Sheared colloids and emulsions studied with confocal microscopy

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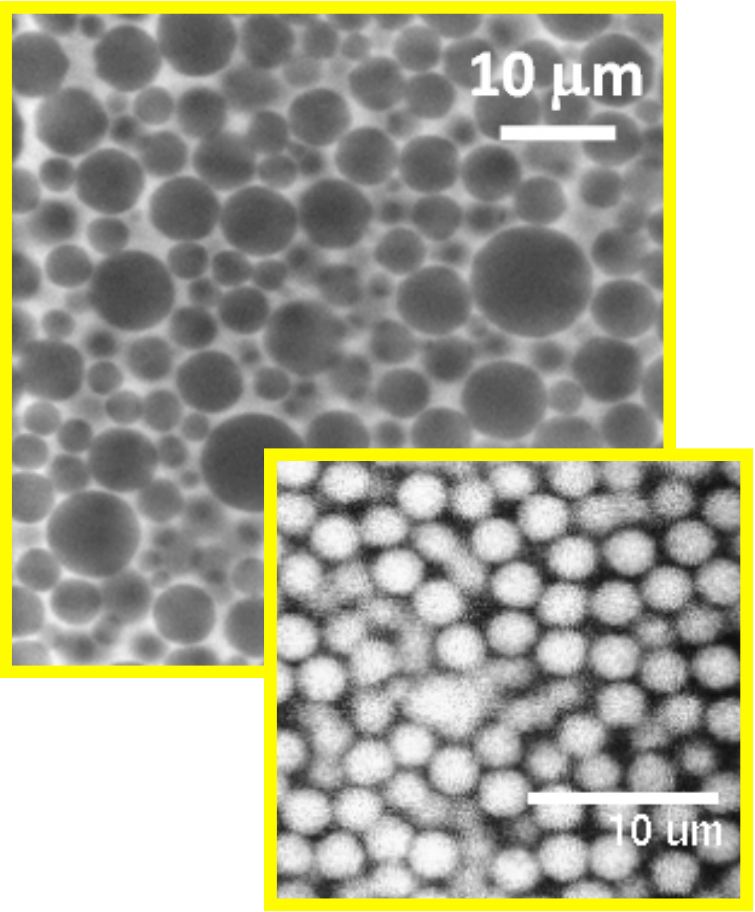
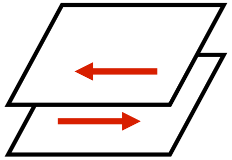
Funding by NSF-DMR

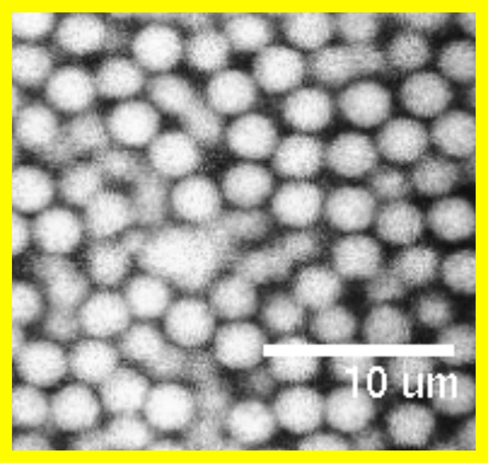
Colloidal particles from Andrew Schofield,
University of Edinburgh



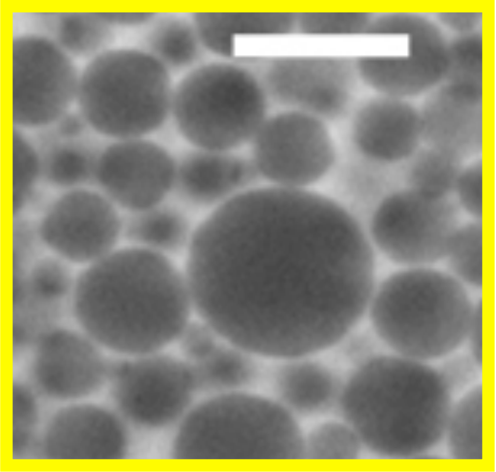
Overview

- Study dense amorphous ("jammed") samples
- How do they deform microscopically under shear?



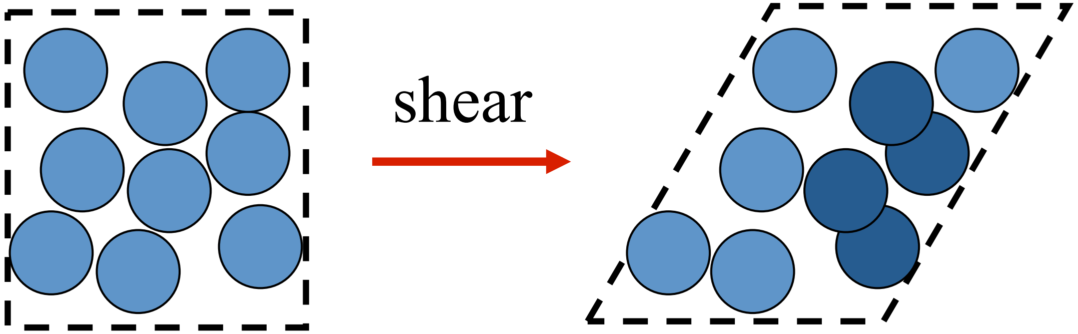


Colloidal particles: hard, monodisperse
(Dandan Chen's work)



Emulsion droplets: soft, polydisperse
(Joaquim Clara Rahola's work)

Basic problem: particles collide, must find way to rearrange

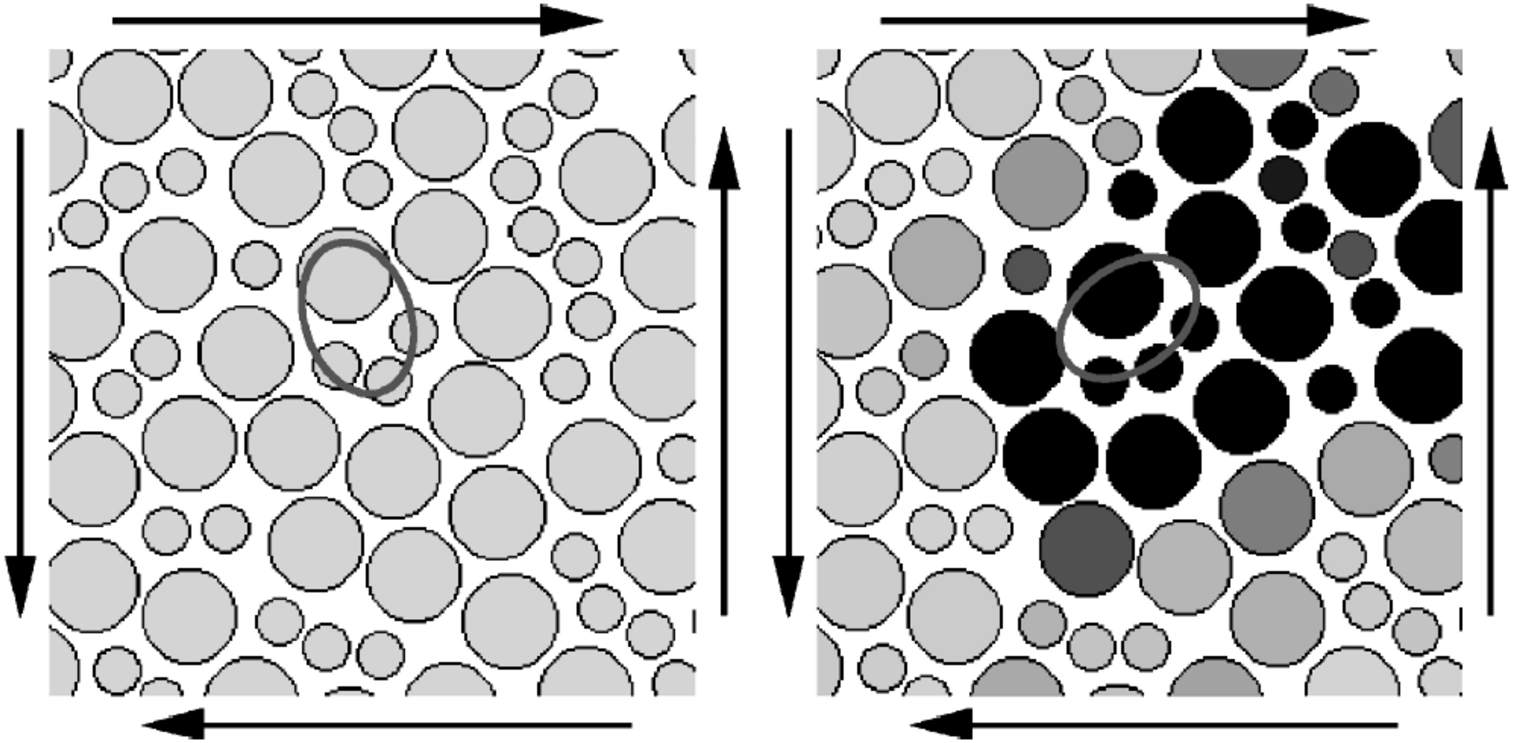


Like the "David Pine" effect. For dense amorphous samples, even small strains cause collisions. Unlike David's talk, our particles are influence by Brownian motion.

