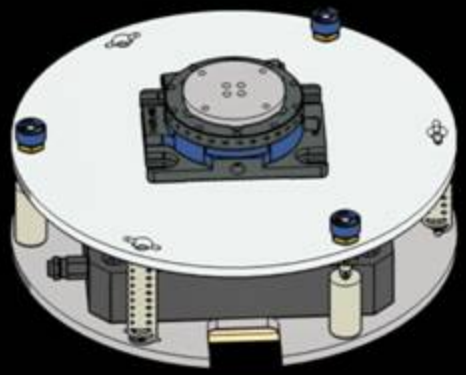
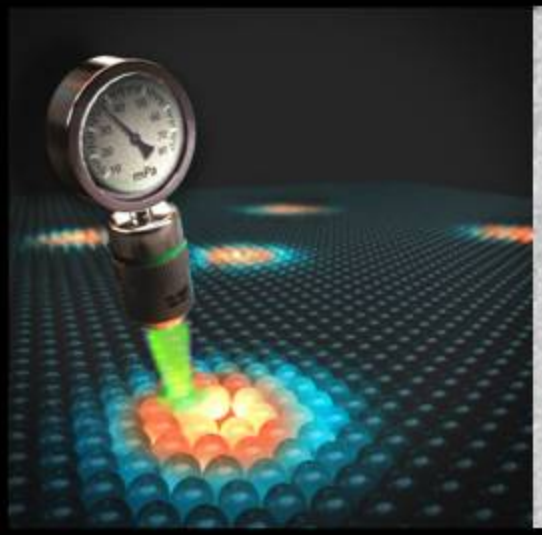
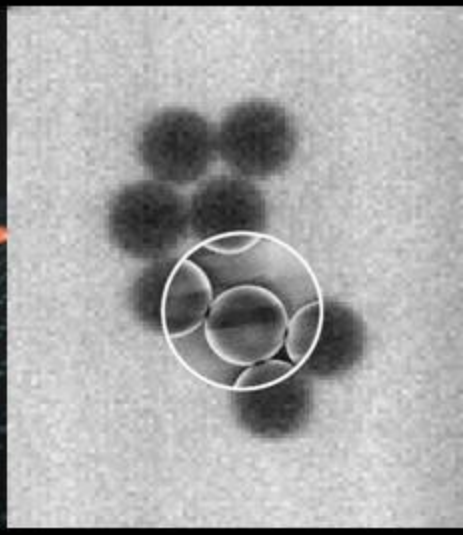
Quantitative Light Microscopy of Dense Suspensions

Colloid Science at the Next Decimal Place



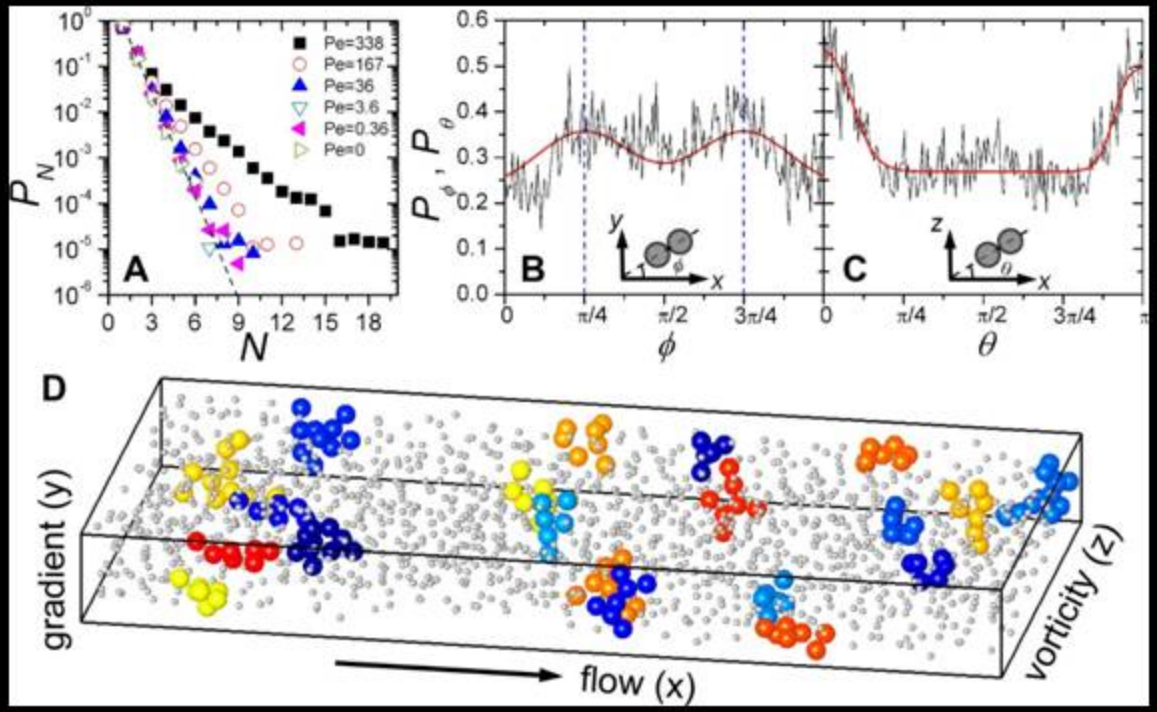




Itai Cohen

Brian Leahy, Matt Bierbaum, Neil Lin, Xiang Cheng, Jonathan McCoy, Jim Sethna, Jacob Israelachvili, Ben Guy, Michiel Hermes, Wilson Poon, Chris Ness, Mike Cates, Jin Sun

Shear thickening



Continuous shear thickening due to clusters

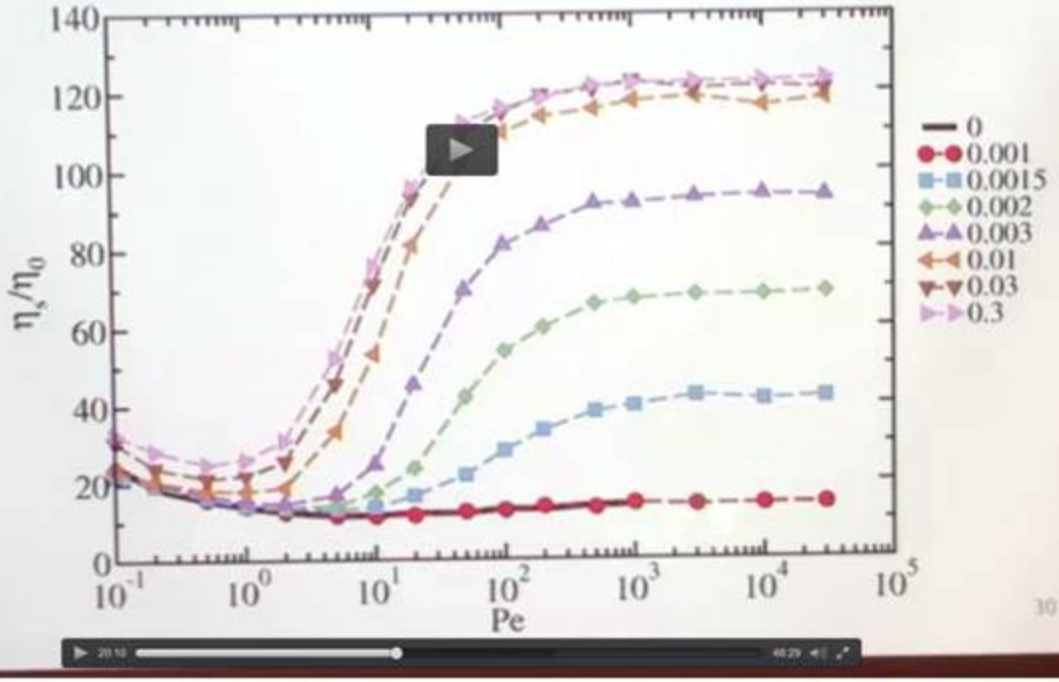
X Cheng, J McCoy, J Israelachvili, I Cohen – *Science*, 2011

John's talk...

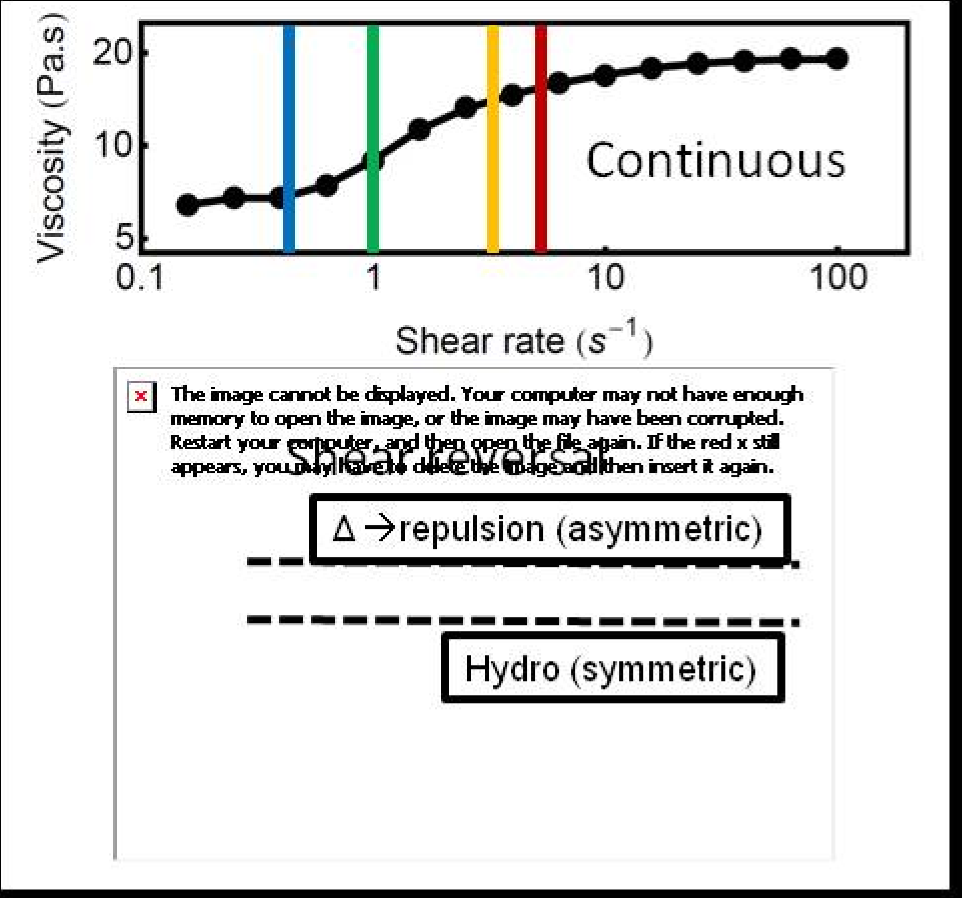
Bidisperse suspensions size ratio 1.4:1, volume ratio 1:1;

$N = 30, \alpha = 0.01, \phi = 0.5$

$$f(h) = \frac{\alpha}{h} + \frac{2\alpha}{h_0^3}h^2 - \frac{3\alpha}{h_0^2}h$$

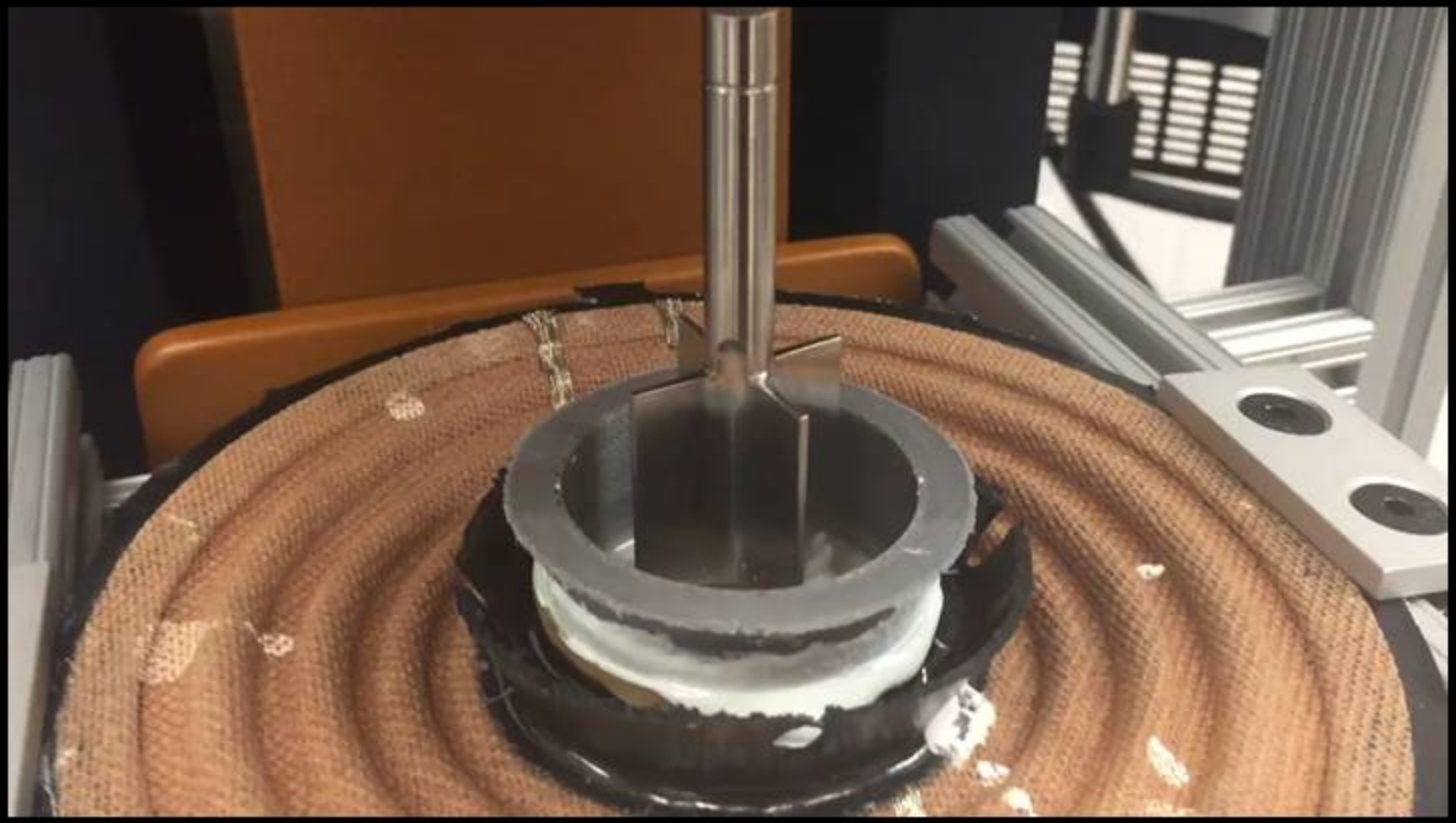


Flow reversal (silica): repulsion!



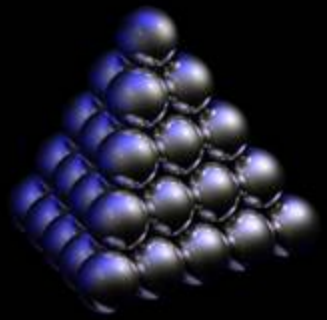
YC Lin, B Guy, M Hermes, W Poon, C Ness, J Sun, I Cohen – *PRL*, 2015

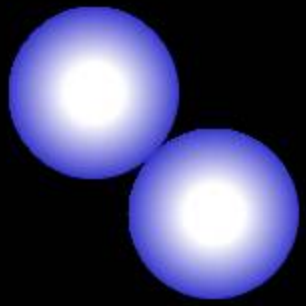
Tunable shear thickening

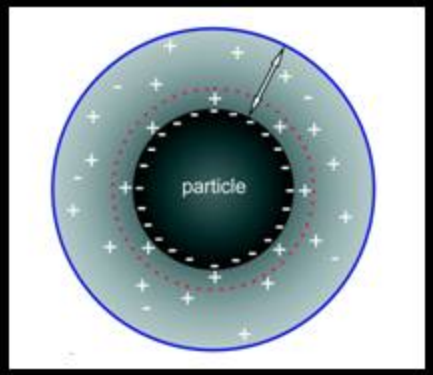


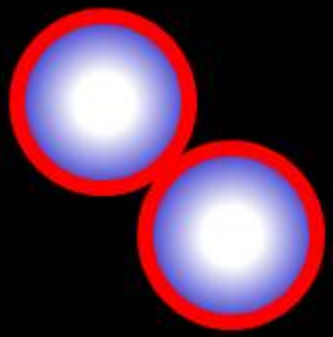
YC Lin, Chris Ness, M Cates, J Sun, I Cohen – *PNAS*, 2016

Repulsion, contact, friction & all that…









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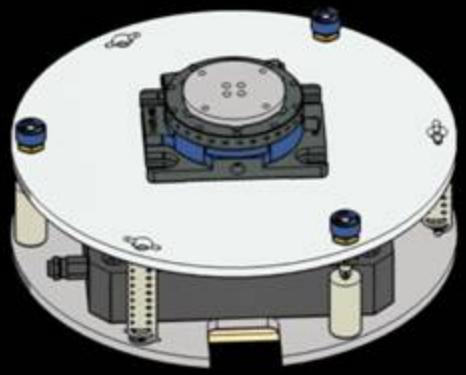
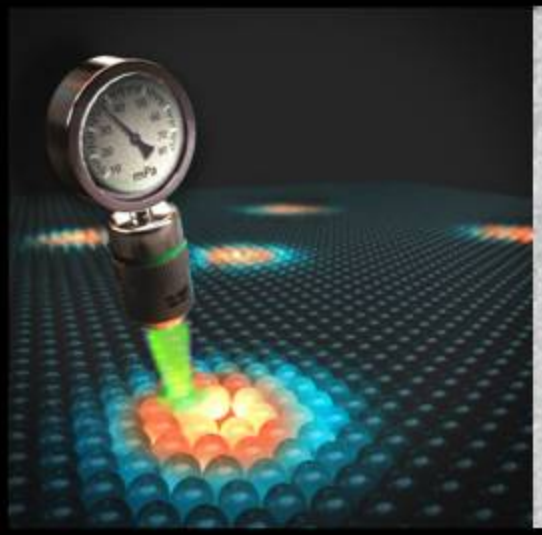
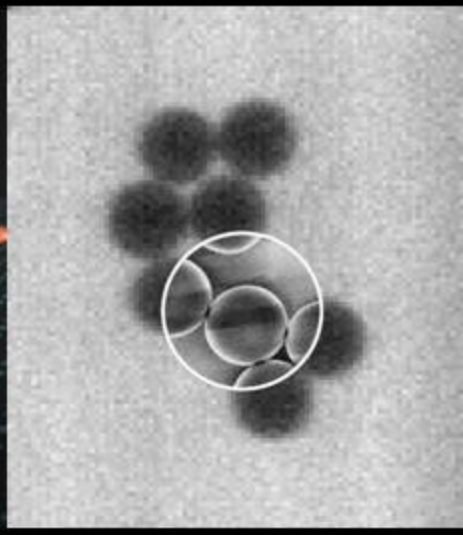






