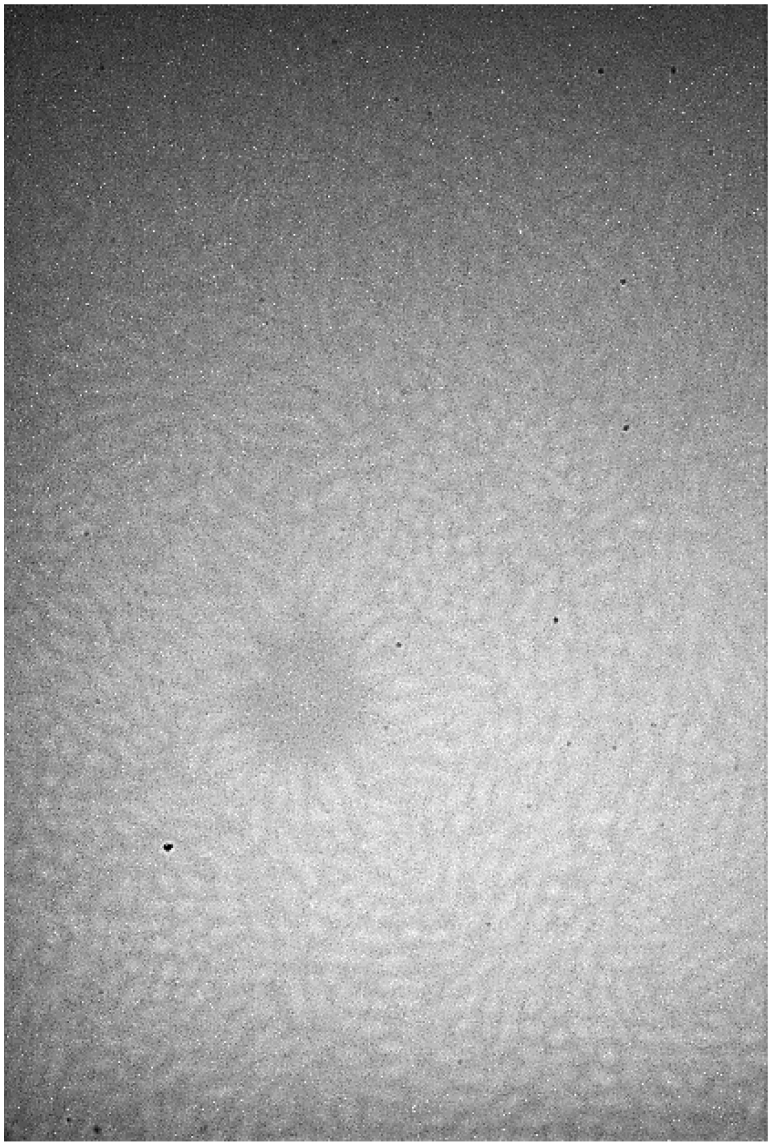


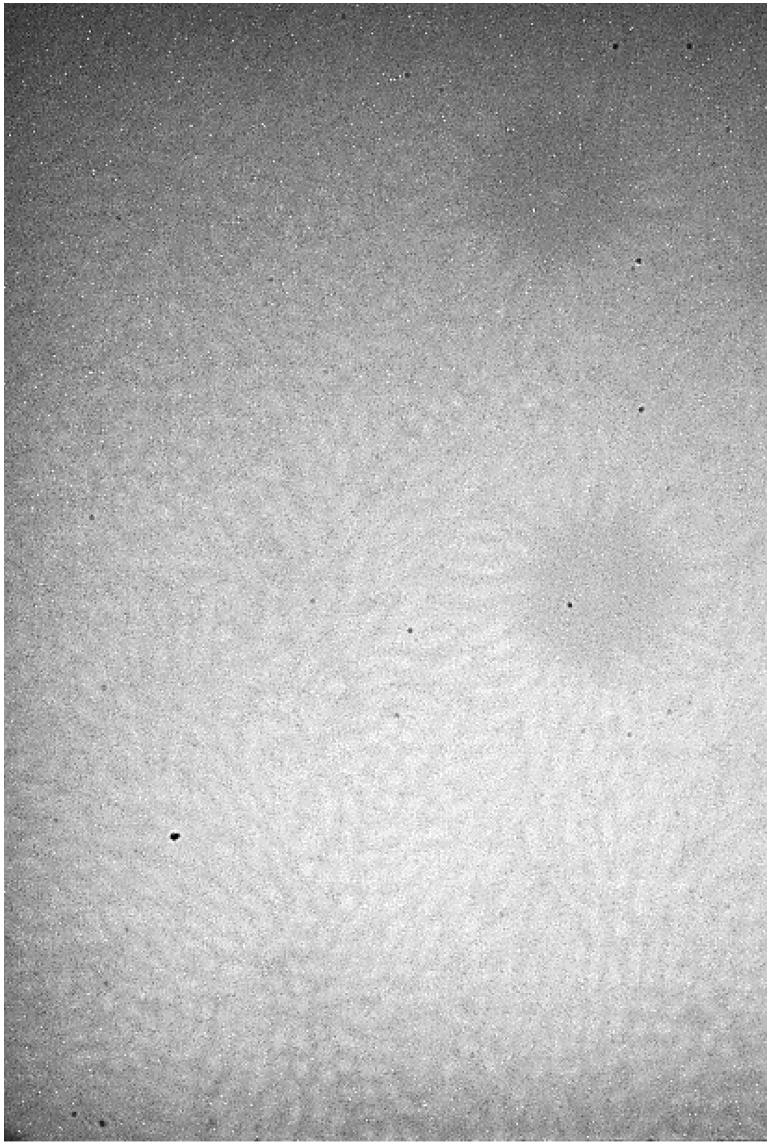
A diagram of Delaunay triangulation corresponding to the center spheres are developed. The maximum difference among each edge of triangles (labeled as *MaxDiff*) and area of the triangles (labeled as *TriArea*) formed as a result of Delaunay triangulation are computed.