"shear-induced cage breaking" – Akira Furukawa

One possibility: shear transformation zones

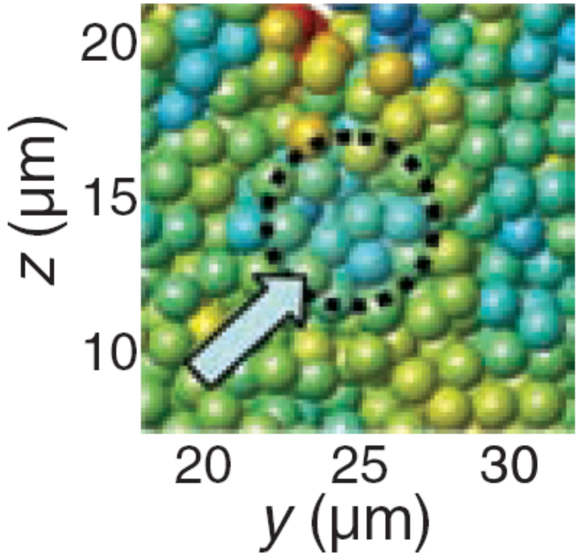
(ML Falk and JS Langer, Phys. Rev. E **57**, 7192 (1998)]

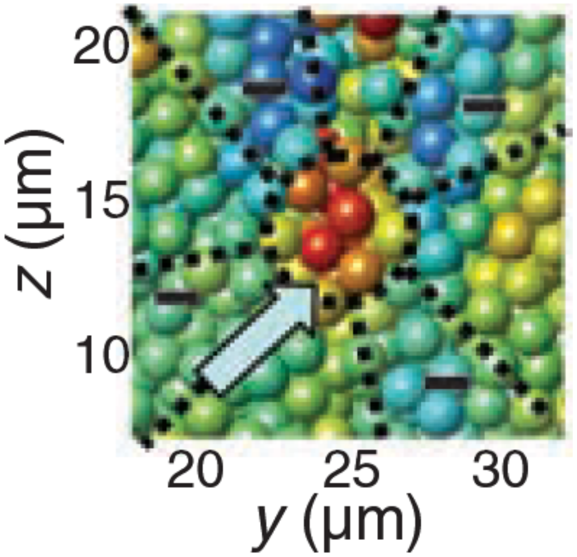


2D Lennard-Jones simulations of Falk & Langer

One possibility: shear transformation zones

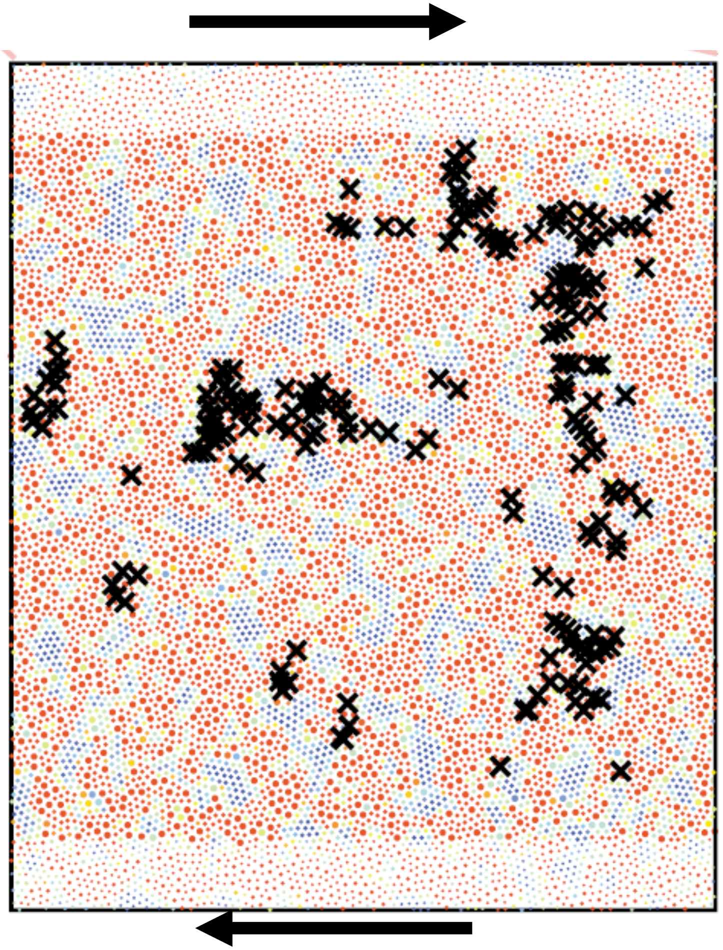
(ML Falk and JS Langer, Phys. Rev. E **57**, 7192 (1998)]





3D colloidal experiments: 1.5 μm dia monodisperse particles
Blue = negative strain, red = position (direction of shear)
Total strain $\gamma = 0.01$
[P Schall, DA Weitz, & F Spaepen, Science (2007)]