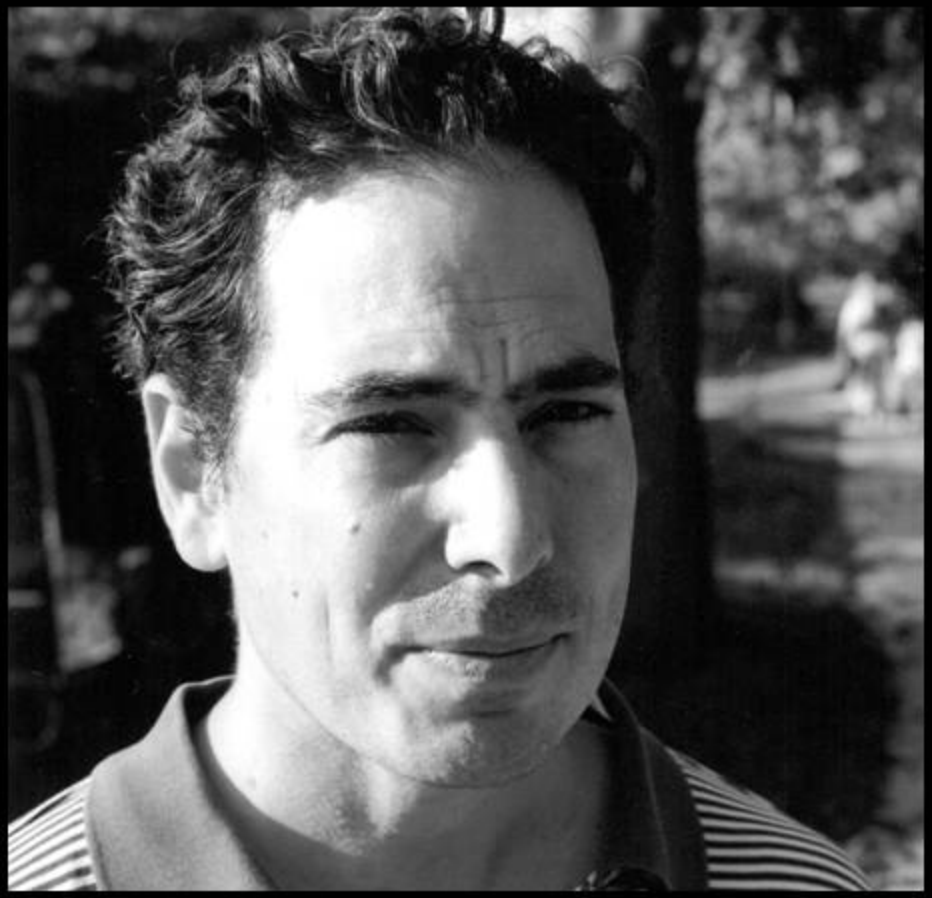
Itai Cohen

Brian Leahy, Matt Bierbaum, Neil Lin, Xiang Cheng, Jonathan McCoy, Jim Sethna, Jacob Israelachvili, Ben Guy, Michiel Hermes, Wilson Poon, Chris Ness, Mike Cates, Jin Sun

"Why should one wish to make measurements with ever increasing precision?

Because the whole history of physics proves that a new discovery is likely to be found lurking in the next decimal place."

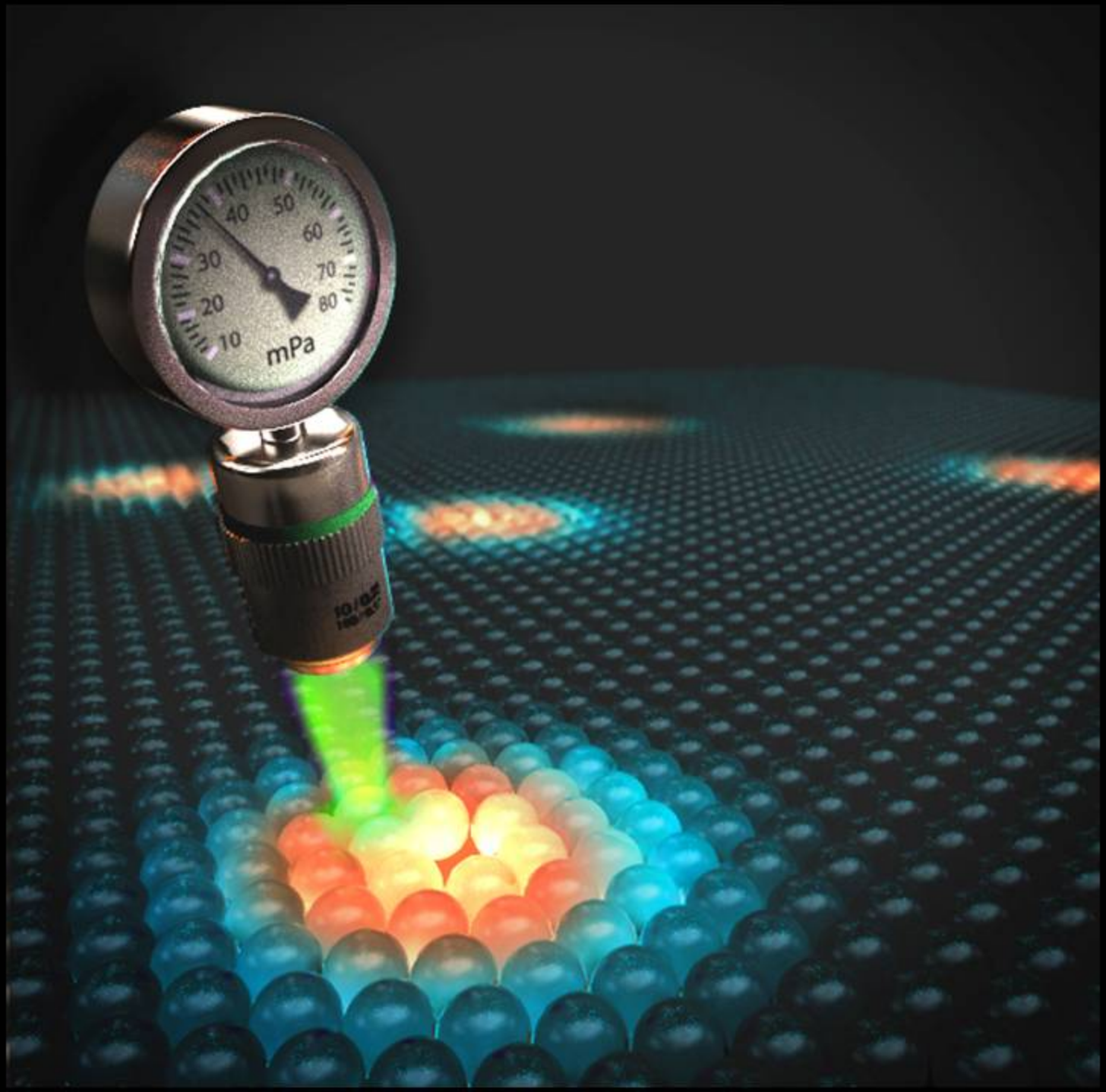
(Floyd K. Richtmyer, 1931)



"Why should one wish to make measurements with ever increasing precision?

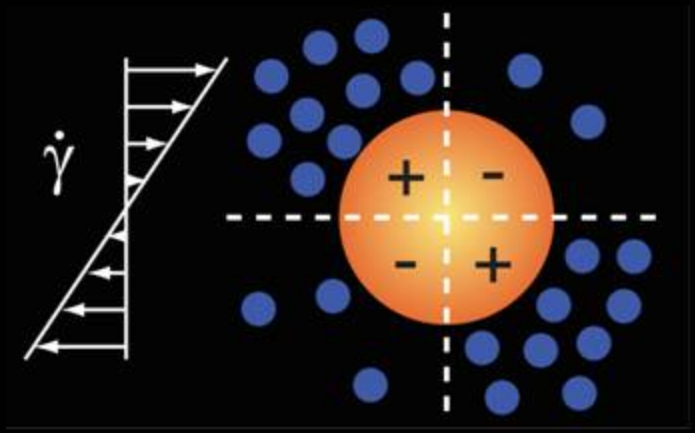
Because everyone has solved everything so I'm going to need to work harder."

(Itai Cohen, 2017)


10
20
30
40
50
60
70
80
mPa

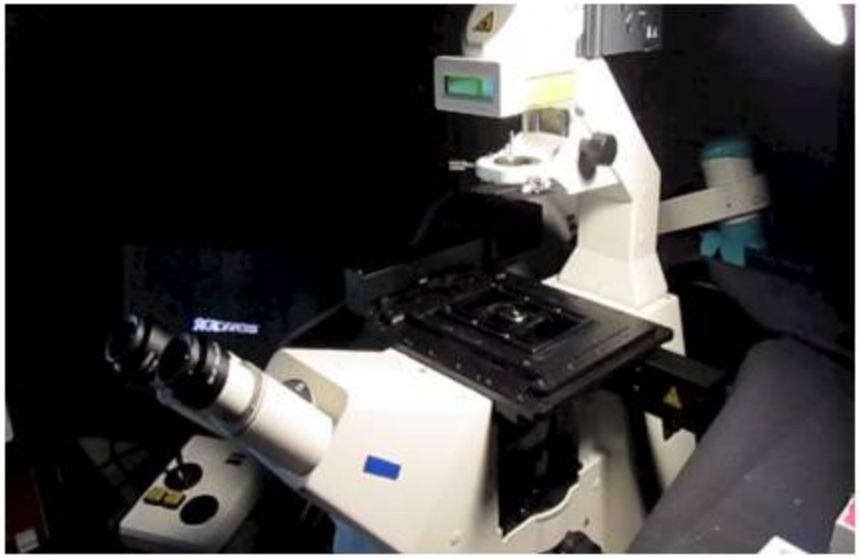
Brownian stress

$$\sigma = -n^2 k_B T a \iint_{r=2a} \hat{r}\hat{r} g(\vec{r}) dS$$



JG Kirkwood, J Chem Phys (1950)
JF Brady, J Chem Phys (1993)
DR Foss & JF Brady, JFM (2000)
C Gao etal, PRE (2010)
X Cheng etal, Science (2011)

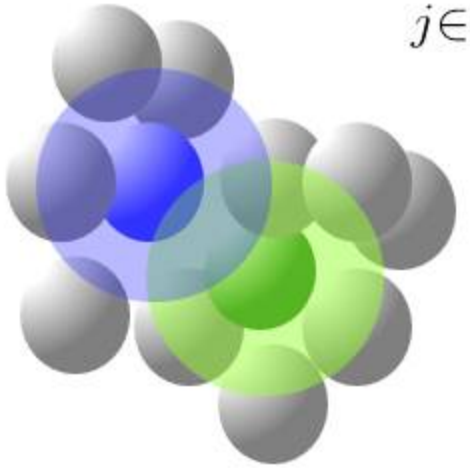
Stress Assessment from Local Structural Anisotropy (SALSA)



1μm silica in water/glycerol

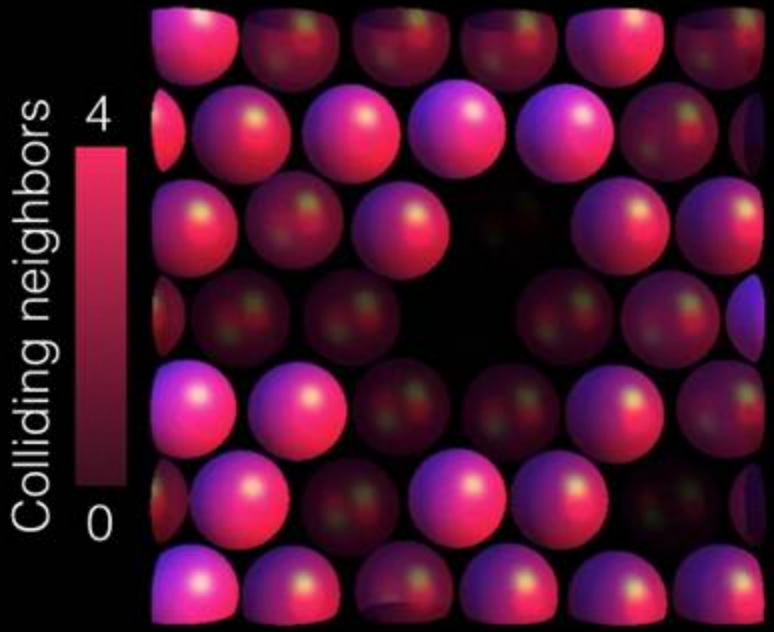
$$\bar{\bar{\sigma}}_i^B = -\frac{k_B T}{\Omega_i}\frac{a}{\Delta}\psi_i(\Delta)$$