2020/11/22, 21:23:50.501, CnFcl, ACET11, S2017, C3, Y-20, Z0+120, X100, X-12186, P11, E1053922, 00010

ELLIPSOIDS

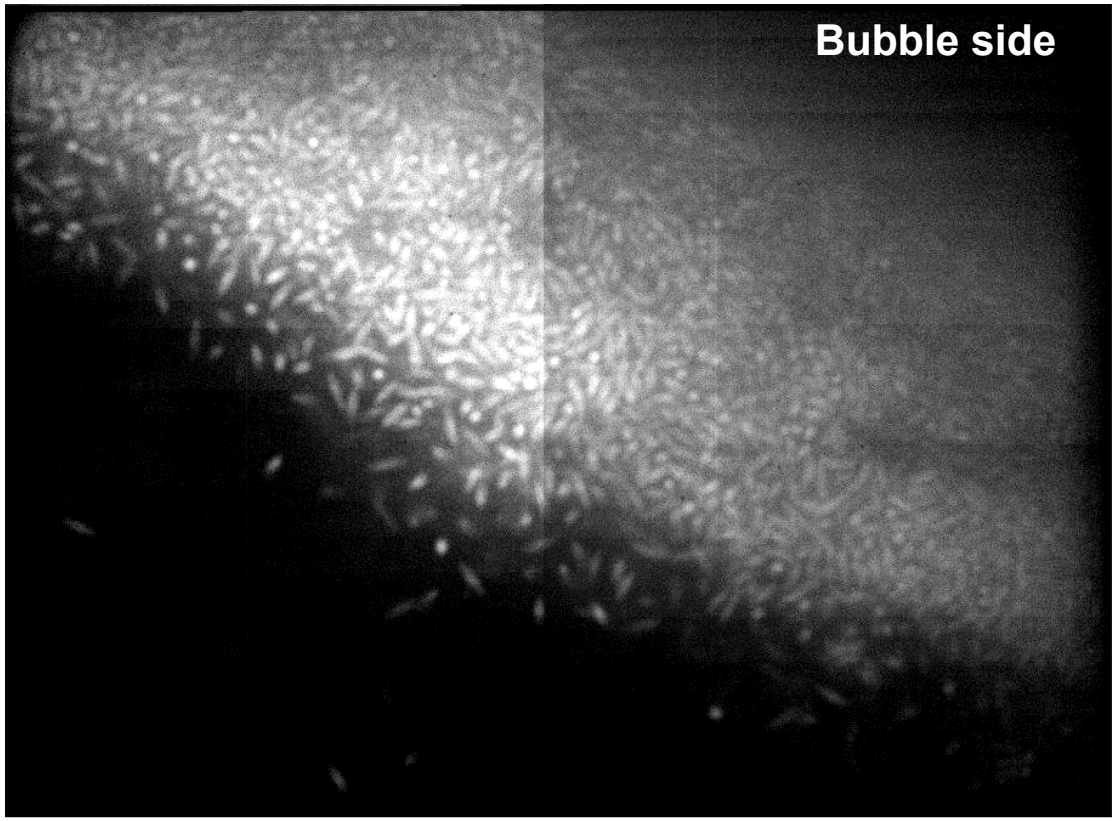
Nematic ordering and associated “hedgehog” defects

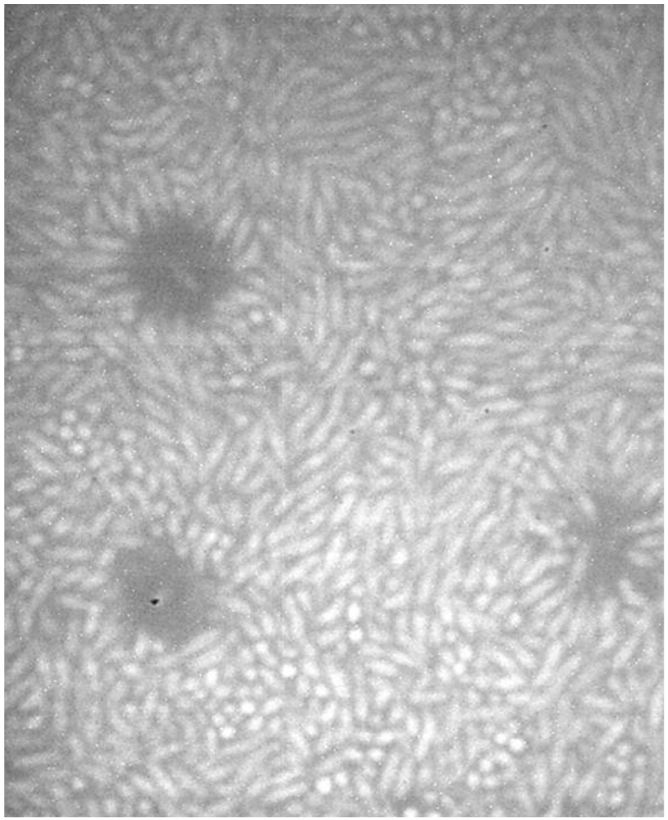




ELLIPSOIDS

Marangoni flow concentrated particles near bubble





SUMMARY OF PRELIMINARY DATA

Spheres

- Large crystal formed in microgravity
- Observed transition between crystalline and non-crystalline structures

Ellipsoids

- Large nematic regions formed in microgravity
- Associated “hedgehog” defects found

Questions

Boris Khusid: khusid@njit.edu