Can also have “avalanches” of rearranging particles

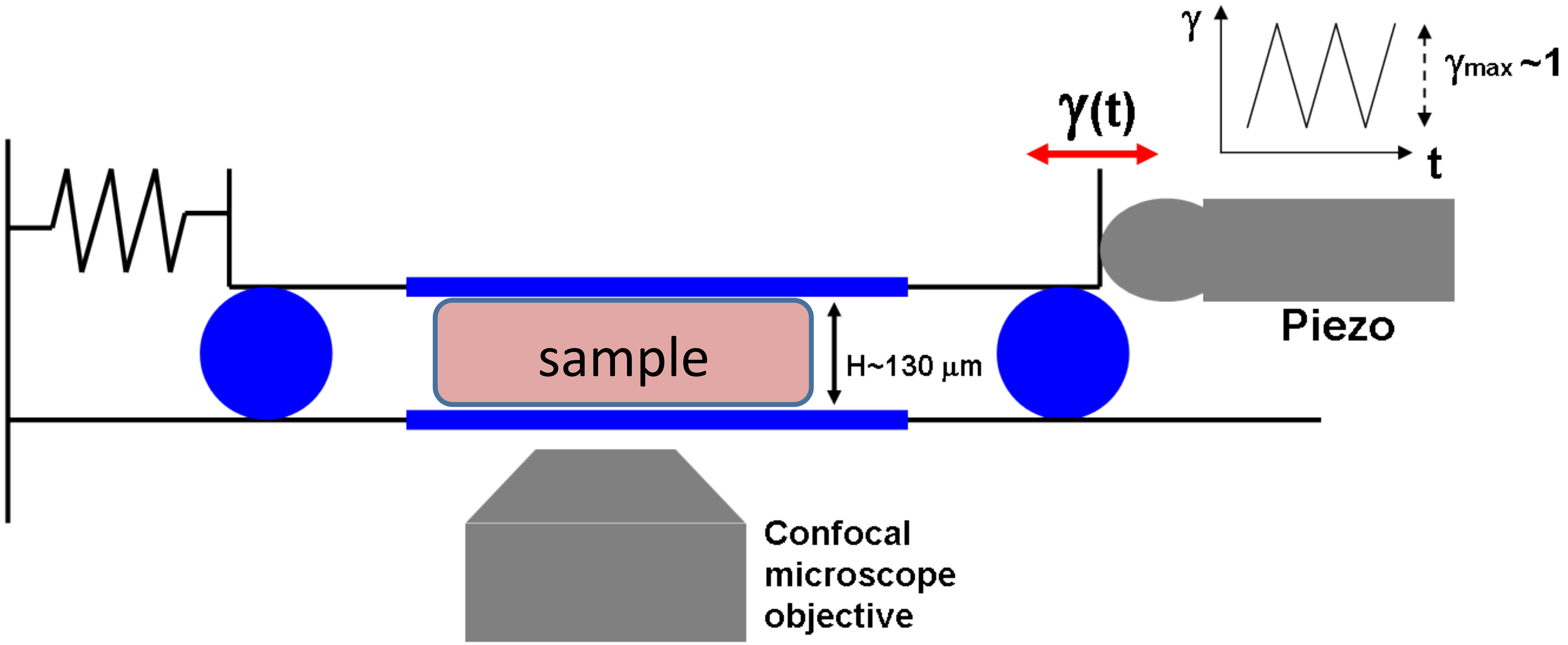


H Shiba & A Onuki PRE 2010

Sheared glassy 2D binary mixture, black X’s mark rearranging particles, $\Delta\gamma = 0.005$

<u>See poster</u>: Hayato Shiba

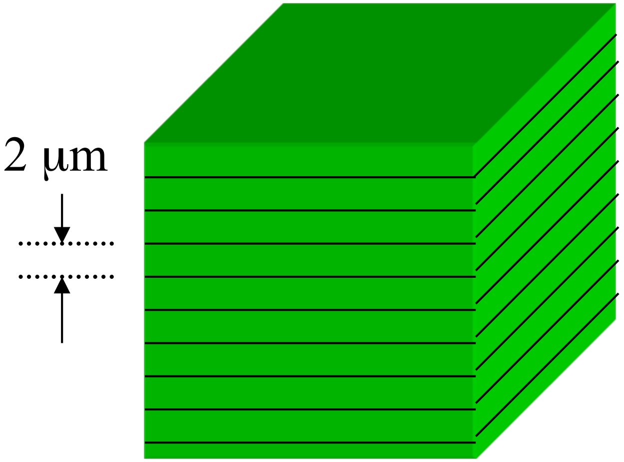
Controlled strain, parallel plate shear cell

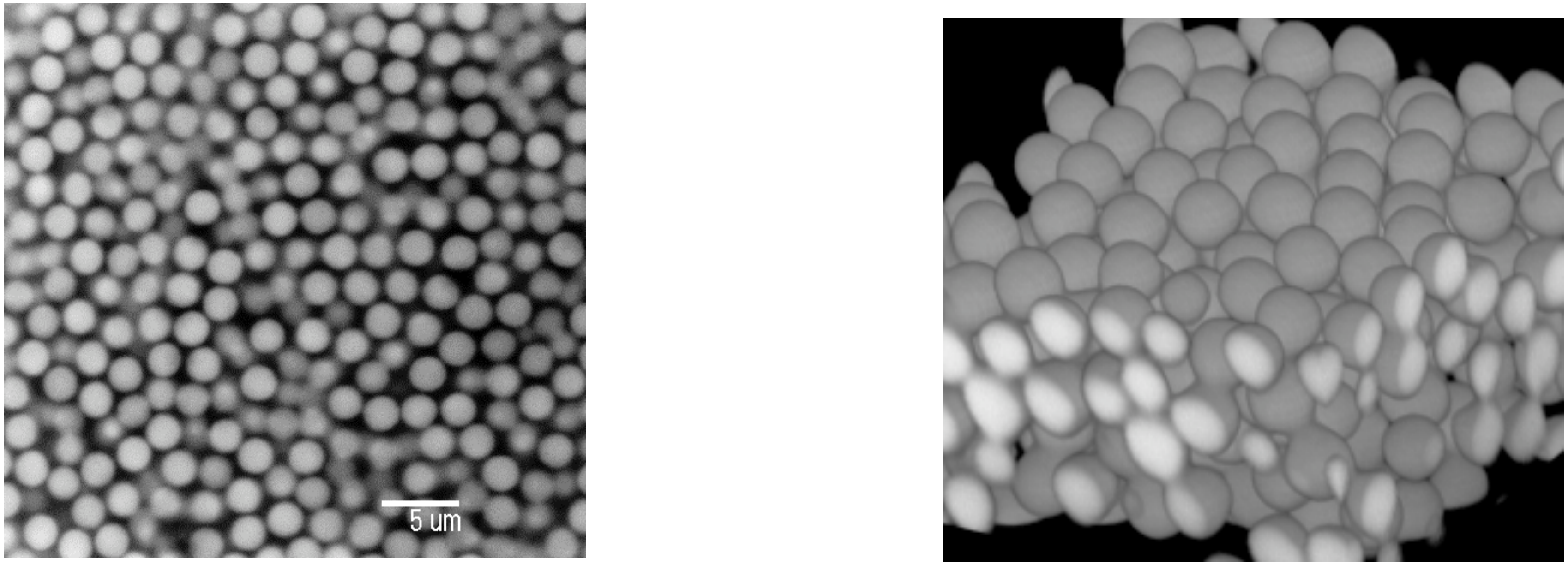


stick colloids and/or Scotchgard to plates to diminish slip

Confocal microscopy for 3D pictures

Scan many 2D slices,
reconstruct 3D image





2D and 3D images of 2.3 μm diameter PMMA particles

Microscopy and Tracking

software: http://www.physics.emory.edu/~weeks/idl/
Dinsmore, Weeks, Prasad, Levitt, & Weitz, Appl. Optics ’01

Microscopy:

- 30 images/s (512×480 pixels, 2D)
- one 3D “chunk” per 2 - 20 s
- $67 \times 63 \times 20\ \mu m^3$
- 100× oil / 1.4 N.A. objective
- Identify particles within 0.03 μm (*xy*), 0.05 μm (*z*)

Particle tracking:

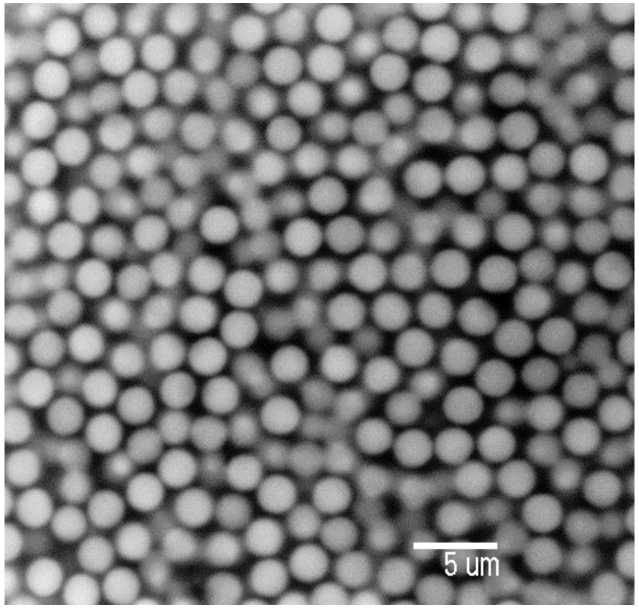
- Follow 3000-5000 particles, in 3D
- 200-1000 time steps = hours to days
- ≈ 4 GB of images per experiment

Part 1: Shear of dense colloidal suspensions

D Chen *et al.*, Phys Rev E **81**, 011403 (2010)

Colloidal System

- 2.3 mm diameter PMMA colloids
- density matched solvent (cyclohexylbromide + decalin)
- slightly charged hard spheres
- provided by A Schofield & WCK Poon, Univ. of Edinburgh



one of Andrew Schofield’s cabinets

Colloidal glass transition

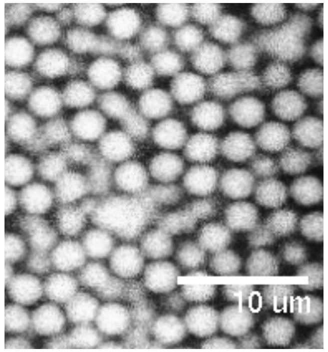
- Control parameter is volume fraction ϕ
- Glass exists when $\phi > \phi_g \approx 0.58$

  (agrees with simulations with slight polydispersity)

- Diffusion constant → 0
- See aging behavior

  (Courtland & Weeks '03; Cianci, Courtland, Weeks '06;
  Lynch, Cianci, & Weeks '08)

Our experiments:
$\phi \approx 0.51\text{-}0.57$



How fast do we shear?