$$\bar{\bar{\psi}}_i = \sum_{j\in\Delta} \vec{r}_{ij}\vec{r}_{ij}$$

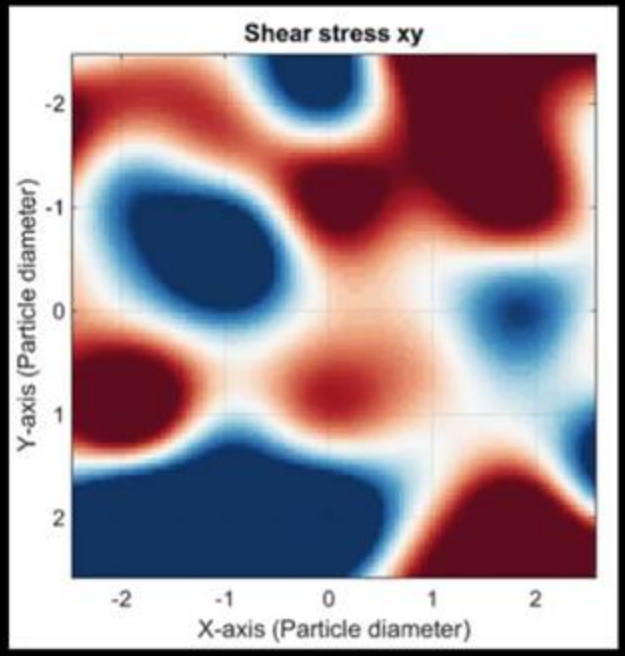


Neil Lin, Matt Bierbaum,
James Sethna, Peter Schall

Step 2
Average stress snapshots over time



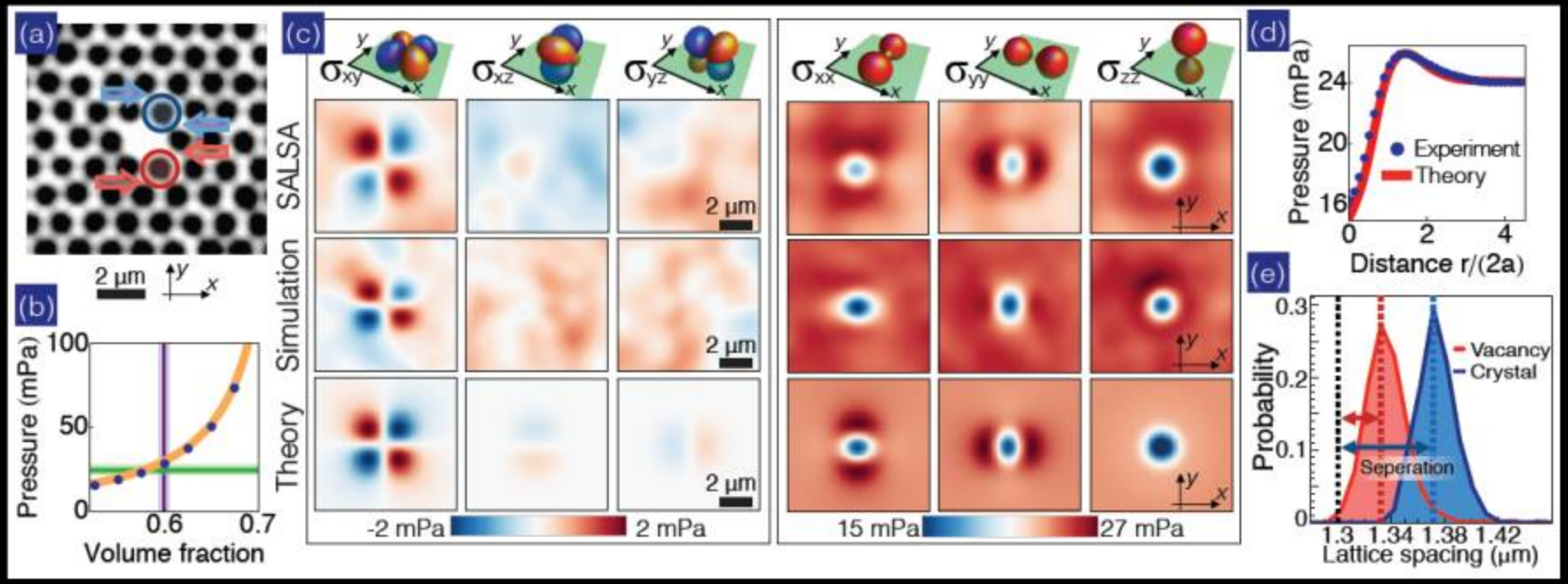
snapshot



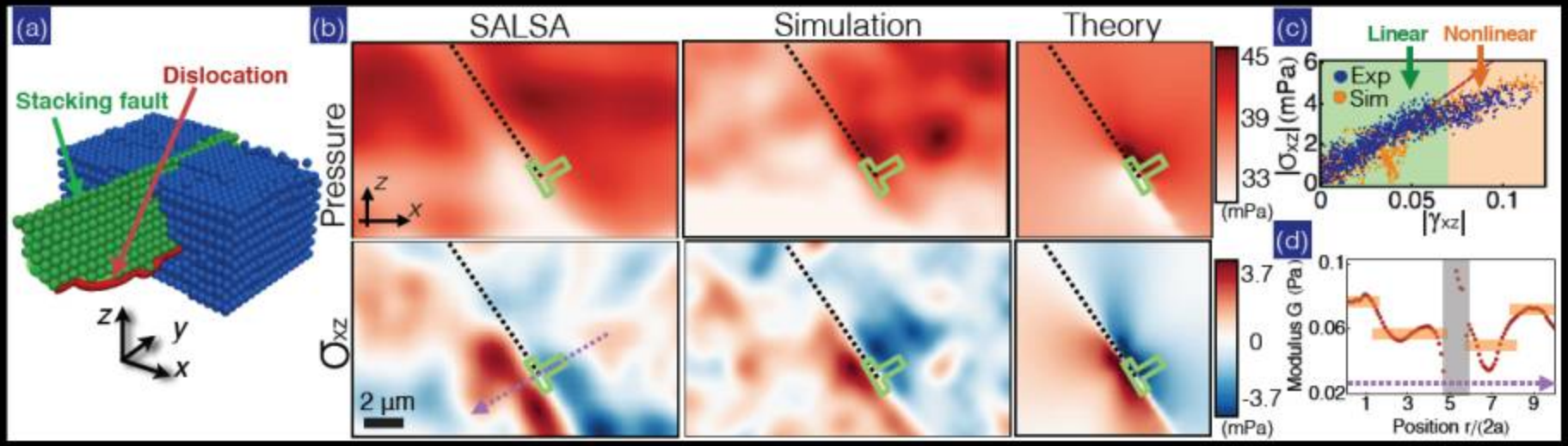
snapshot

Apply to crystals, liquids and glasses

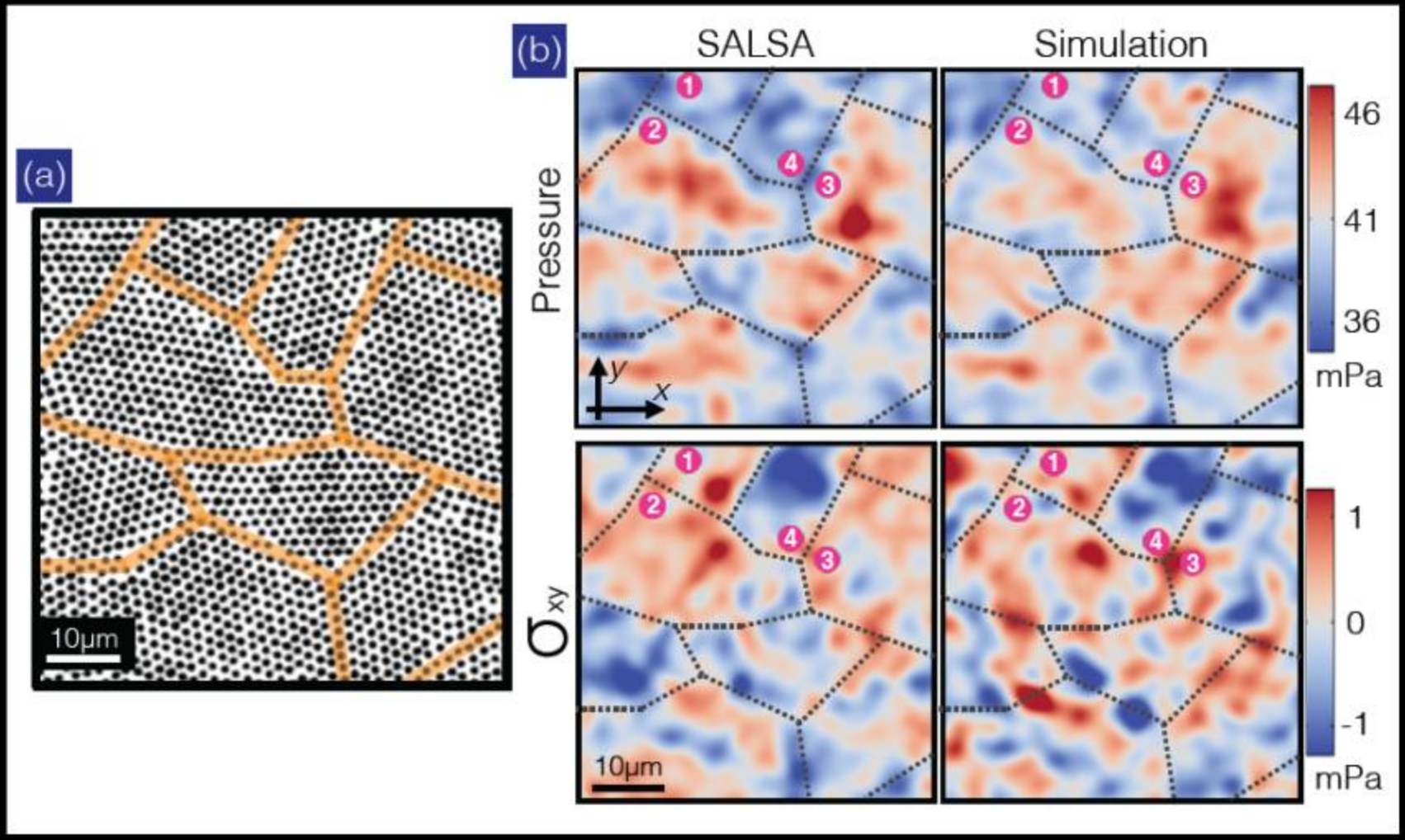
Vacancy


(a)
2 μm
y
x
(b)
Pressure (mPa)
100
50
0
0.6
0.7
Volume fraction
(c)
σxy
σxz
σyz
σxx
σyy
σzz
SALSA
Simulation
Theory
2 μm
-2 mPa
2 mPa
15 mPa
27 mPa
(d)
Pressure (mPa)
24
20
16
0
2
4
Experiment
Theory
Distance r/(2a)
(e)
Probability
0.3
0.2
0.1
0
Vacancy
Crystal
Seperation
1.3
1.34
1.38
1.42
Lattice spacing (μm)

Dislocation

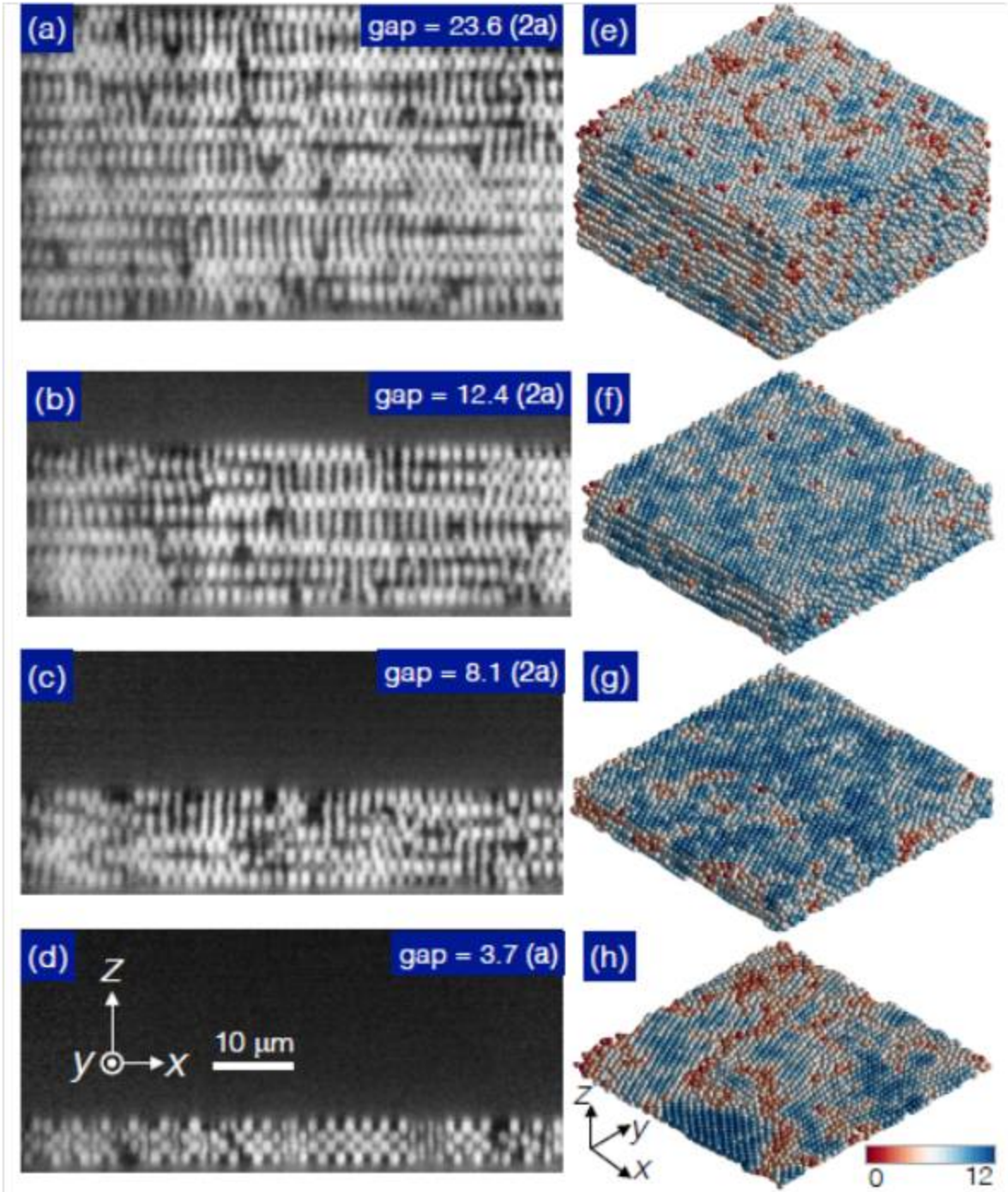


Polycrystal



N Lin*, M Bierbaum*, J Sethna, I Cohen – Nature Materials, 2016

- Confinement
- Yield
- Plasticity
- Defect interaction
- Cracking
- …



N Lin, I Cohen –
Soft Matter, 2016

Liquids: "The noise is the signal"

Rolf Landauer

Green-Kubo relation

Volume

Viscosity

$$\eta = \frac{V}{k_B T} \int_0^\infty \langle \sigma_{xy}(t_0)\sigma_{xy}(t_0 + \Delta t)\rangle d\Delta t,$$

Thermal energy

Stress-stress correlation (Δt)

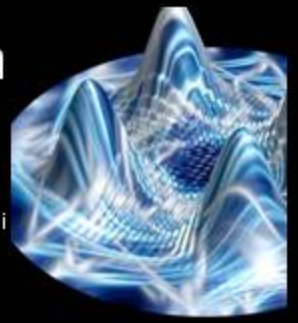
Simple liquid

W. G. Hoover, D. J. Evans, R. B. Hickman, A. J. C. Ladd, W. T. Ashurst, & B. Moran, Phys. Rev. A 22, 1690 (1980)

Supercooled liquid

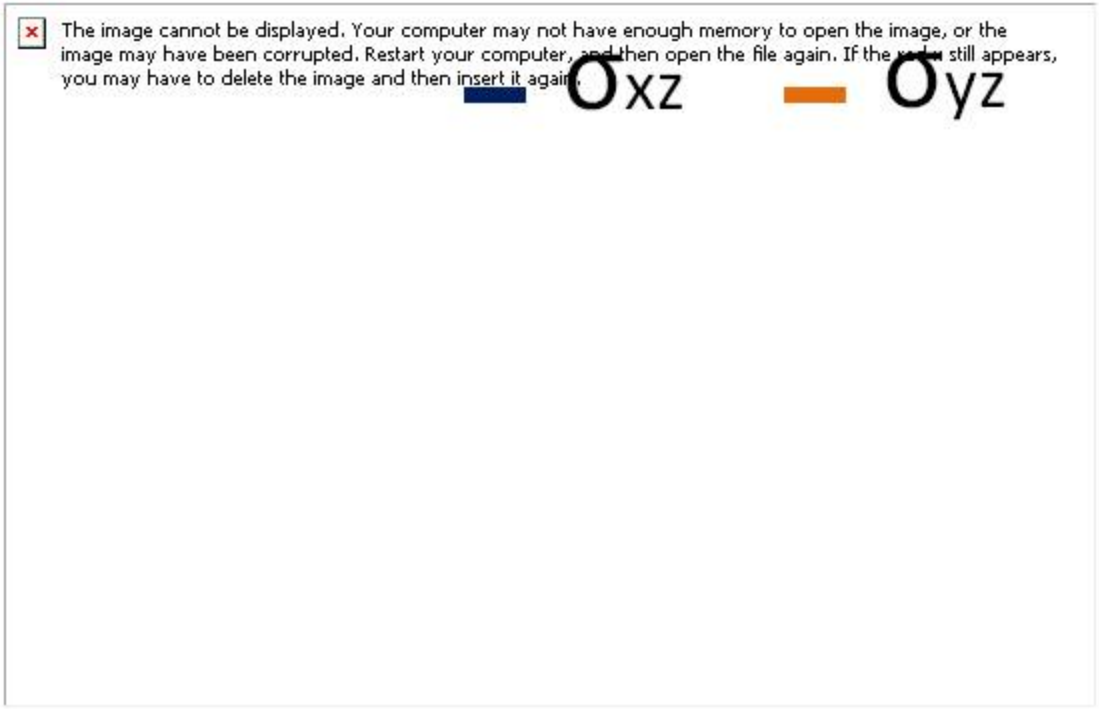
Akihiro Kushima, et al. J. Chem. Phys. 130, 224504 (2009)

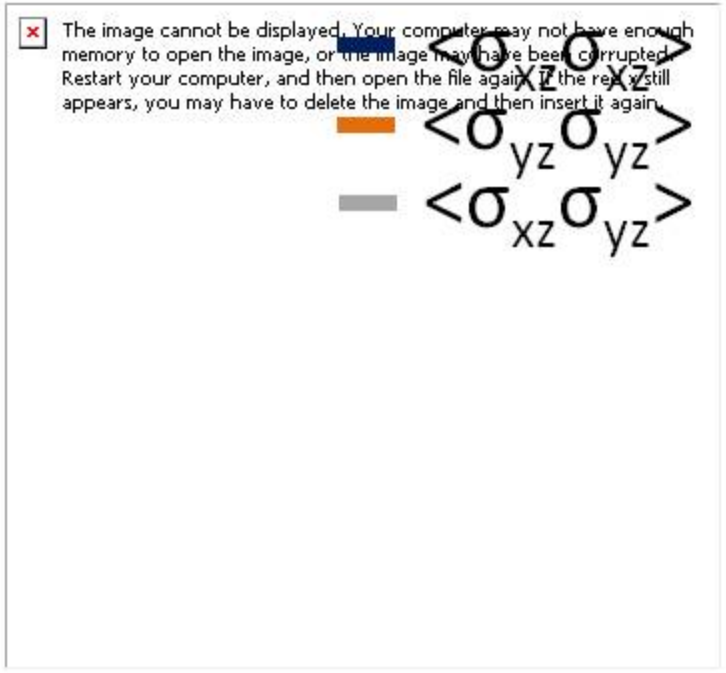
Quantum liquid



D. R. Reichman & E. Rabani *PRL* 87, 265702 (2001)

“The noise is the signal”





Directly image the Brownian stress!

The image cannot be displayed. Your computer may not have enough memory to open the image, or the image may have been corrupted. Restart your computer, and then open the file again. If the red x still appears, you may have to delete the image and then insert it again.