Use triangle wave driving, strain rate $\dot{\gamma}$, period 150-450 s

Compare with time scale to diffuse radius *a* in unsheared sample,

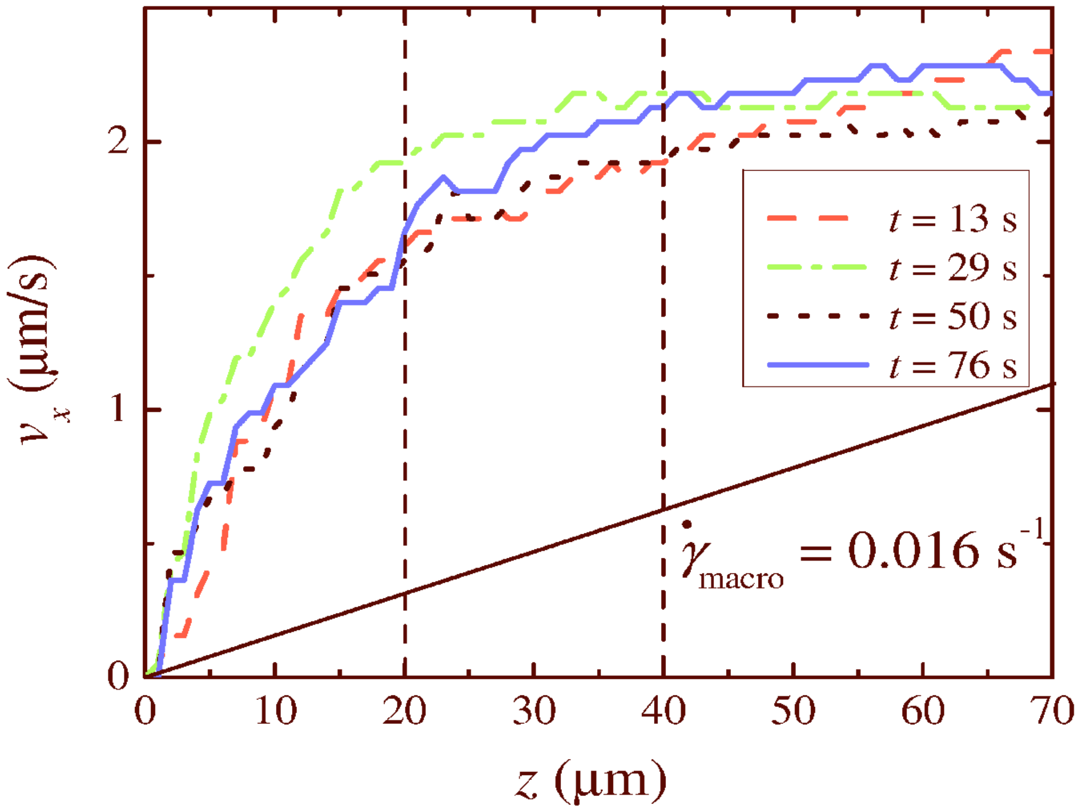
$$\tau_D = \frac{a^2}{2D_\infty} \approx 3000 \text{ s}$$

Define Peclet number $\boxed{\mathrm{Pe} = \frac{\dot{\gamma} a^2}{2D_\infty} = 7-21}$

Shear-induced motion is more significant than thermal motion!

(same idea as $\dot{\gamma}\tau > 1$)

Aside: we have shear bands



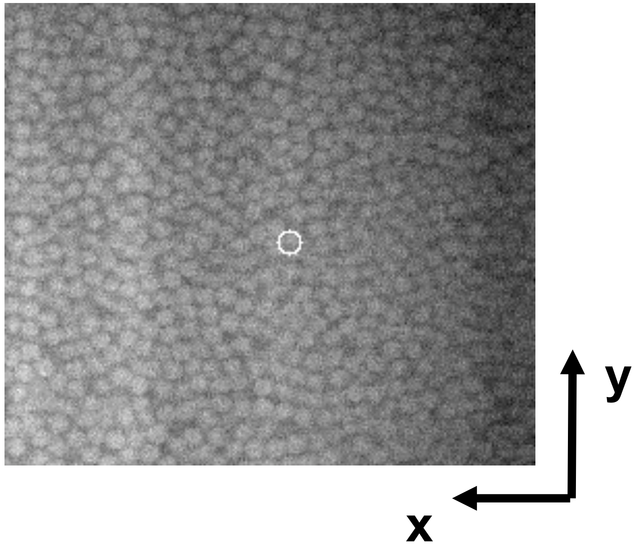
- Focus on region where velocity gradient is linear
- Define local (mesoscopic) strain rate:

$$\dot{\gamma}_{meso}(t) \equiv \frac{v_x(z_2,t) - v_x(z_1,t)}{z_2 - z_1}$$

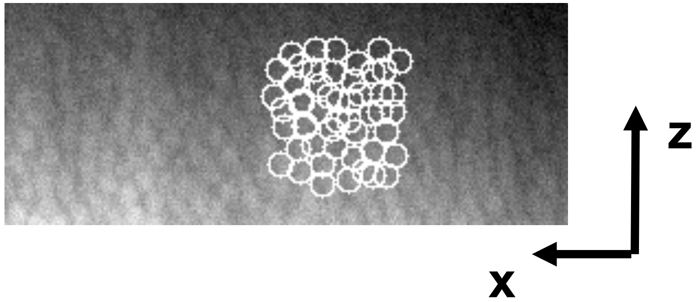
- $\dot{\gamma}_{meso}$ is control parameter

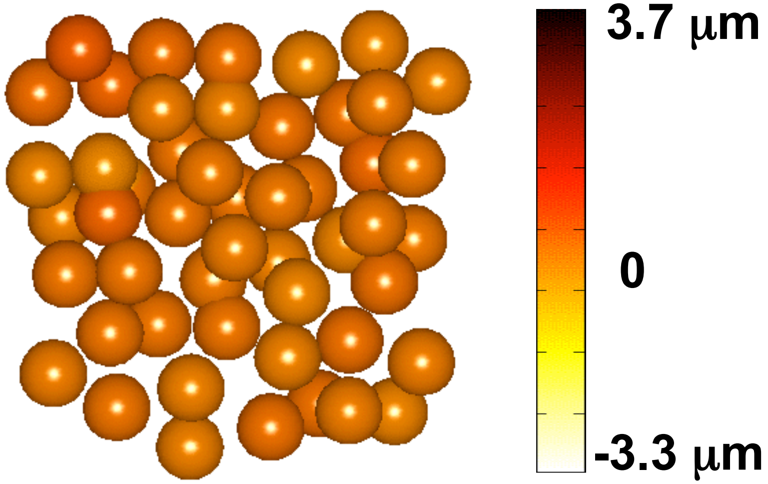
Movie and tracking

Top view



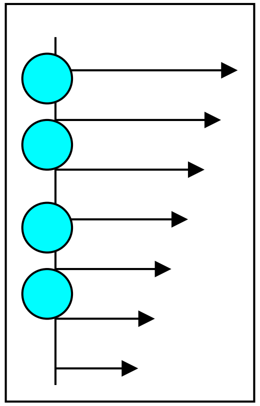
Side view



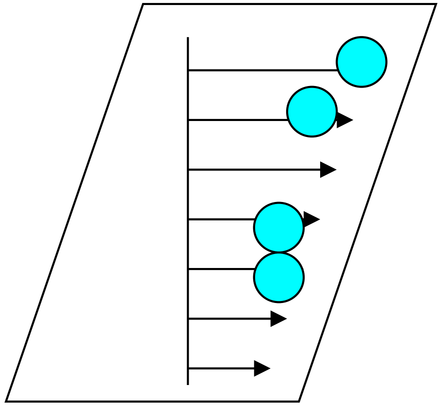


Relative rearrangements between neighbors makes structure change.

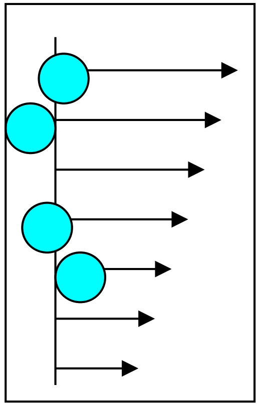
Examine nonaffine motion: $\Delta\tilde{x}$



1. Initial unstrained sample



2. Strained sample



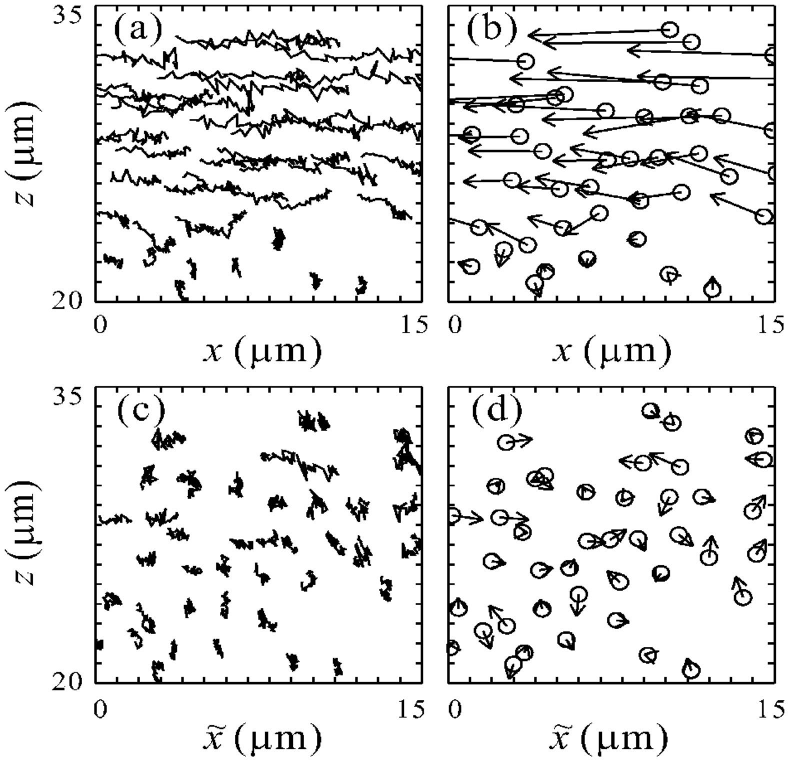
3. Remove affine motion of strain field

4. Motion in y, z left unchanged

Examine nonaffine motion: $\Delta\tilde{x}$

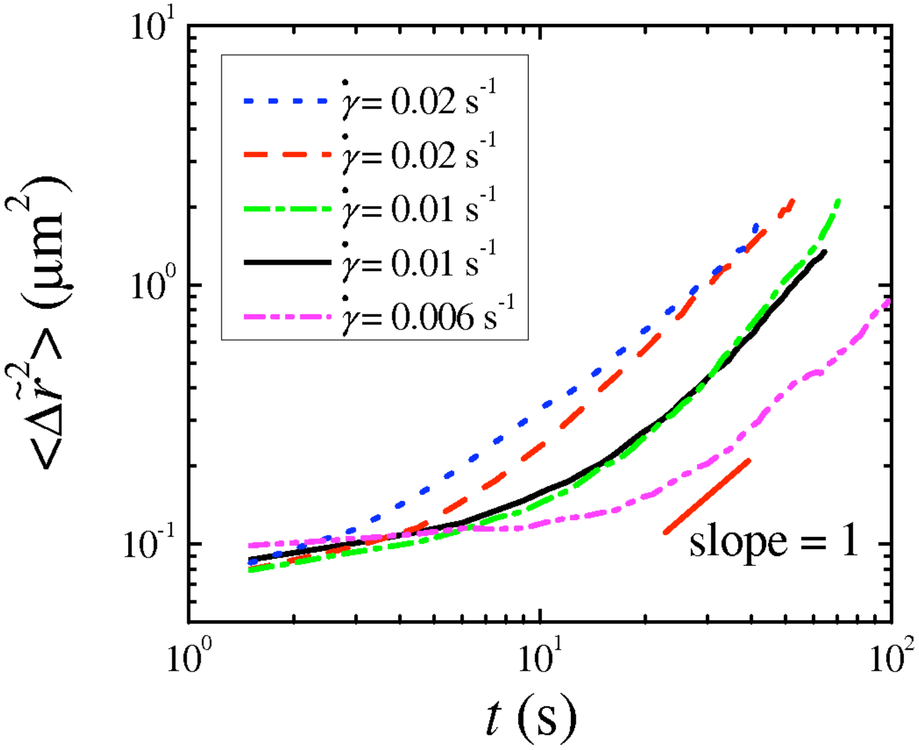
2. Strained sample

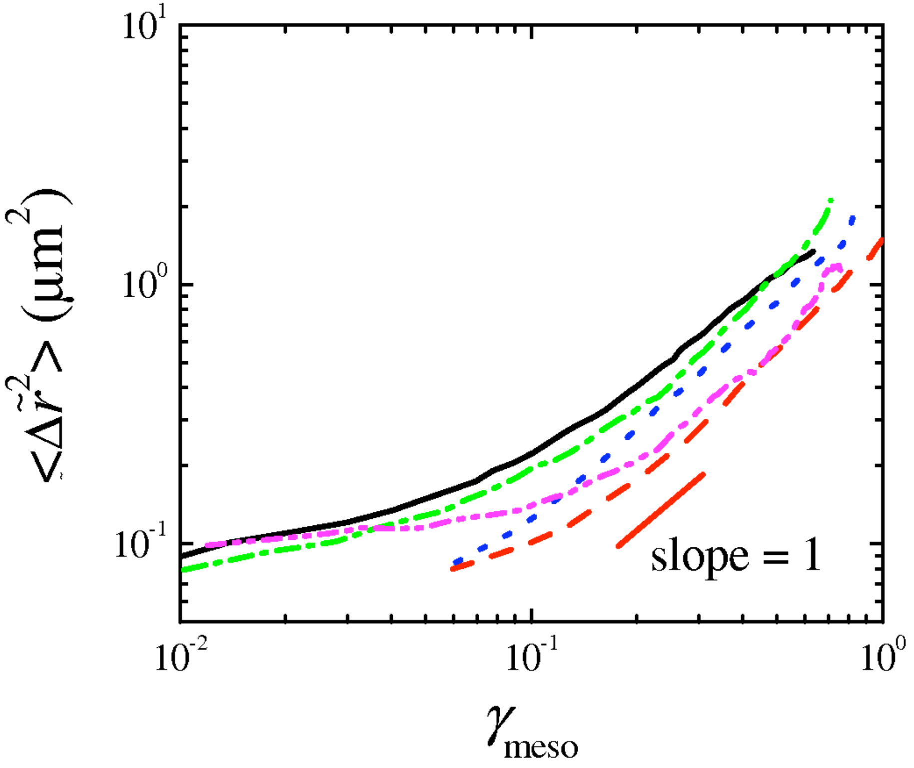
3. Remove affine motion of strain field



$\phi = 0.51$, $\gamma_{\mathrm{meso}} = 0.43$, Pe ≈ 20

Shear-induced motion: accumulated strain is key





$$\gamma_{meso}(t) = \int_0^t \dot{\gamma}_{meso}(t')dt'$$

Shear-induced motion: accumulated strain is key

Comments:

- Has been seen before

(Yamamoto & Onuki 1998; Pine 2005; Maloney & Robbins 2008)