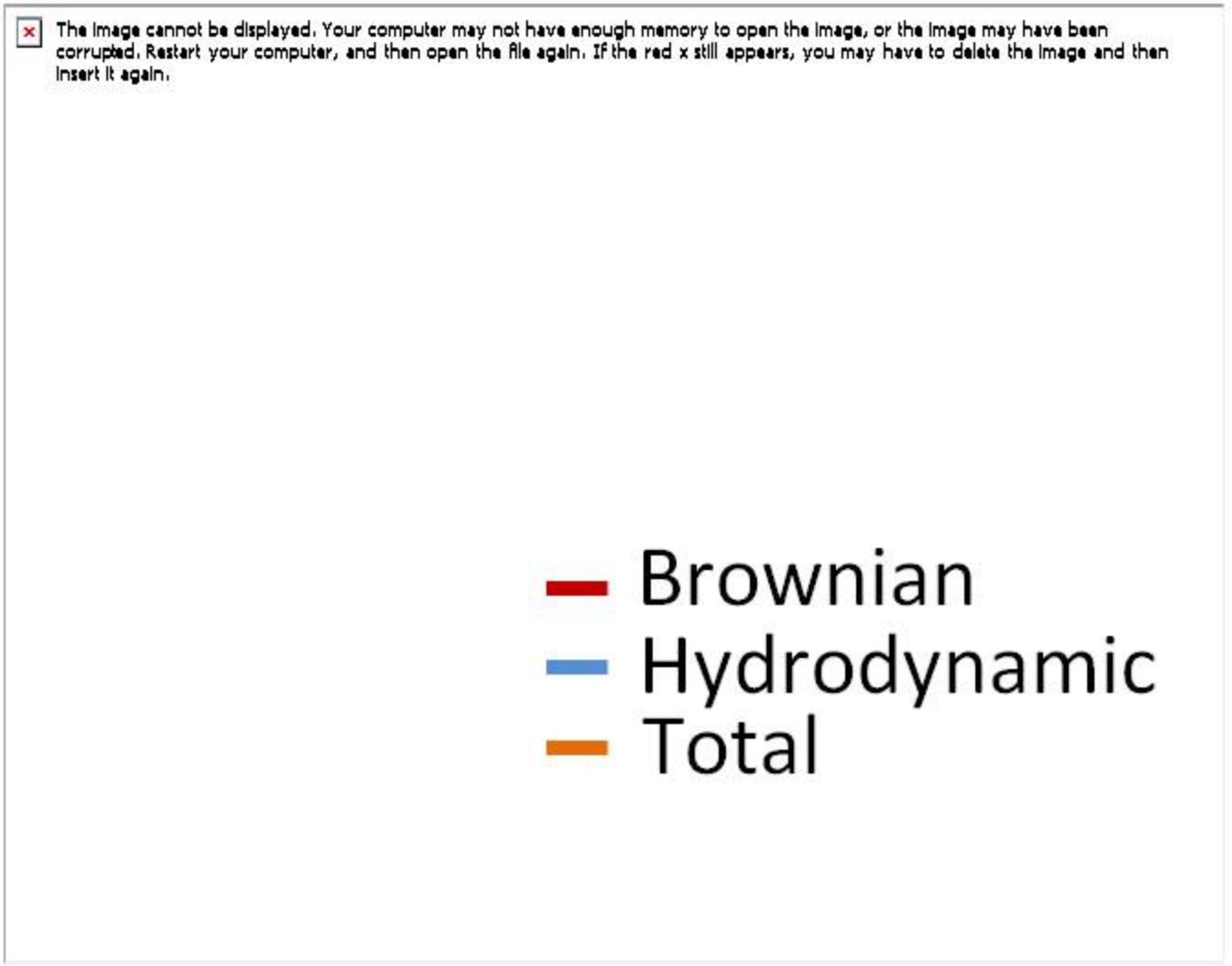
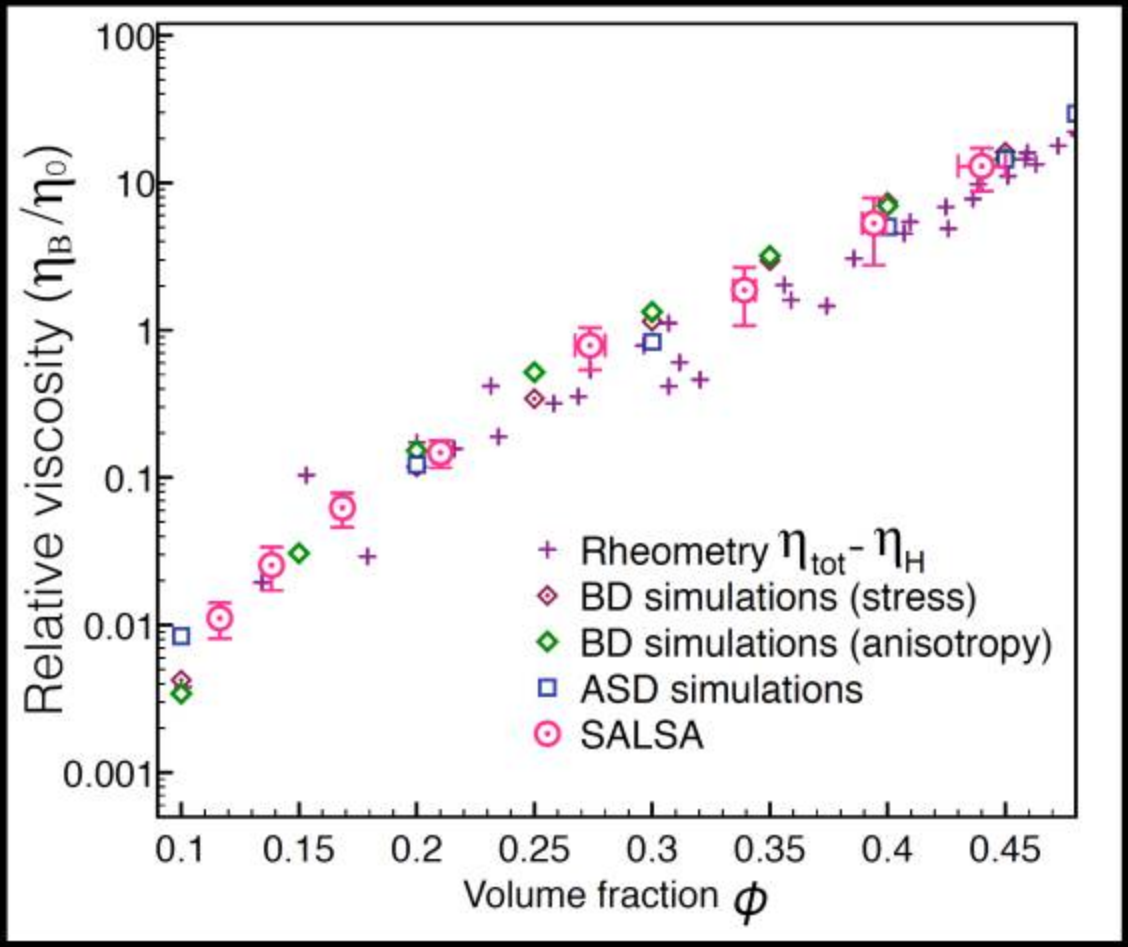


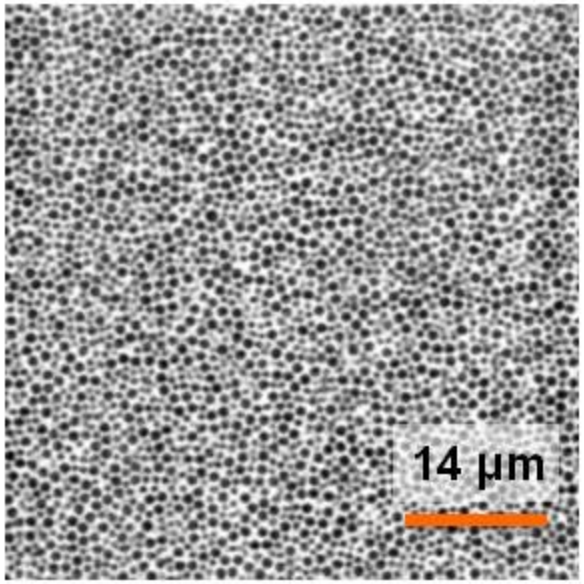
Image the Brownian stress!

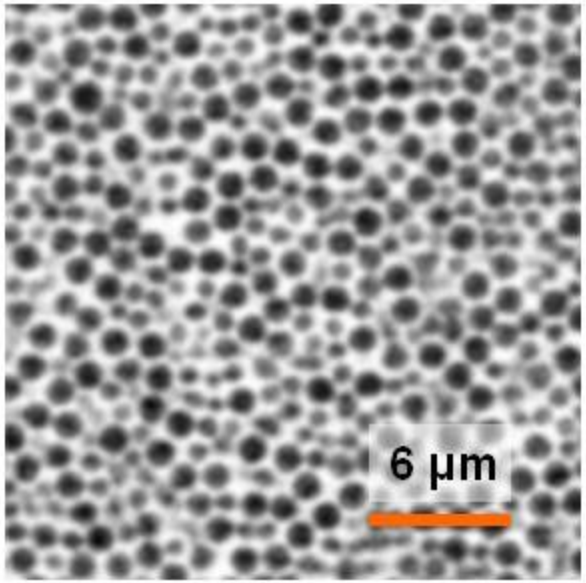


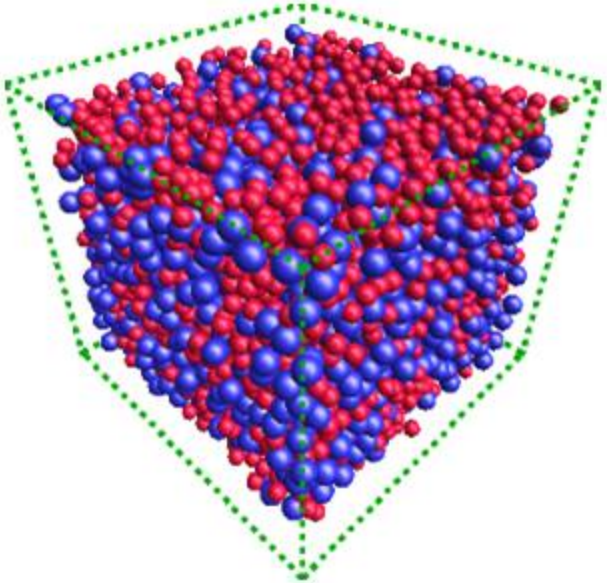
N Lin, M Bierbaum, I Cohen – PRL, 2017

What about high densities?

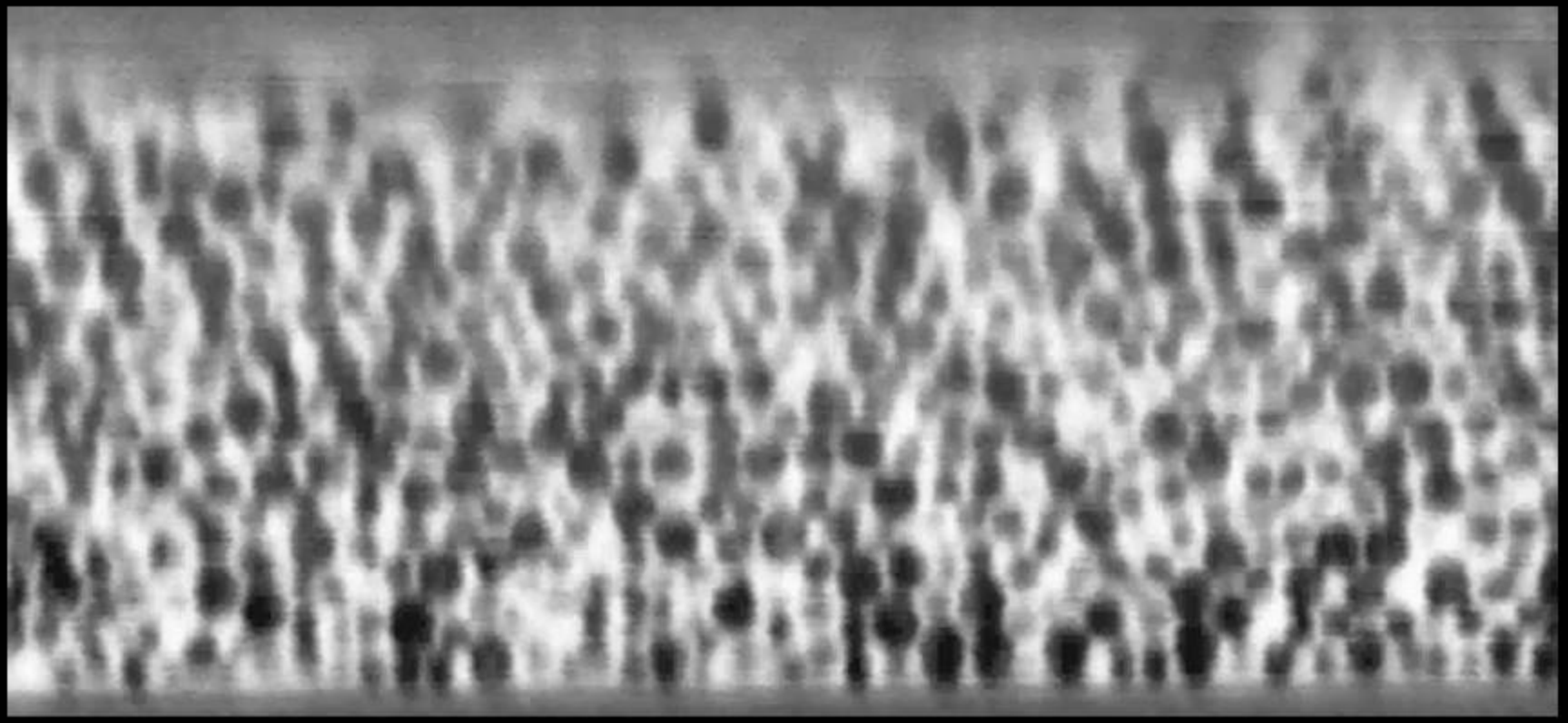
System no longer in equilibrium
Local stresses vary
SALSA still applies







What about high densities?



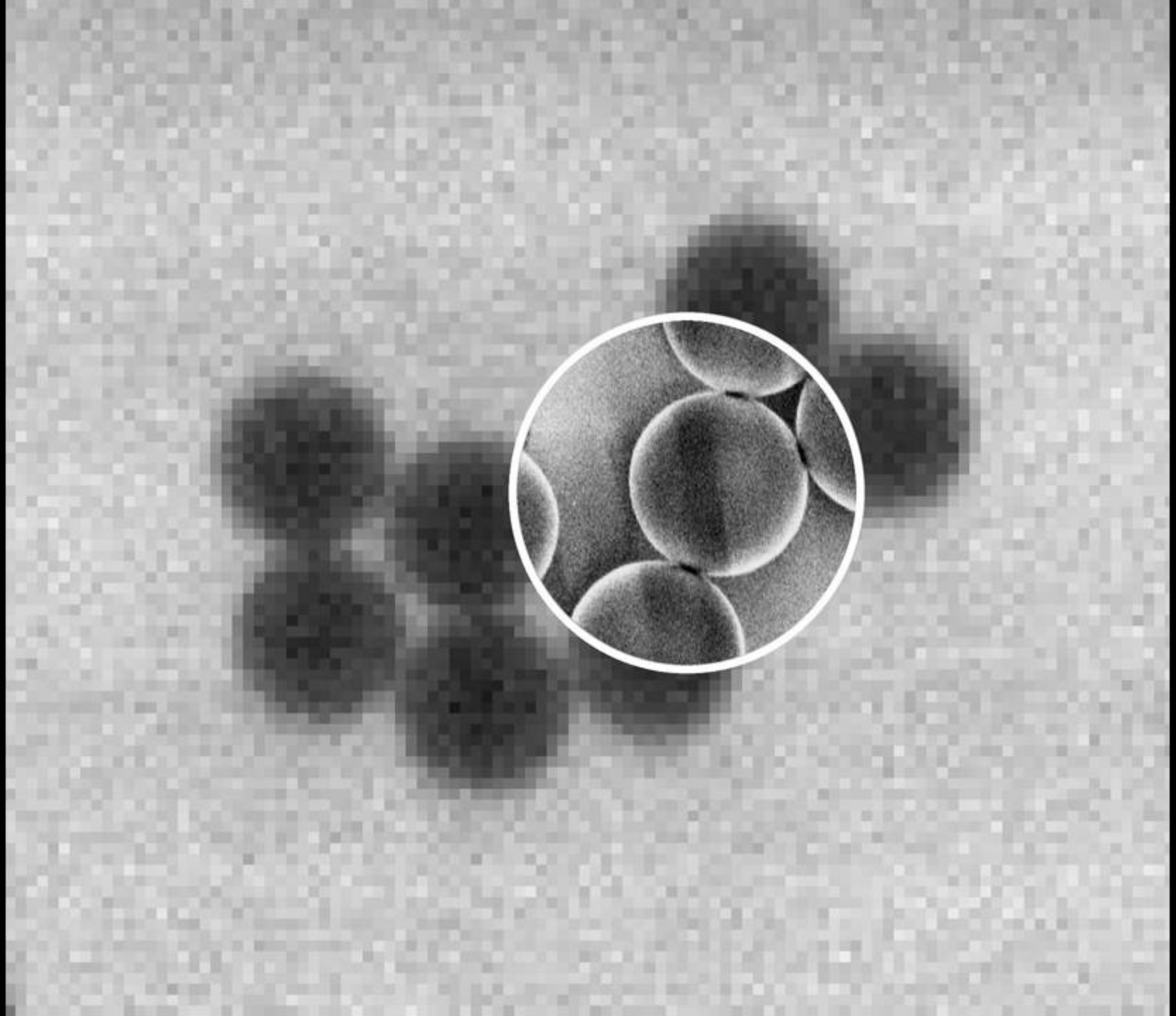
But… accurately locating all particles is a problem

Current techniques: centroid location, σ_x ~ 100 nm,
no R measurement, missing particles

Extend force measurement to high Pe?

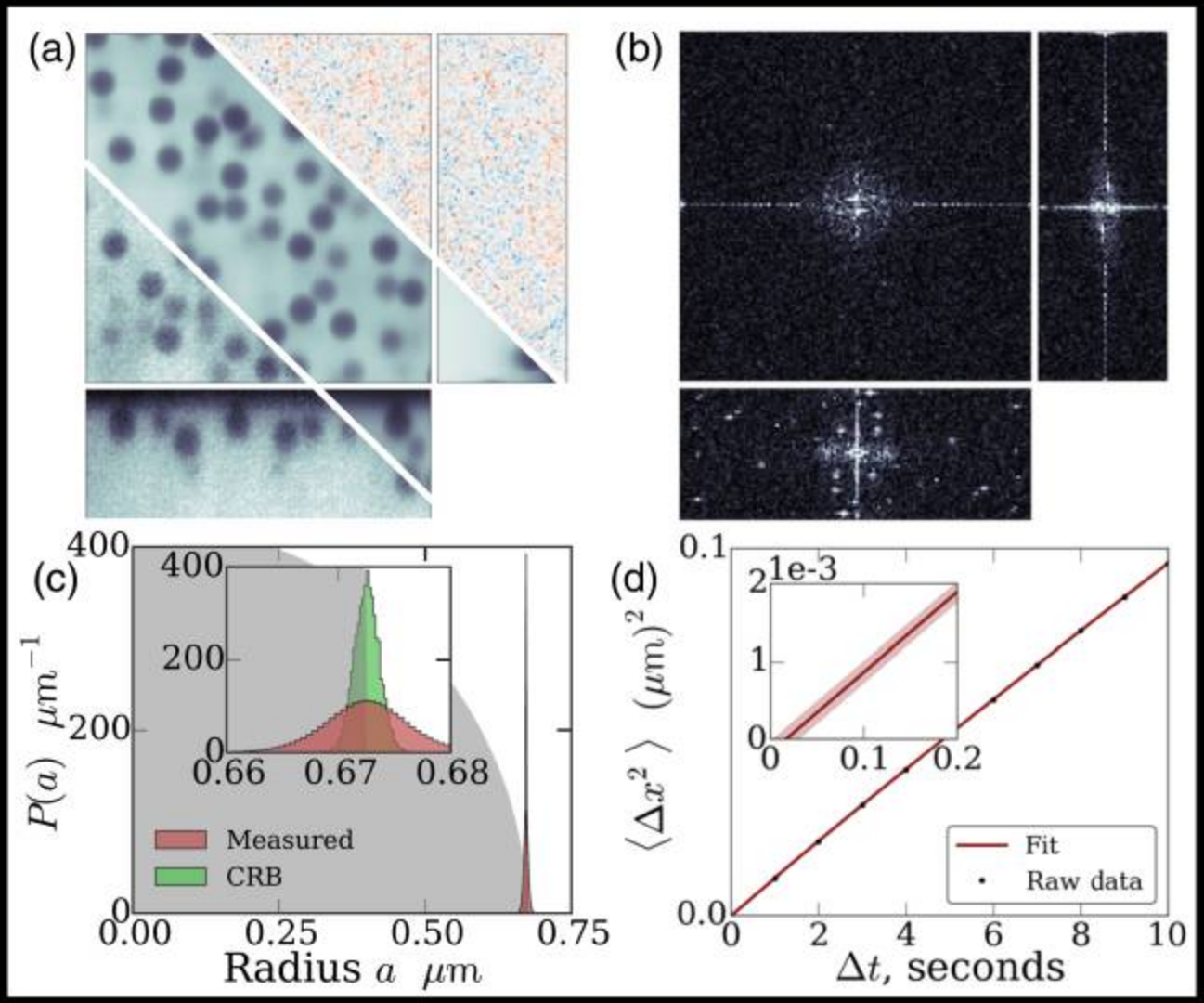
Robert Brown

Jean Claude Eugène Péclet



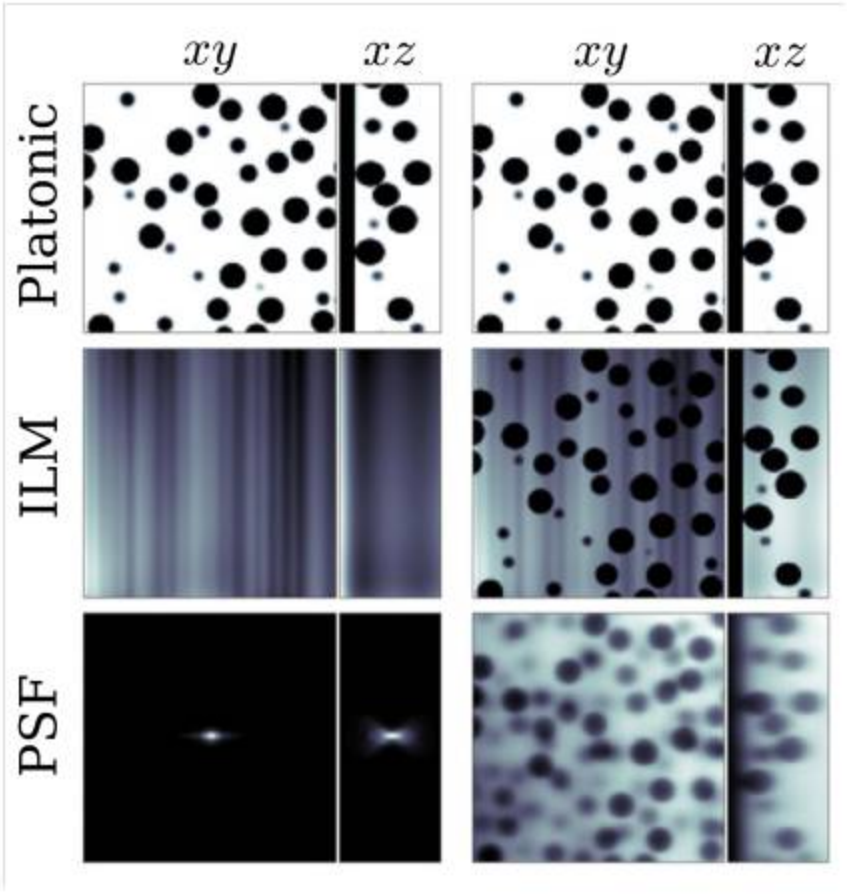
Images are complicated

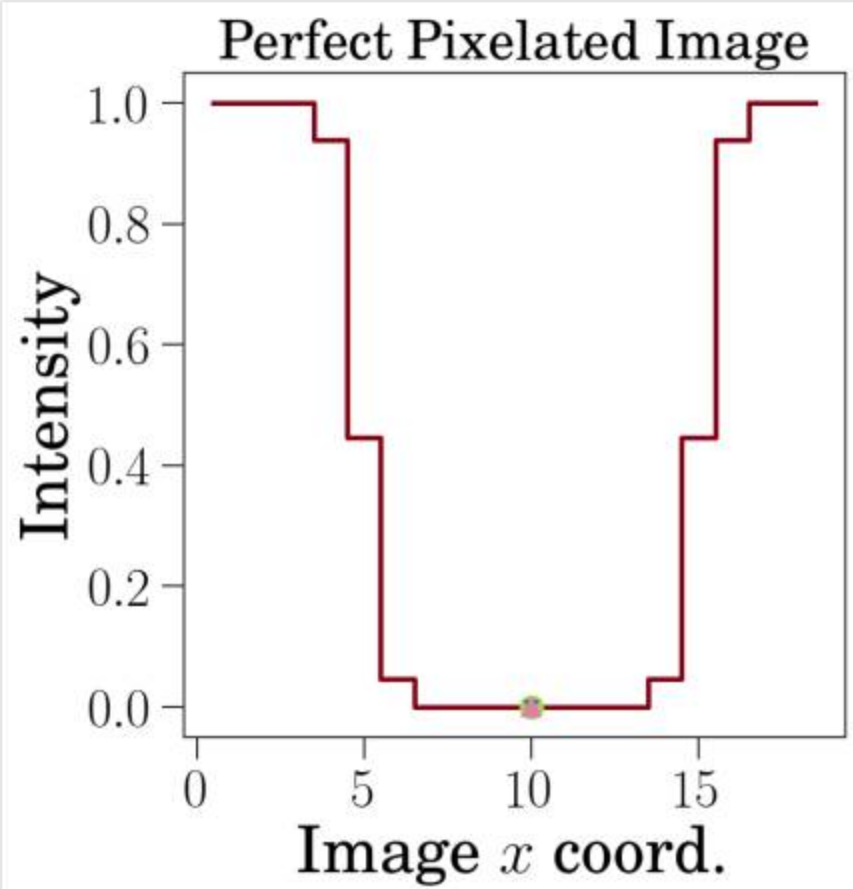
Can extract radii and volume fractions

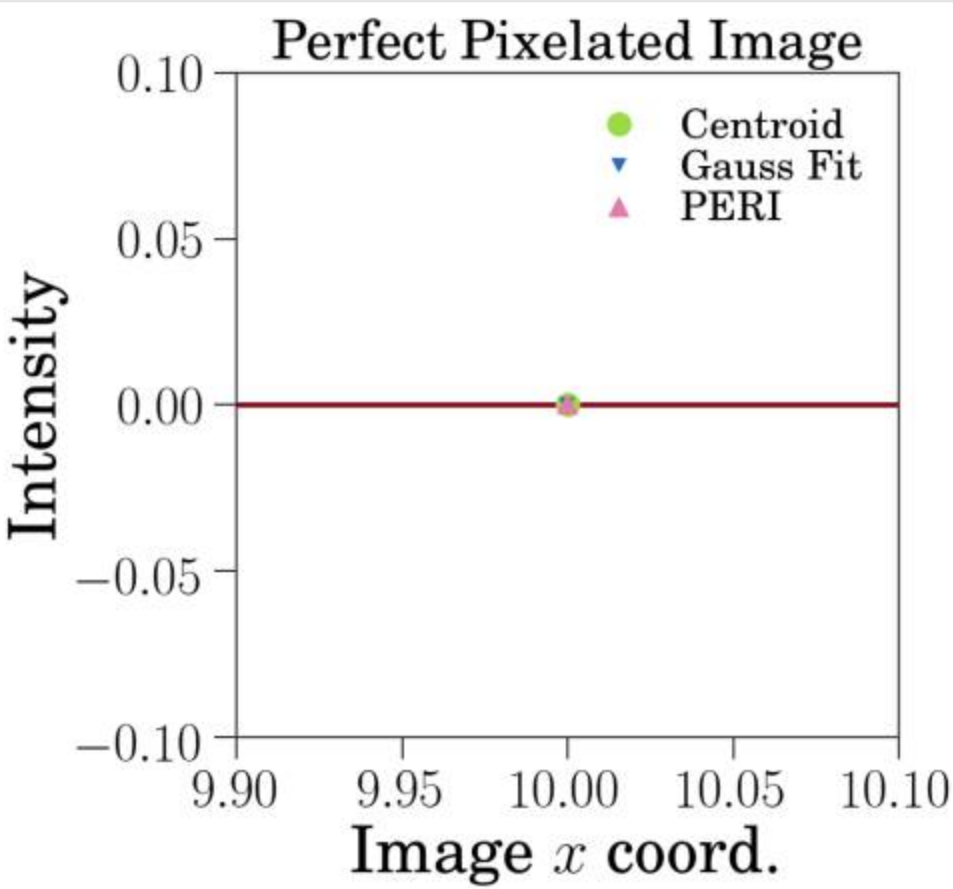


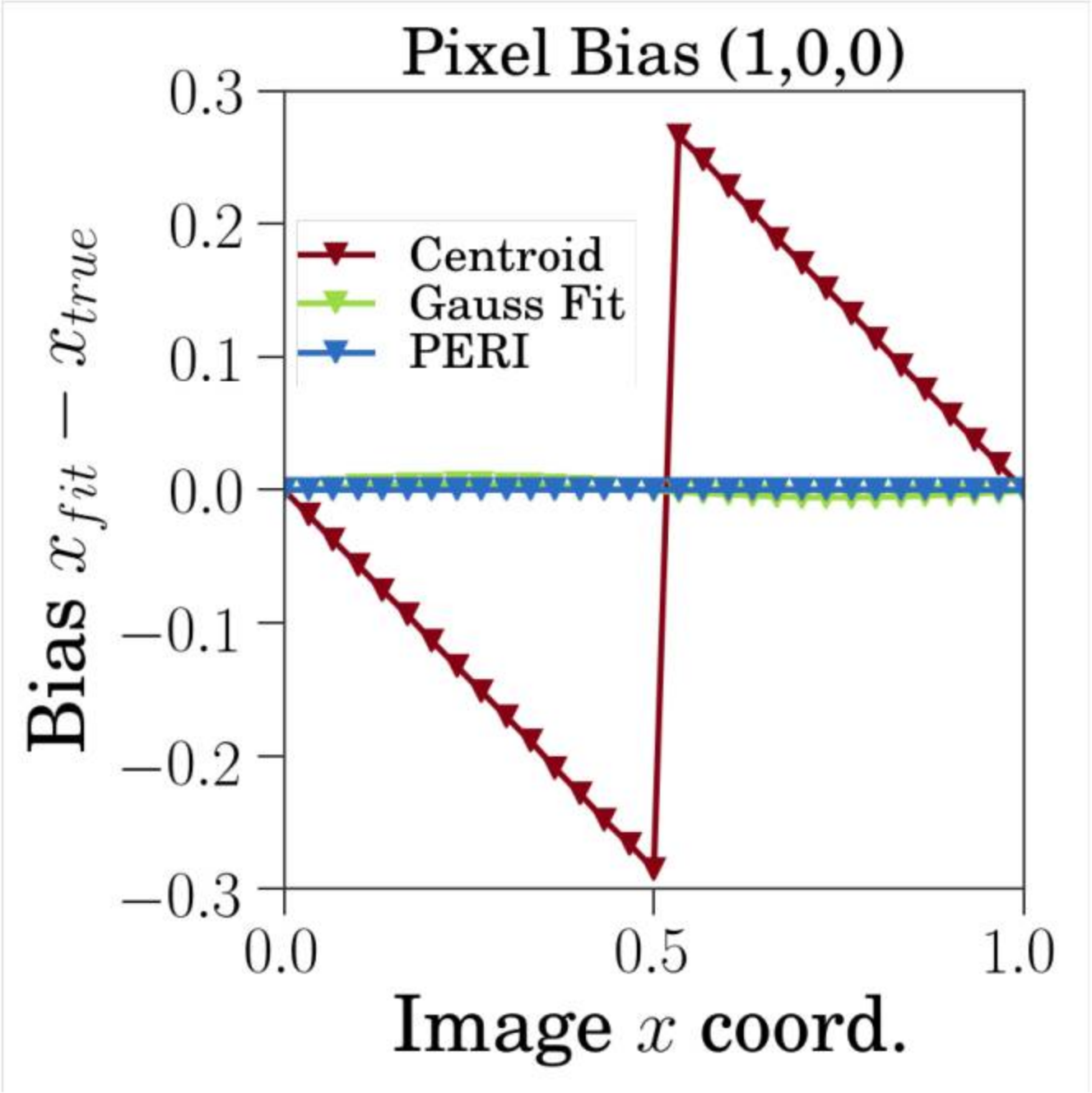
Aside: comparison to other methods

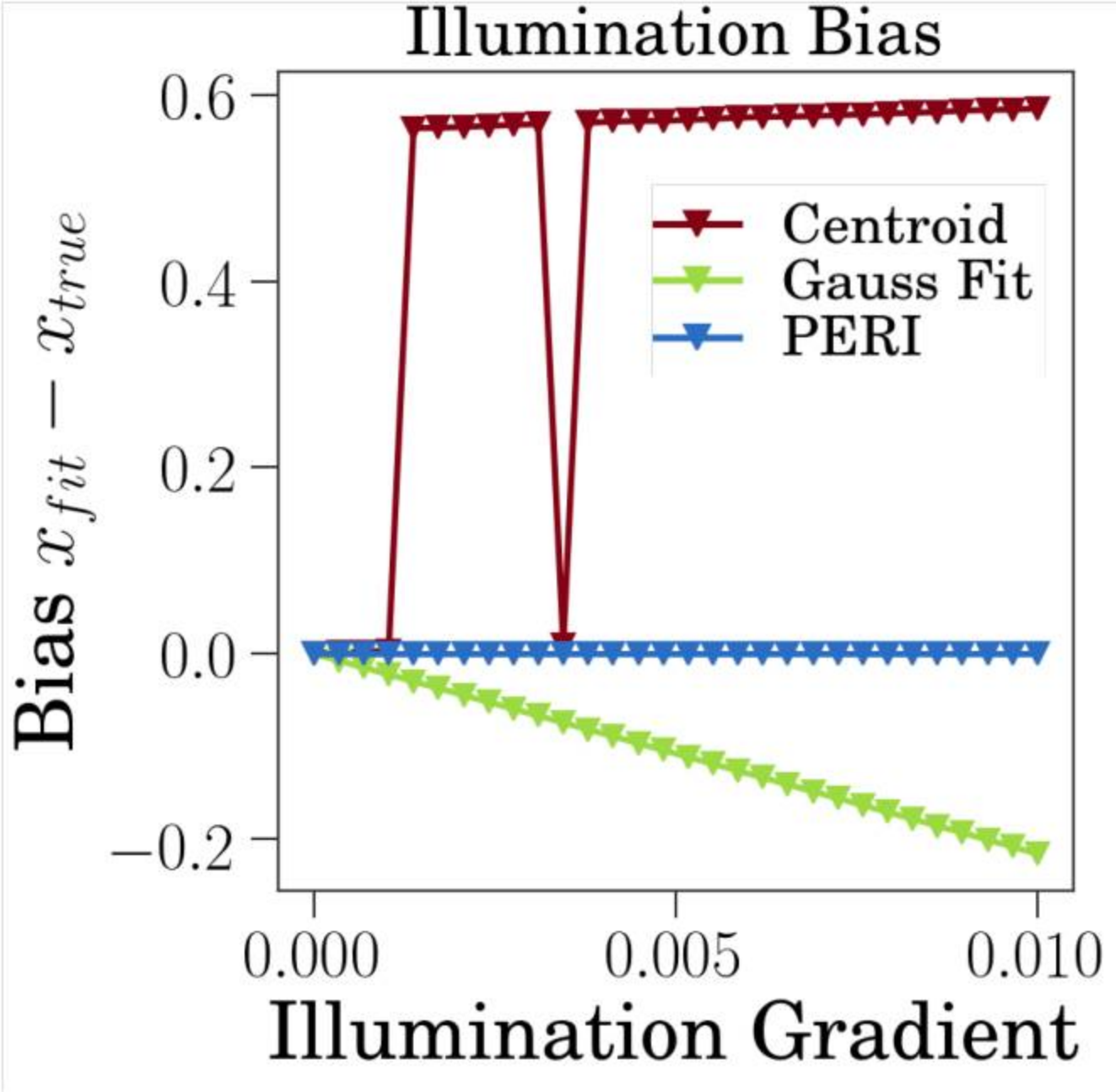
Build a fake image…

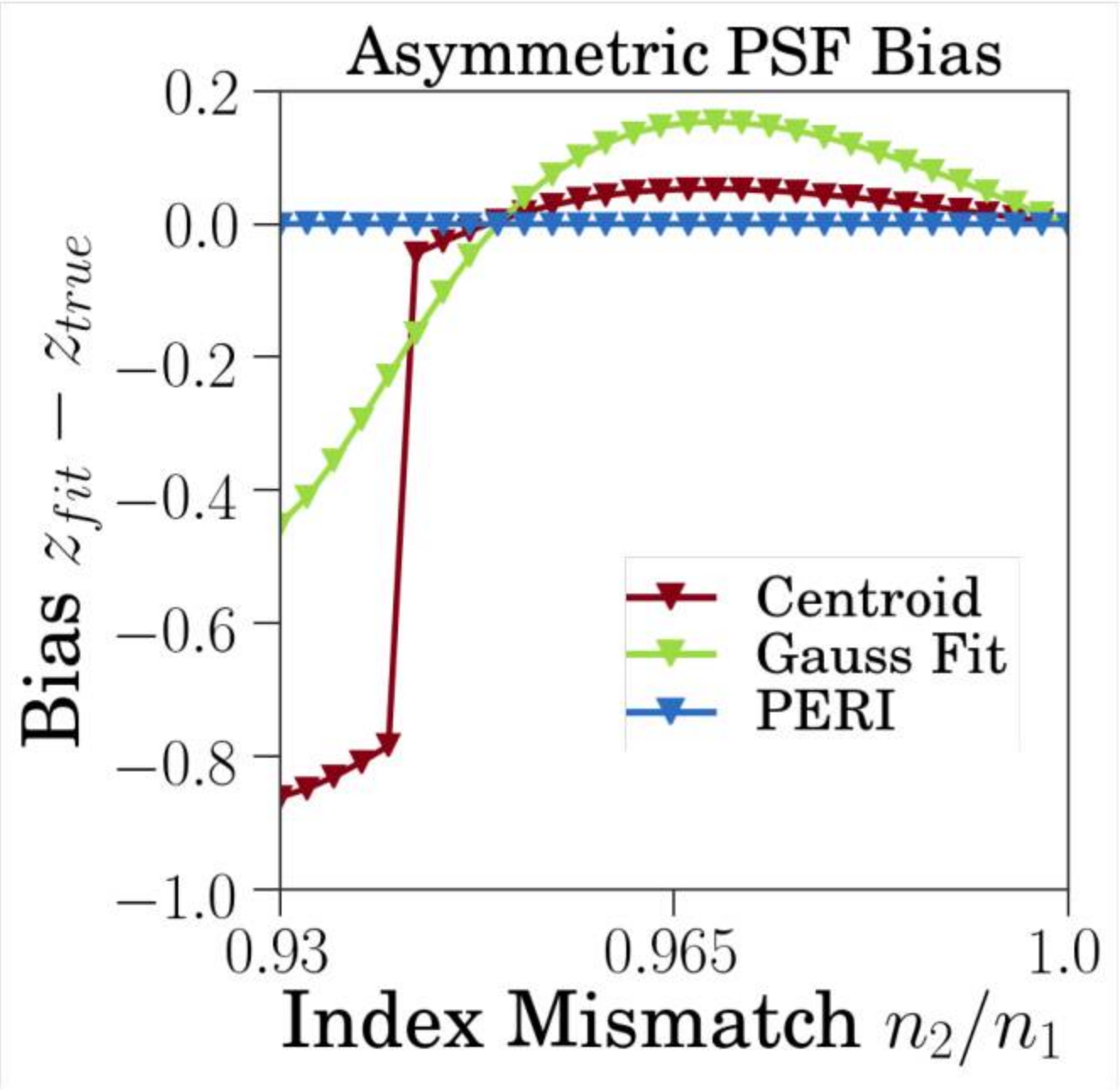


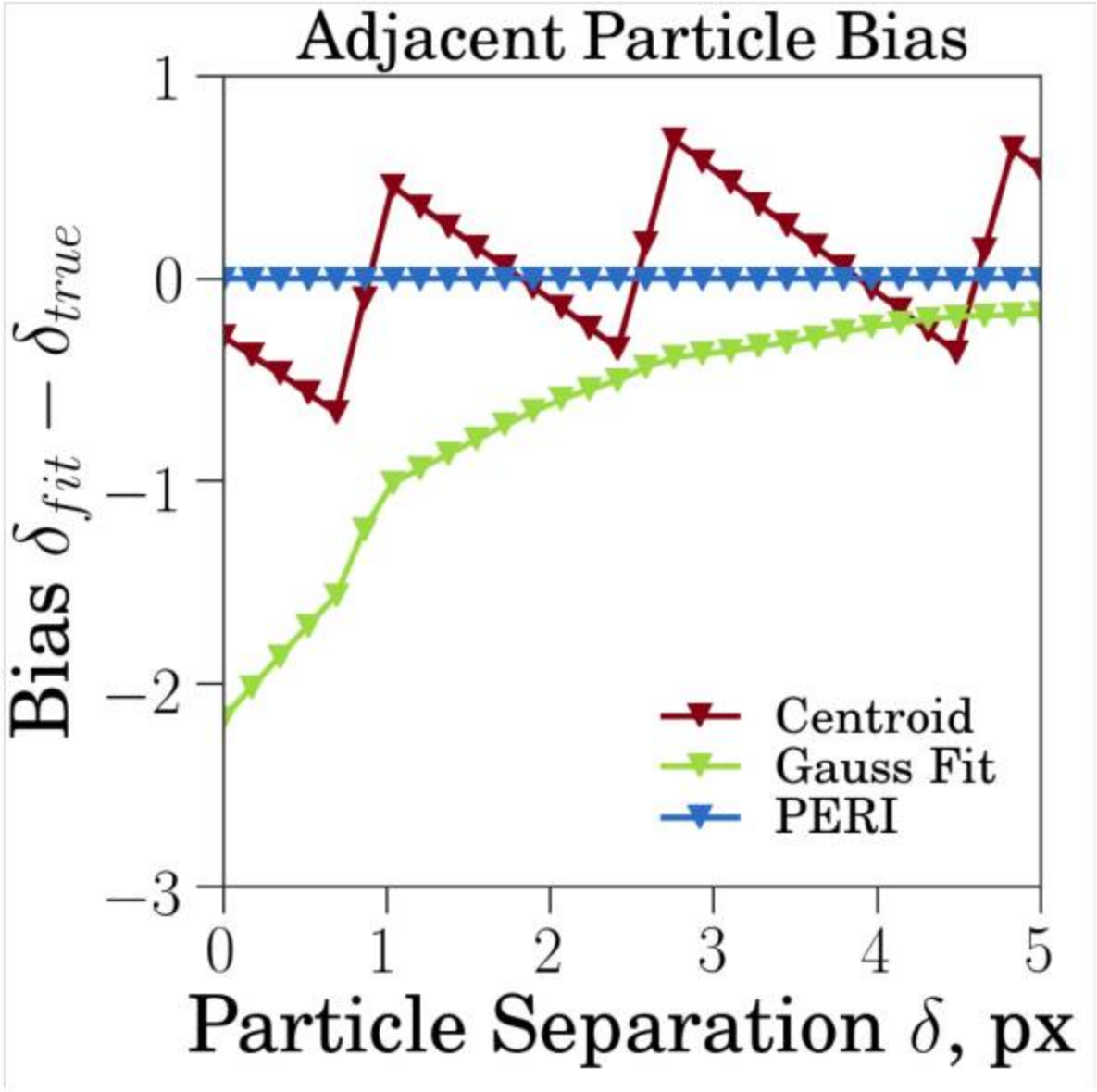

Perfect Pixelated Image
Intensity
1.0
0.8
0.6
0.4
0.2
0.0
0
5
10
15
Image x coord.

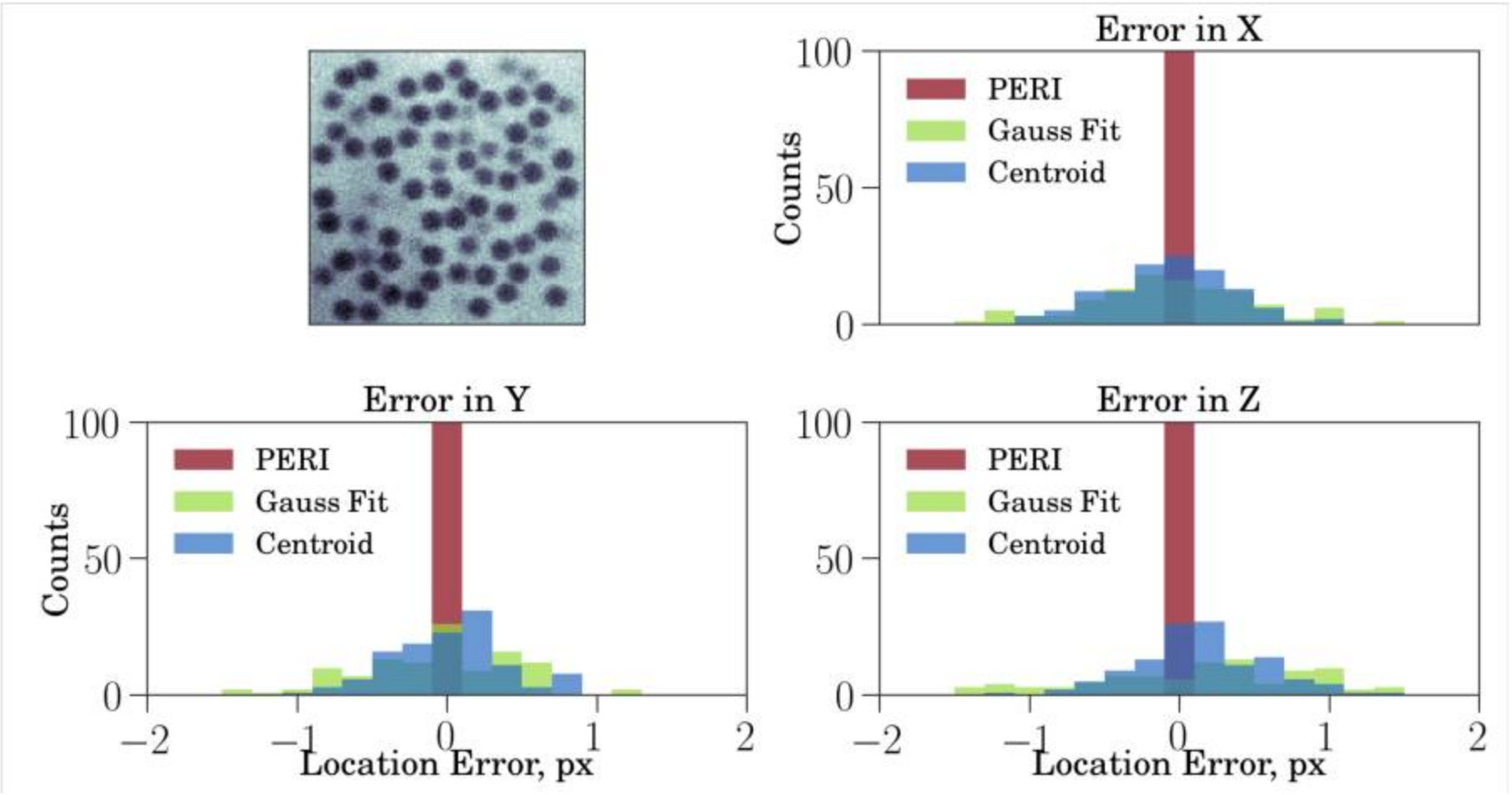

Perfect Pixelated Image
Centroid
Gauss Fit
PERI
Intensity
0.10
0.05
0.00
−0.05
−0.10
9.90
9.95
10.00
10.05
10.10
Image x coord.