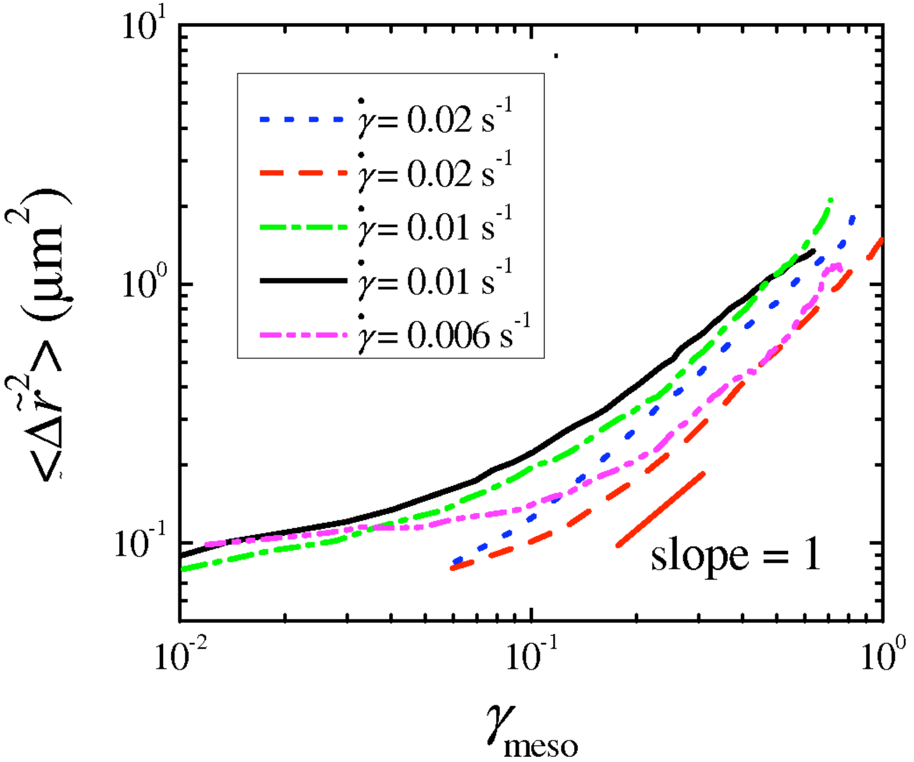
- Our result implies $D \sim (\dot{\gamma})^{1.0}$

(agrees with Eisenmann 2010, Ovarlez 2010)

- Prior work found $D \sim (\dot{\gamma})^{0.8}$

(Besseling, Weeks, Schofield & Poon 2007)

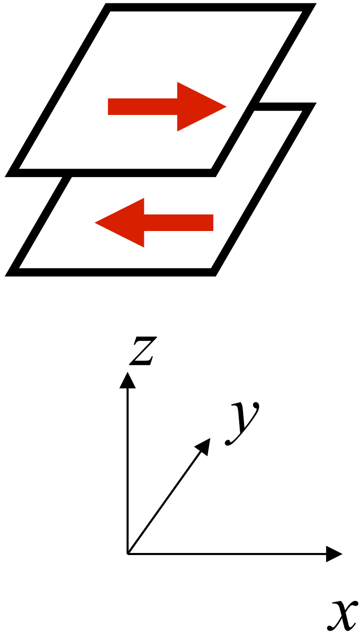
- Prior work was larger strains, glassy samples

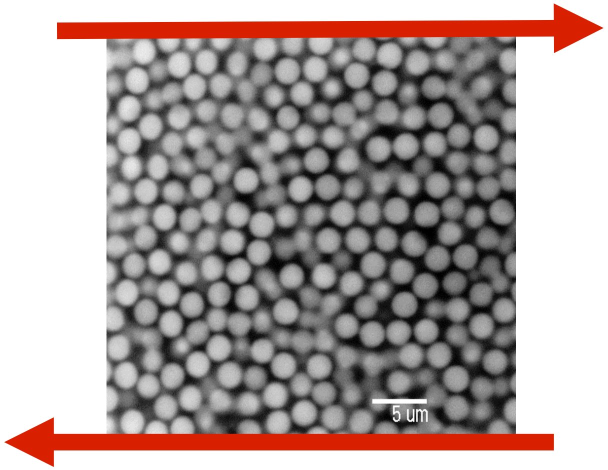


$$\gamma_{meso}(t) = \int_0^t \dot{\gamma}_{meso}(t')dt'$$

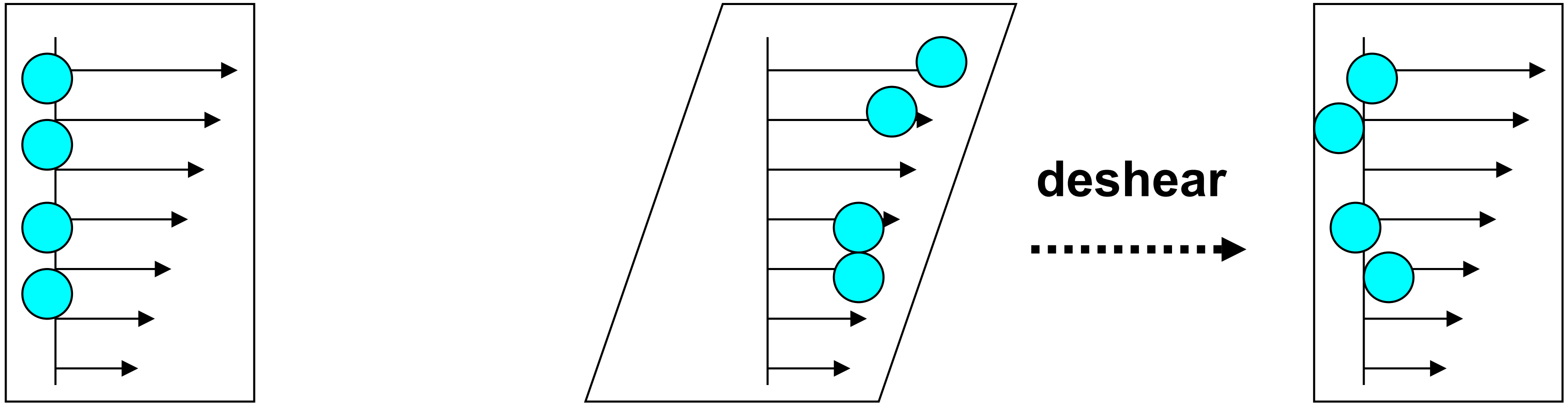
question:

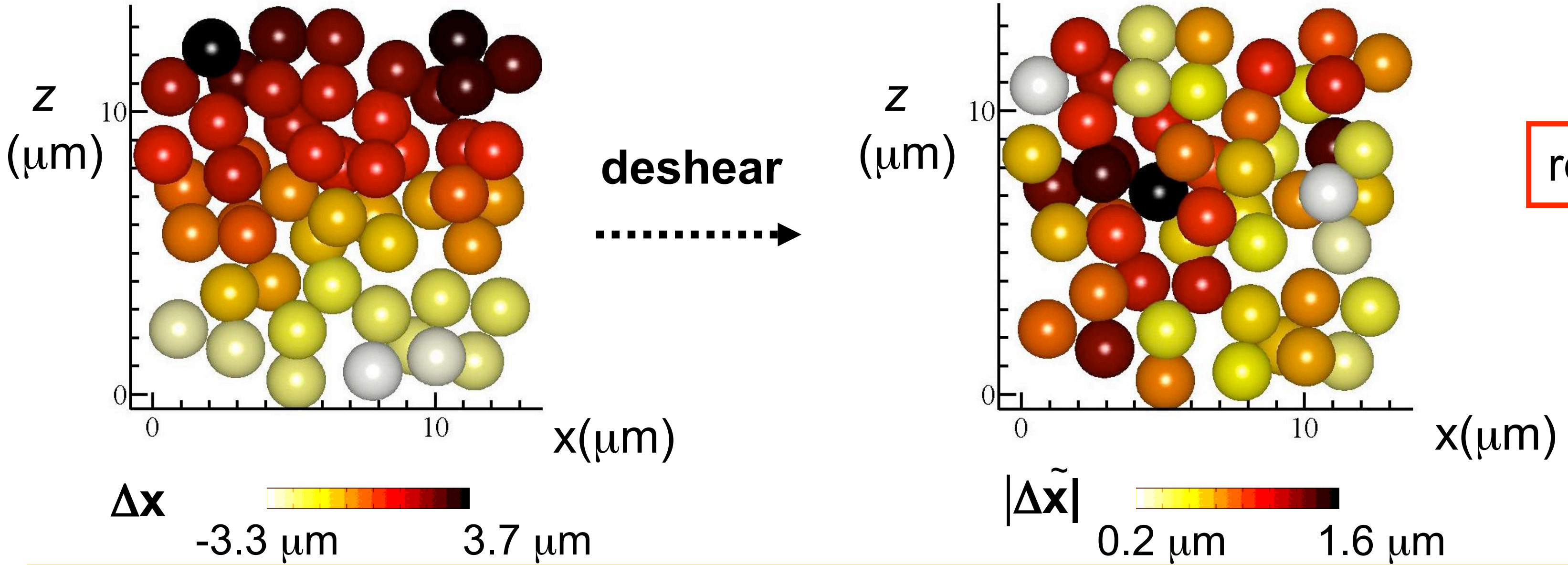
Are shear-induced rearrangements spatially isotropic?