Pixel Bias (1,0,0)
Centroid
Gauss Fit
PERI
Bias $x_{fit} - x_{true}$
Image x coord.
0.3
0.2
0.1
0.0
−0.1
−0.2
−0.3
0.0
0.5
1.0


Illumination Bias
Bias $x_{fit} - x_{true}$
0.6
0.4
0.2
0.0
−0.2
Centroid
Gauss Fit
PERI
0.000
0.005
0.010
Illumination Gradient

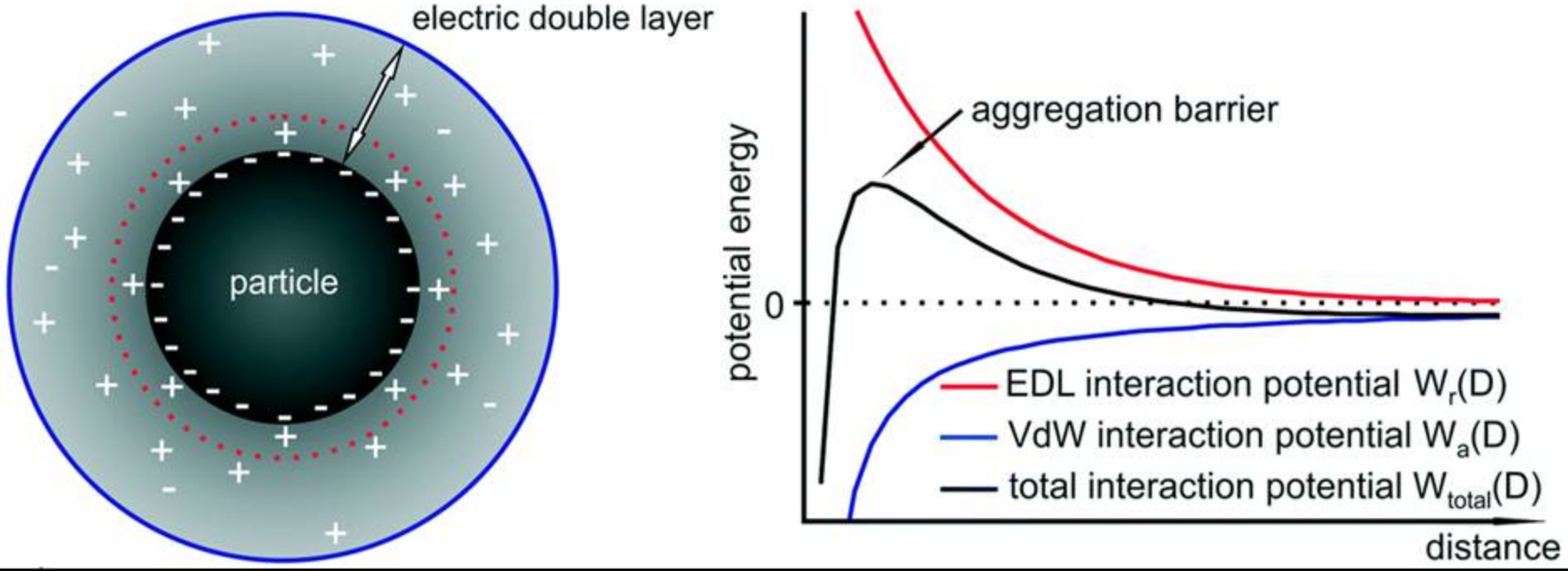

Asymmetric PSF Bias
0.2
0.0
−0.2
−0.4
−0.6
−0.8
−1.0
Bias $z_{fit} - z_{true}$
0.93
0.965
1.0
Index Mismatch n_2/n_1
Centroid
Gauss Fit
PERI

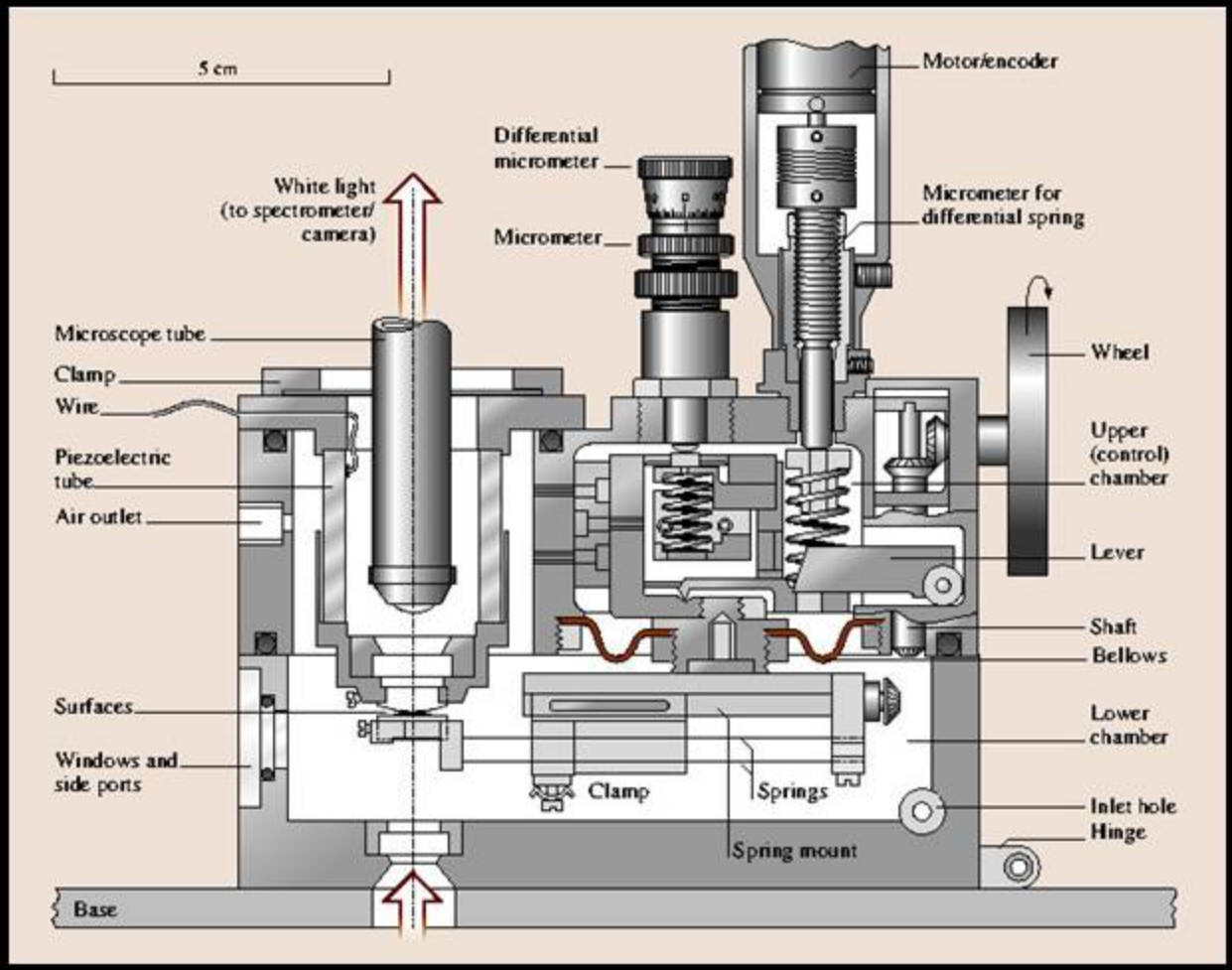

Adjacent Particle Bias
Bias $\delta_{fit} - \delta_{true}$
1
0
−1
−2
−3
0
1
2
3
4
5
Particle Separation δ, px
Centroid
Gauss Fit
PERI



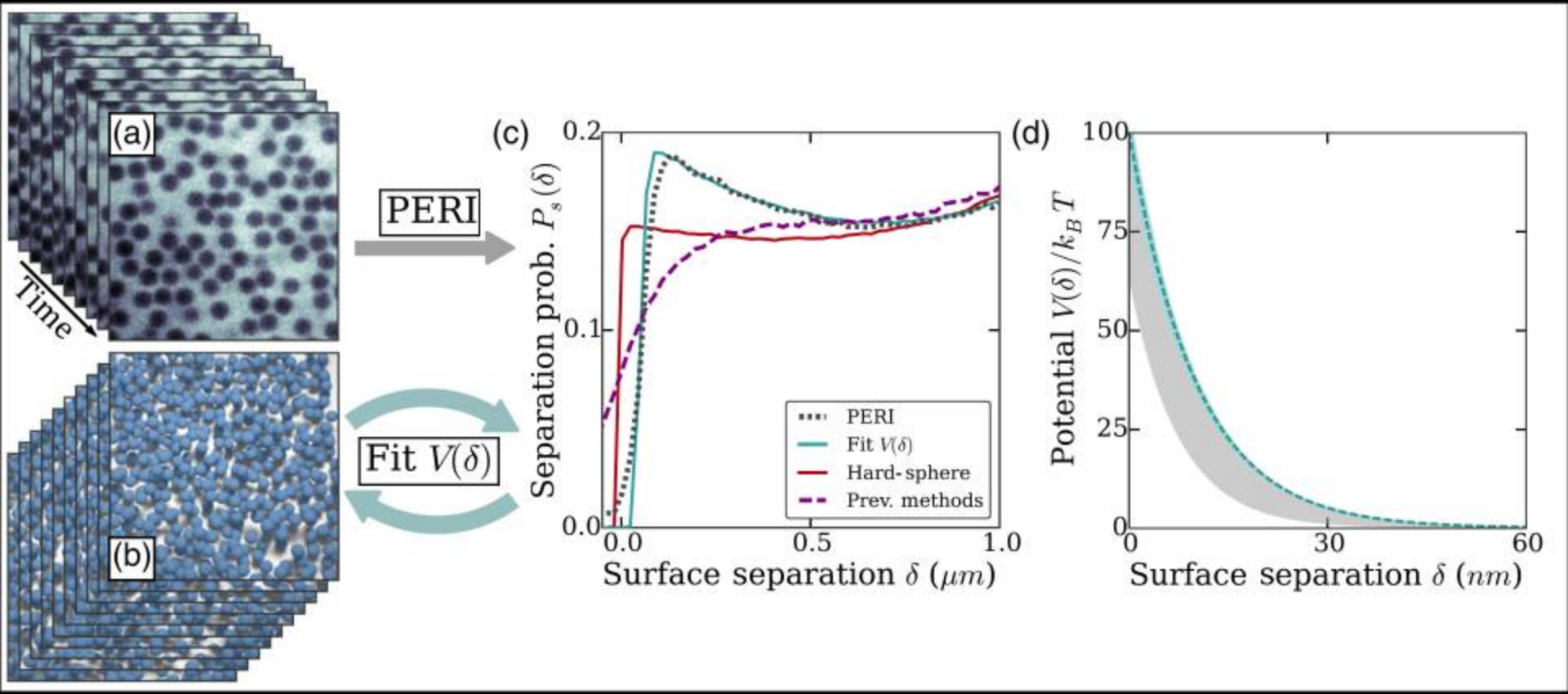
B. Leahy, M Bierbaum, J. Sethna, I Cohen – ArXiv, 2018

Can we measure DLVO potential?




5 cm
Motor/encoder
Differential micrometer
Micrometer
White light (to spectrometer/ camera)
Micrometer for differential spring
Microscope tube
Clamp
Wire
Piezoelectric tube
Air outlet
Wheel
Upper (control) chamber
Lever
Shaft
Bellows
Surfaces
Windows and side ports
Lower chamber
Clamp
Springs
Inlet hole
Hinge
Spring mount
Base

The DLVO potential



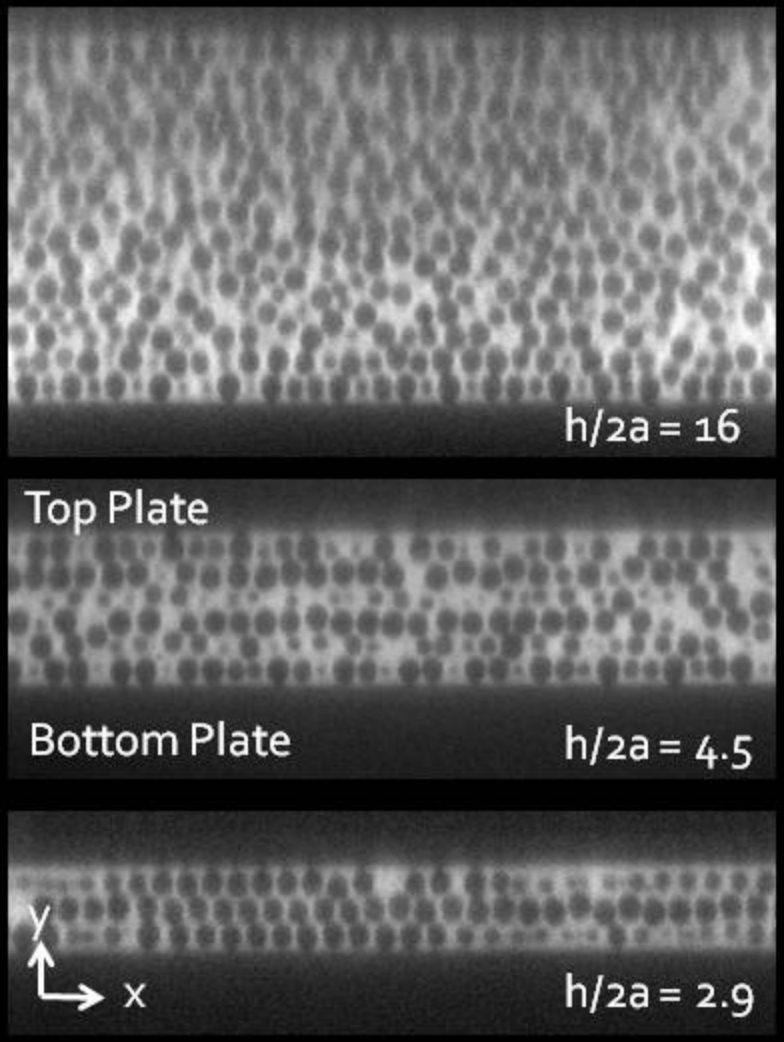
B. Leahy, M Bierbaum, I Cohen, J. Sethna – PRX, 2017

Back to Kirkwood

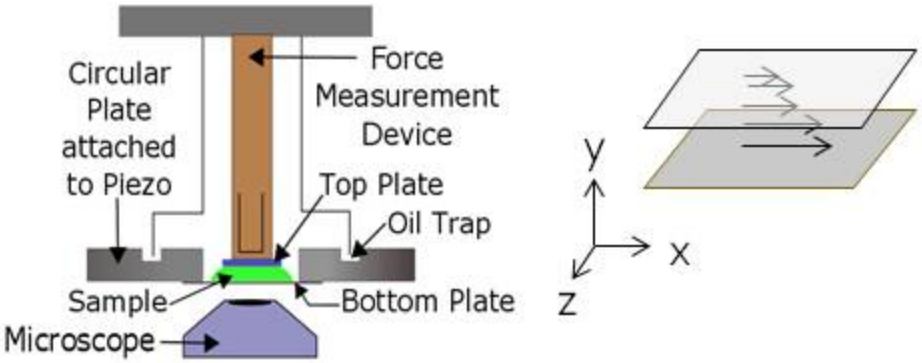
Sum the force contributions

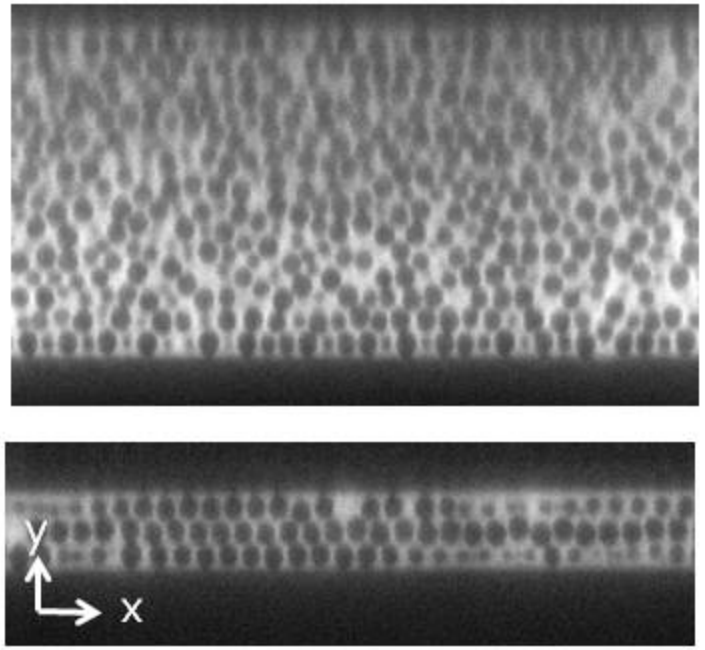

John Gamble Kirkwood
1907 — 1959
Physical Chemist
S.B.–University of Chicago (1926); Ph.D.–Massachusetts Institute of Technology (1929); Sc.D. (Honoris Causa)–University of Chicago (1954) and Universite Libre de Bruxelles (1959)
Served: Yale University as Sterling Professor of Chemistry, Chairman of the Chemistry Department (1951 — 1959) and Director of Division of Sciences (1956 — 1959); Leiden University as Lorentz Professor of Theoretical Physics (1959); California Institute of Technology as Noyes Professor of Chemistry (1947 – 1951); Cornell University as Todd Professor of Chemistry (1938 – 1947); National Academy of Sciences as Foreign Secretary (1954 – 1958); The United States Government as Scientific Consultant (1941 – 1959)
Scientific Honors: American Chemical Society Award in Pure Chemistry (1936); Richards Medal (1950); Lewis Medal (1955)

Example: shear under confinement



Experimental details





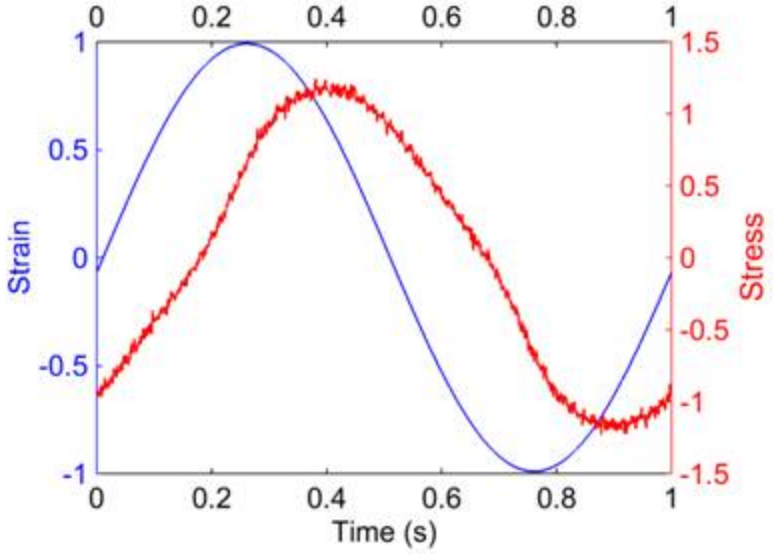
Silica spheres in 80-20 glycerol-water – index matched

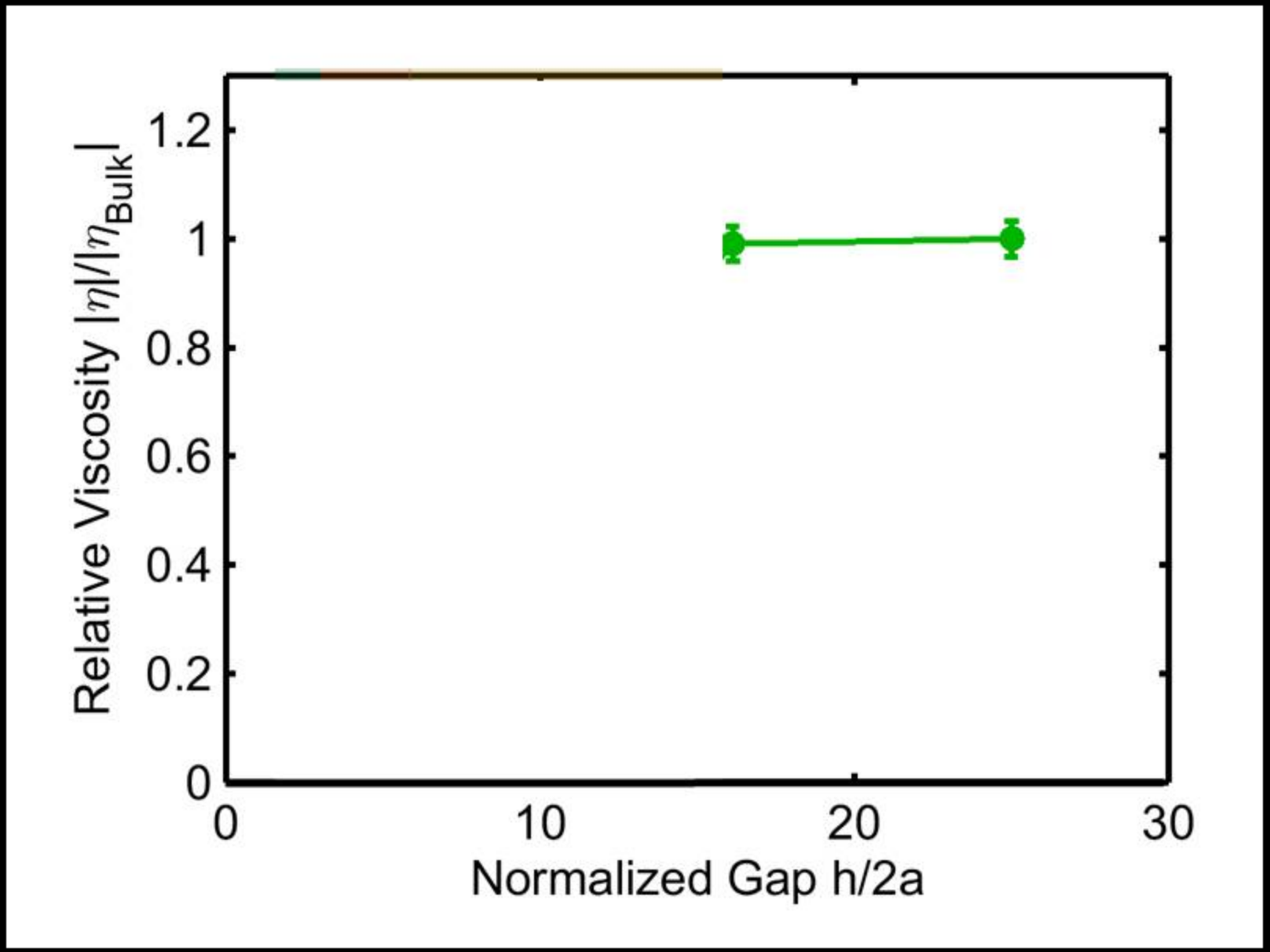
Volume fraction = 0.52

Oscillatory Shear

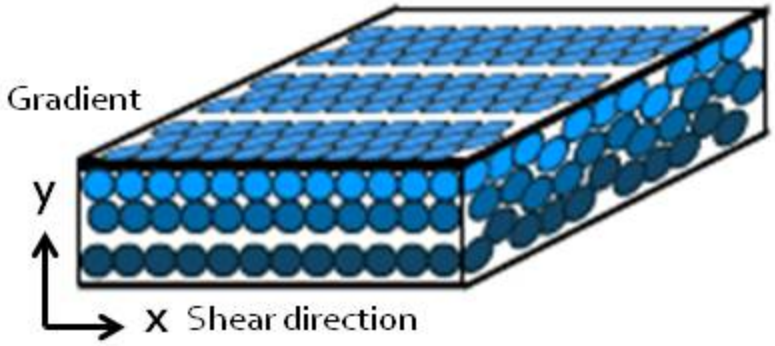
Frequency 1Hz

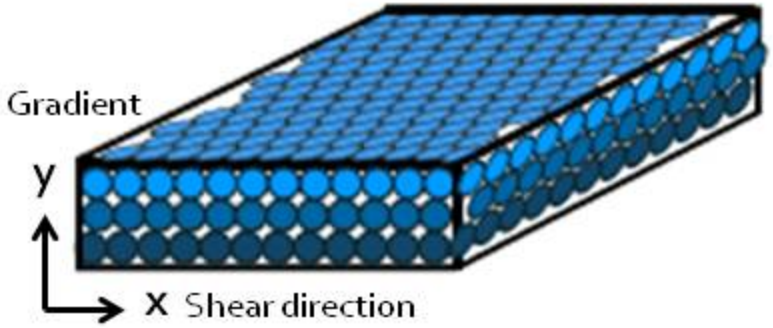
Constant Shear amplitude

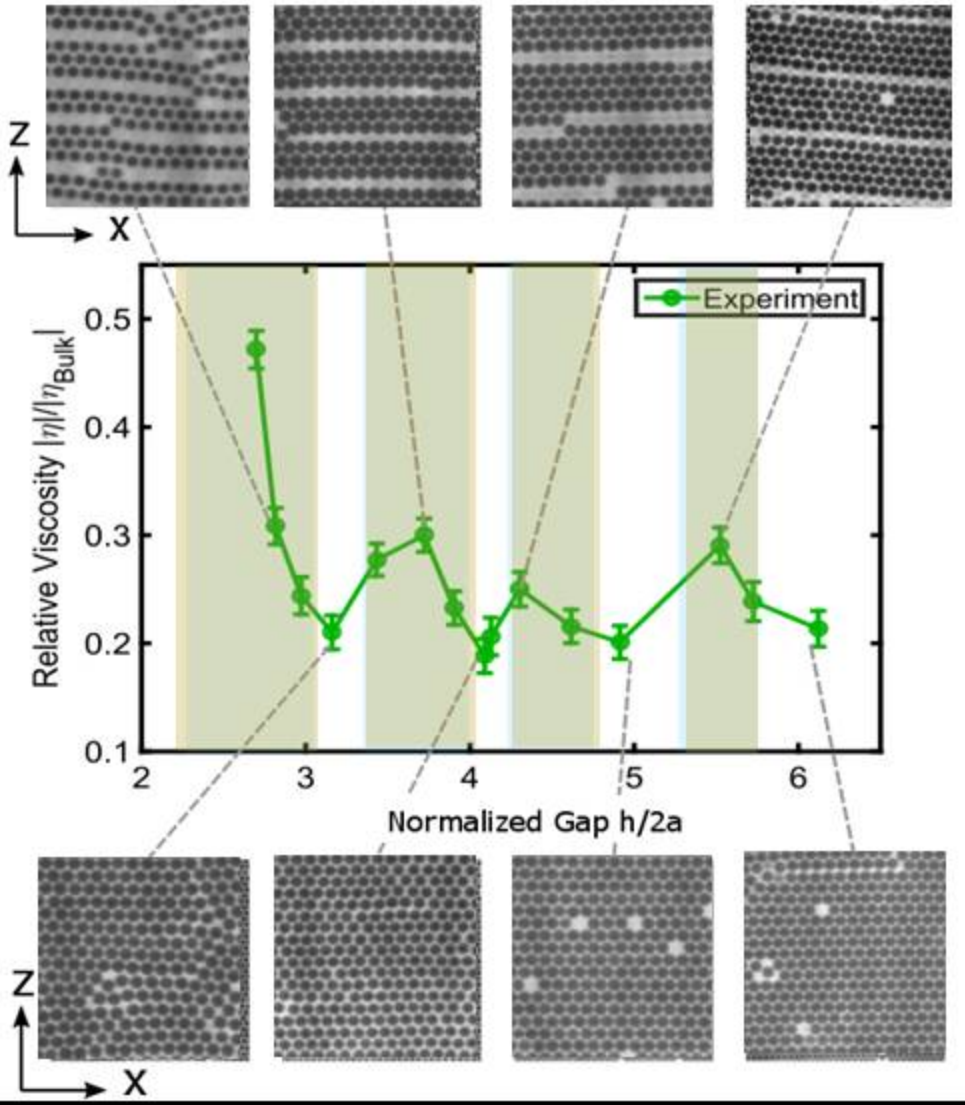



Relative Viscosity $|\eta|/|\eta_{Bulk}|$
1.2
1
0.8
0.6
0.4
0.2
0
0
10
20
30
Normalized Gap h/2a