Nonaffine motion: $\Delta\tilde{x}$

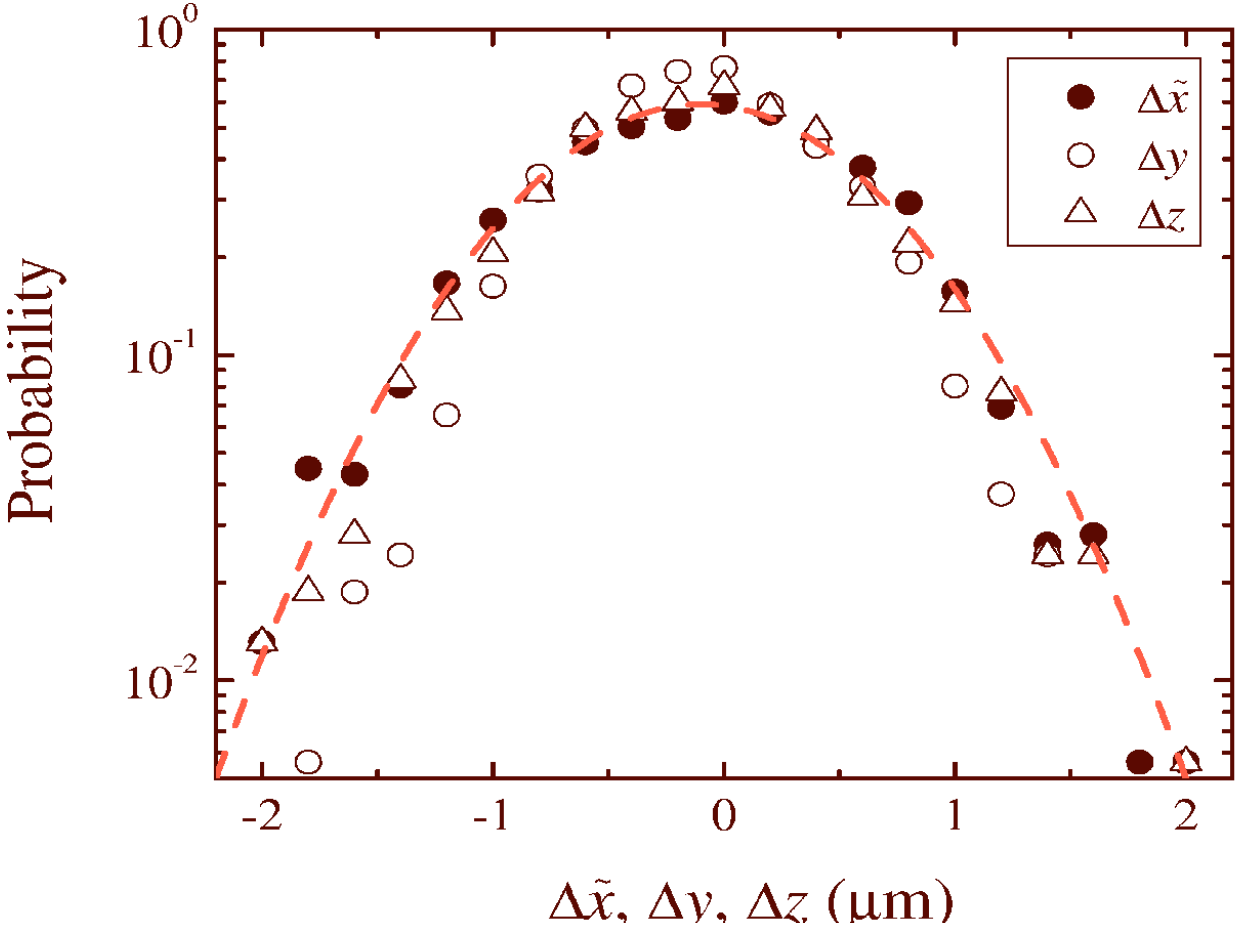


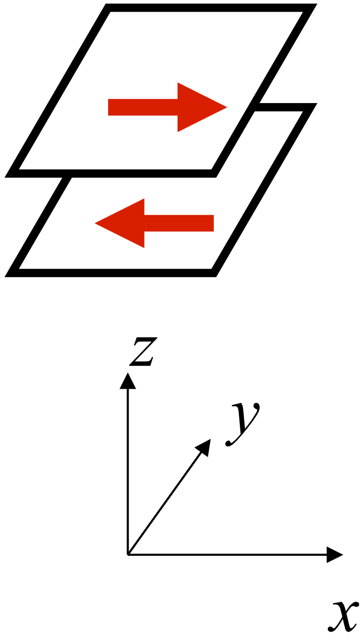


real data

$\phi = 0.51,\ \gamma_{\text{meso}} = 0.43,\ \text{Pe} \approx 20$

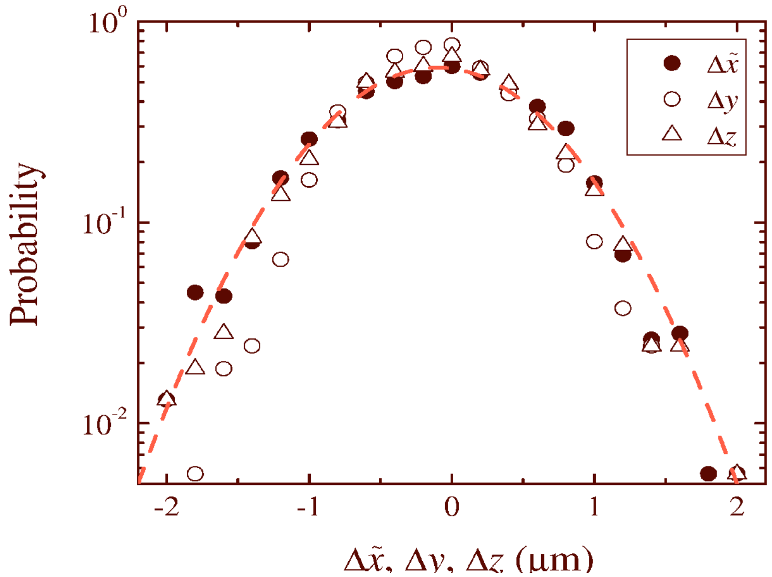
Nonaffine displacements have isotropic distribution