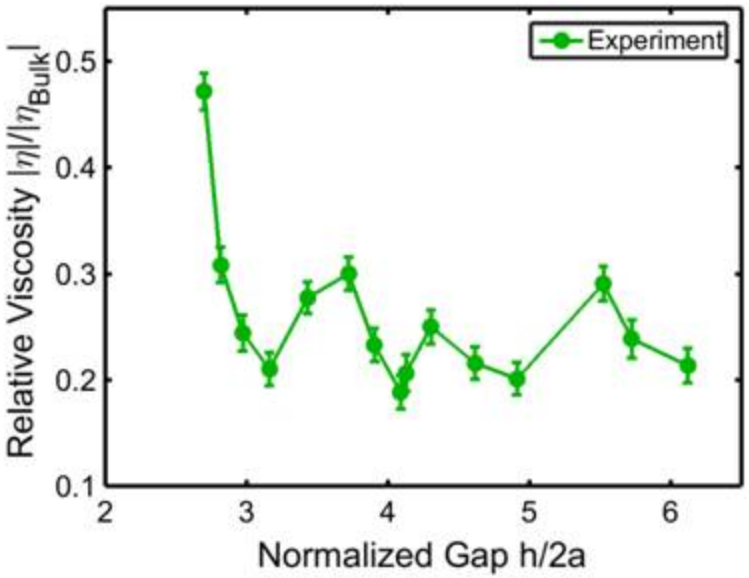
Viscosity of buckled phase

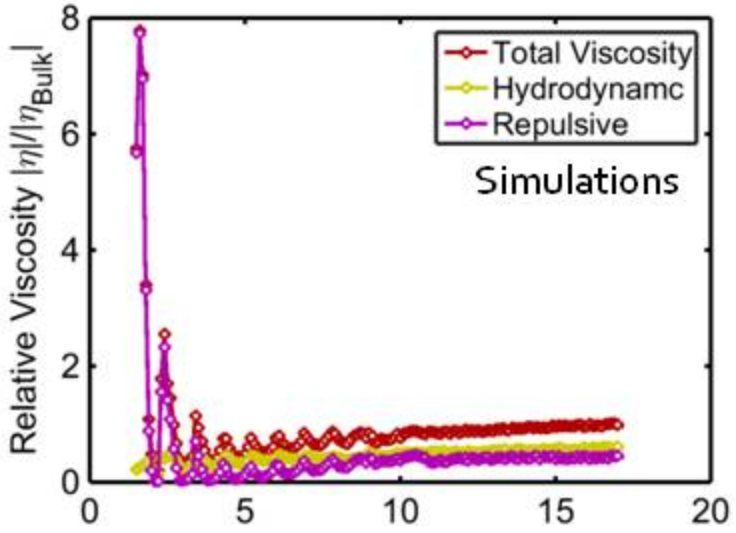




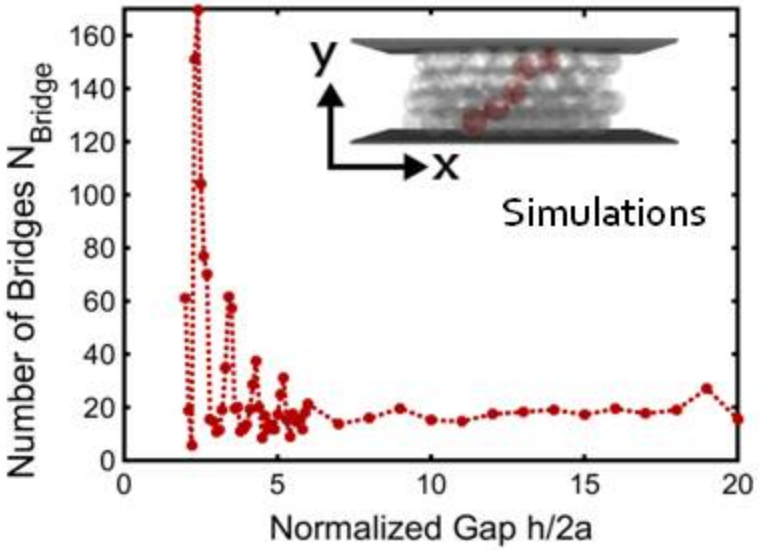


Sharp increase at extreme confinement

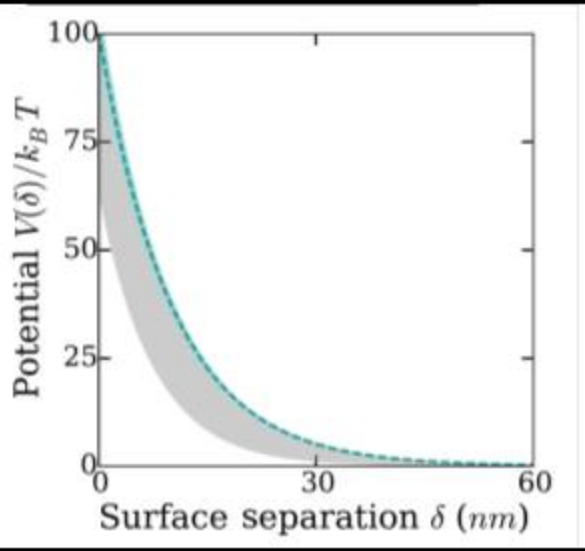


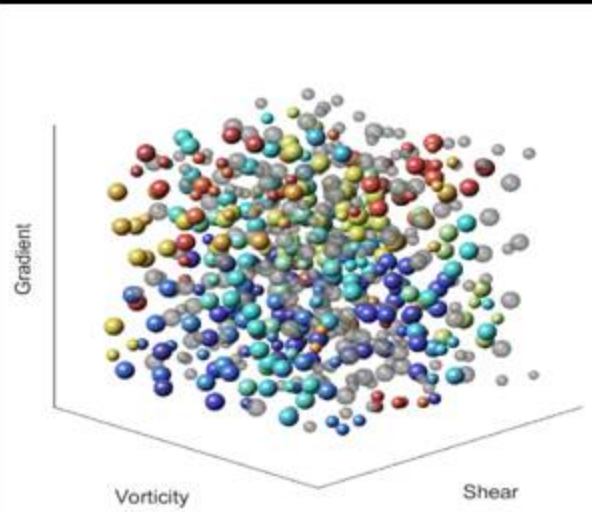


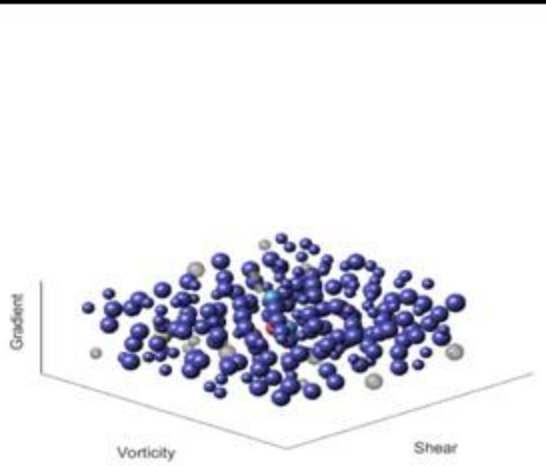
At small gaps, the viscosity increases sharply due to an increase in the short range repulsive forces



Force chains in confined suspensions

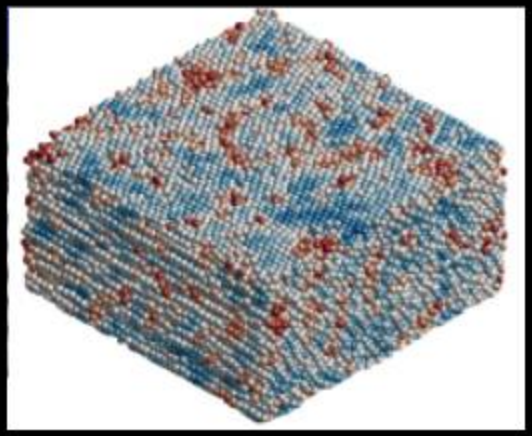


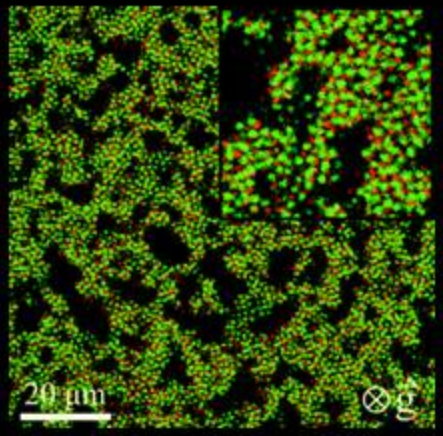


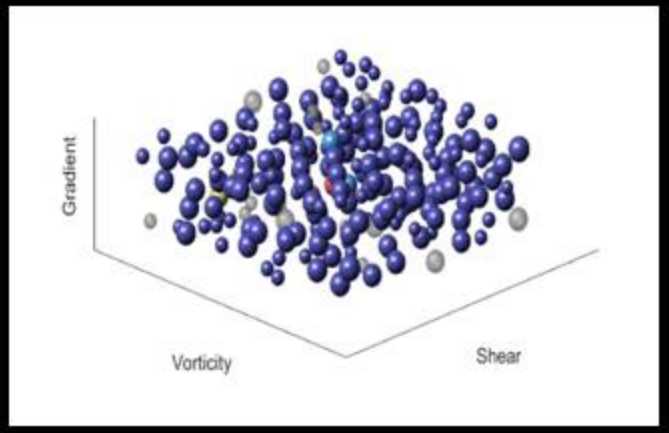


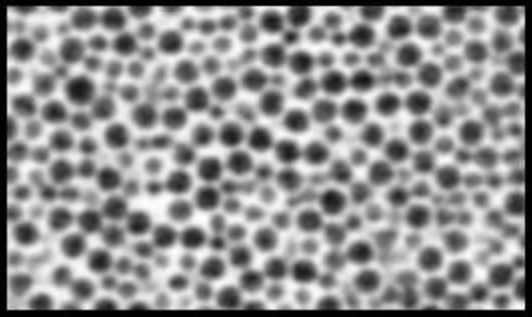
How do they evolve under shear?

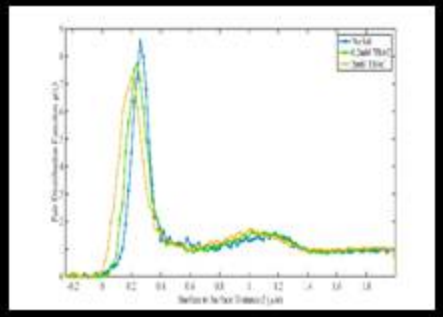
Stay tuned...

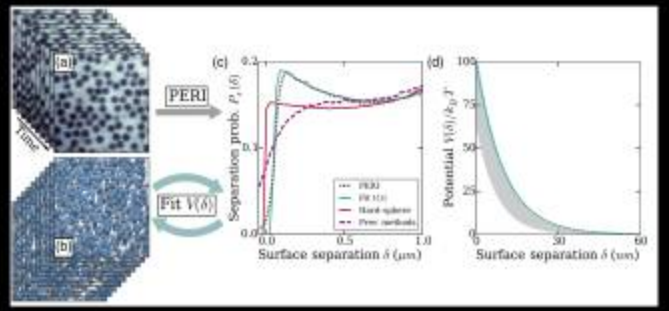


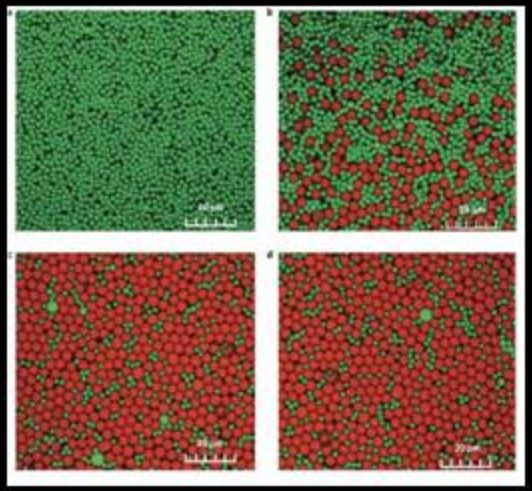

20 μm
⊗g

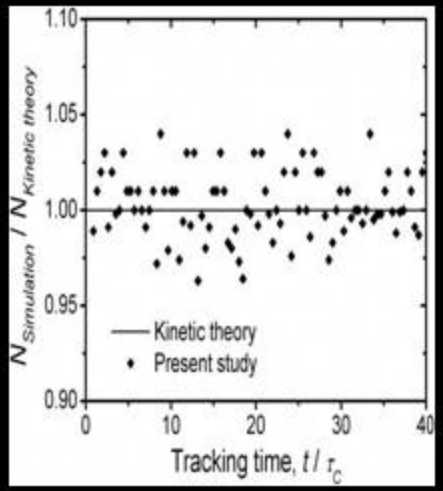

Gradient
Vorticity
Shear

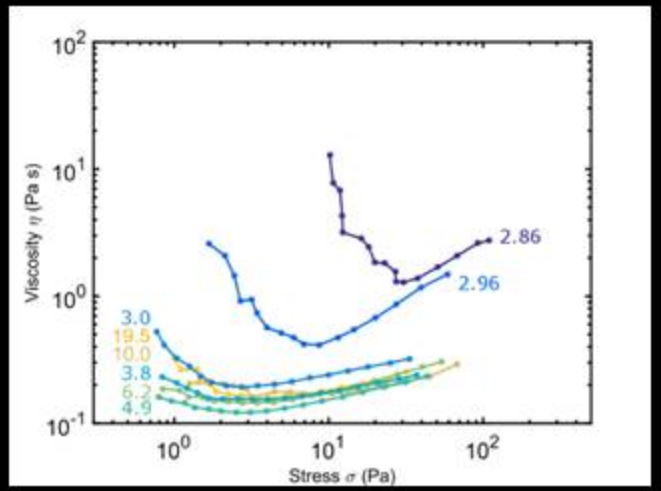





(a)
(b)
PERI
Fit V(δ)
(c)
(d)
Separation prob. P(δ)
Potential V(δ)/k_BT
Surface separation δ (μm)
Surface separation δ (nm)




N_Simulation / N_Kinetic theory
Kinetic theory
Present study
Tracking time, t / τ_c

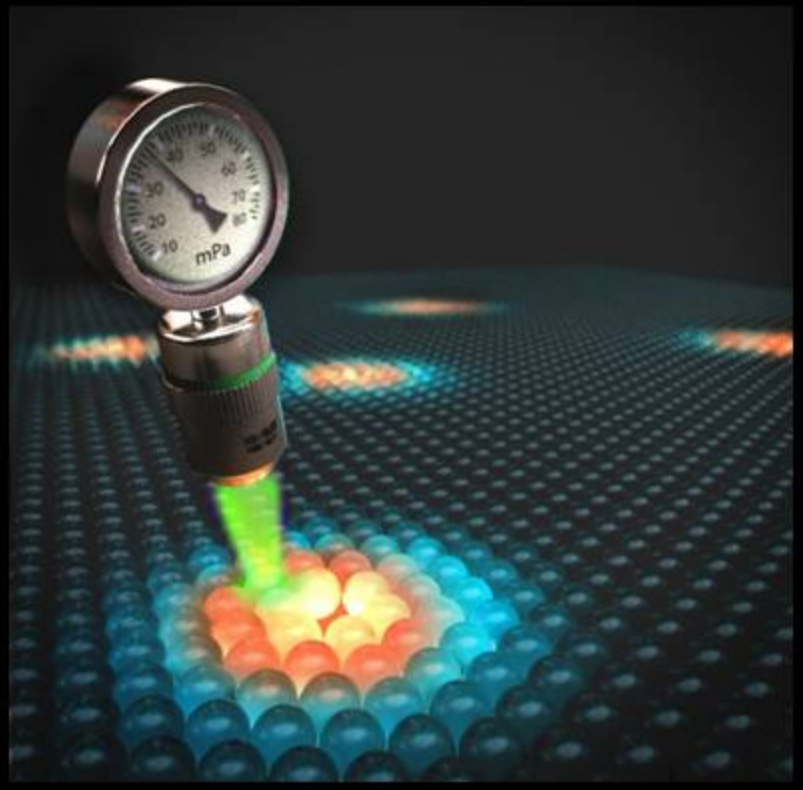

Viscosity η (Pa s)
Stress σ (Pa)
2.86
2.96
3.0
19.5
10.0
3.8
6.2
4.9

The Cohen Group

"You can observe a lot by watching" Yogi Berra

Papers: structure to rheology

- Imaging the microscopic structure of shear thinning and thickening colloidal suspensions
  X Cheng et al. Science (2011)
- Assembly of vorticity aligned hard-sphere colloidal strings
  X Cheng et al. PNAS (2012)
- Enhancing Rotational Diffusion Using Oscillatory Shear
  BD Leahy et al. PRL (2013)
- Biaxial shear of confined colloidal hard spheres
  YC Lin et al. Soft Matter (2014)
- A multi-axis confocal rheoscope
  YC Lin et al. Rev Sci. Inst. (2014)
- The effect of shear flow on the rotational diffusion of a single axisymmetric particle
  BD Leahy et al. JFM (2015)
- Hydrodynamic and Contact Contributions to Continuous Shear Thickening in Colloidal Suspensions
  YC Lin et al. PRL (2015)
- Stress Assessment from Local Structural Anisotropy (SALSA) YC
  YC Lin et al. Nature Materials (2016)
- Tunable shear thickening
  YC Lin et al. PNAS (2016)
- Maximum Resolution Microscopy (MRM)
  MK Bierbaum et al. PRX (2017)
- Determining Viscosities Using the Green-Kubo Relation
  YC Lin et al. PRL (2017)
- How confinement induced structures alter viscosity contributions
  M Ramaswamy et al. PRX (2017)



Thanks: Gareth Mckinley, John Brady, Don Koch, Roseanna Zia, Heinrich Jaeger, Eric Brown, and the Cohen group.