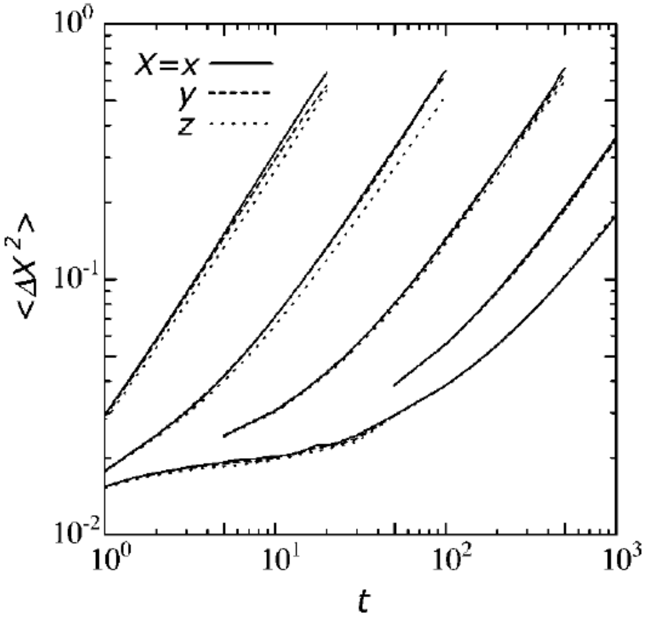


$\phi = 0.51$, $\gamma_{\mathrm{meso}} = 0.43$, Pe ≈ 20

Nonaffine displacements have isotropic distribution



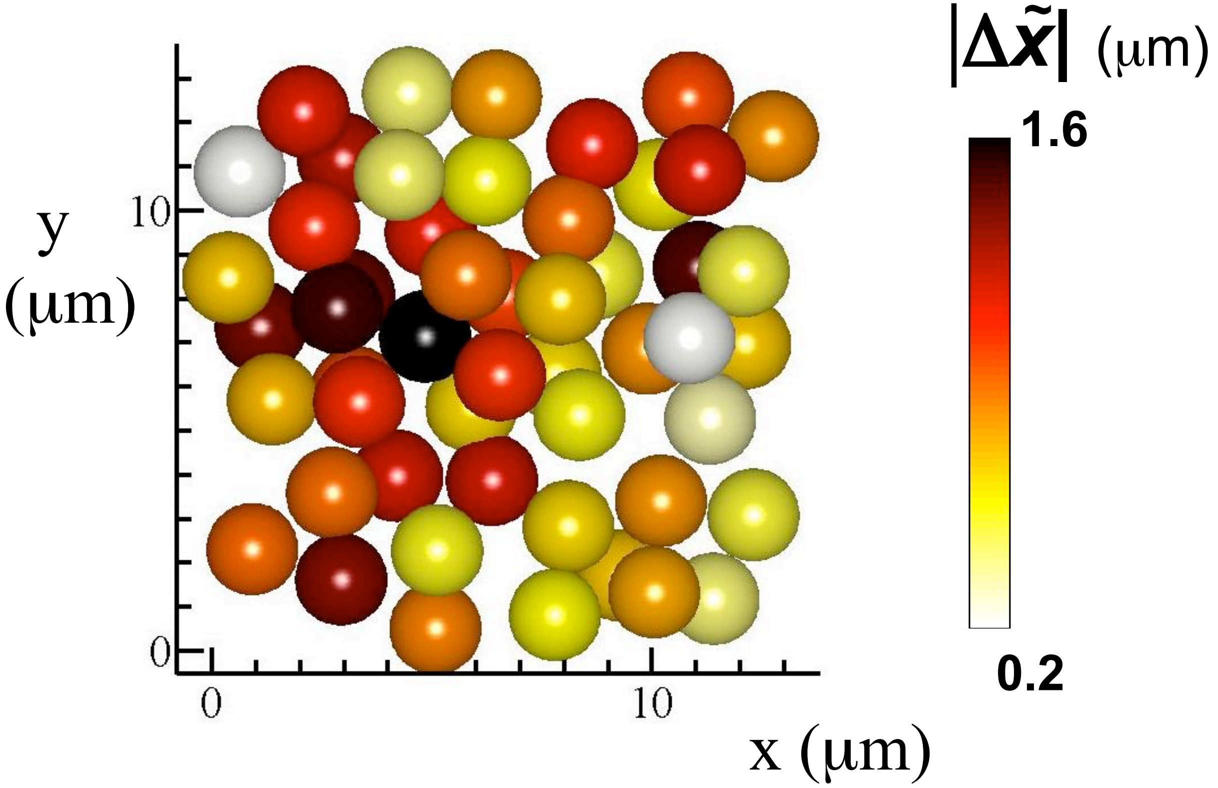
our data

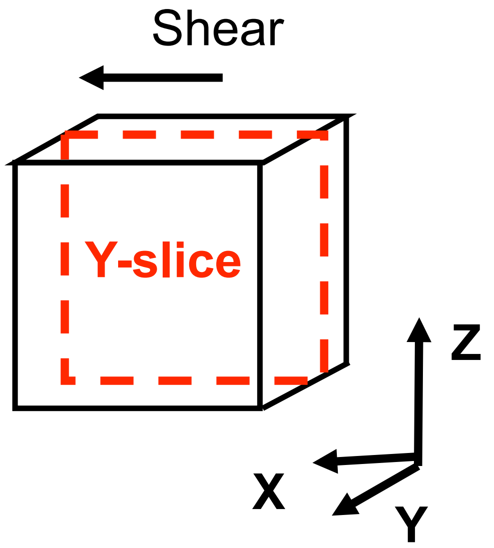


Yamamoto & Onuki simulations
PRE 1998
(also, Miyazaki *et al* PRE 2004)

$\phi = 0.51$, $\gamma_{\text{meso}} = 0.43$, $\text{Pe} \approx 20$

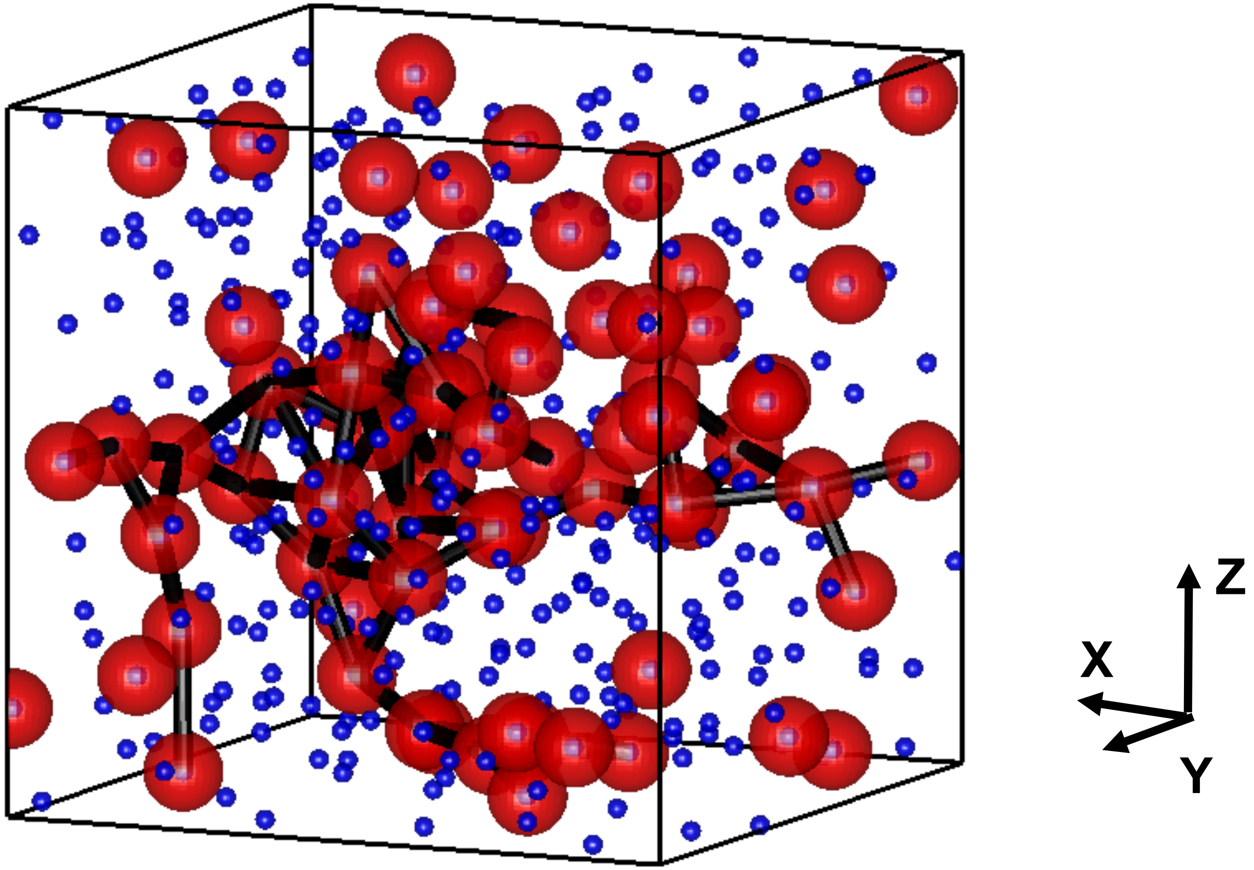
Nonaffine rearrangement is spatially heterogeneous





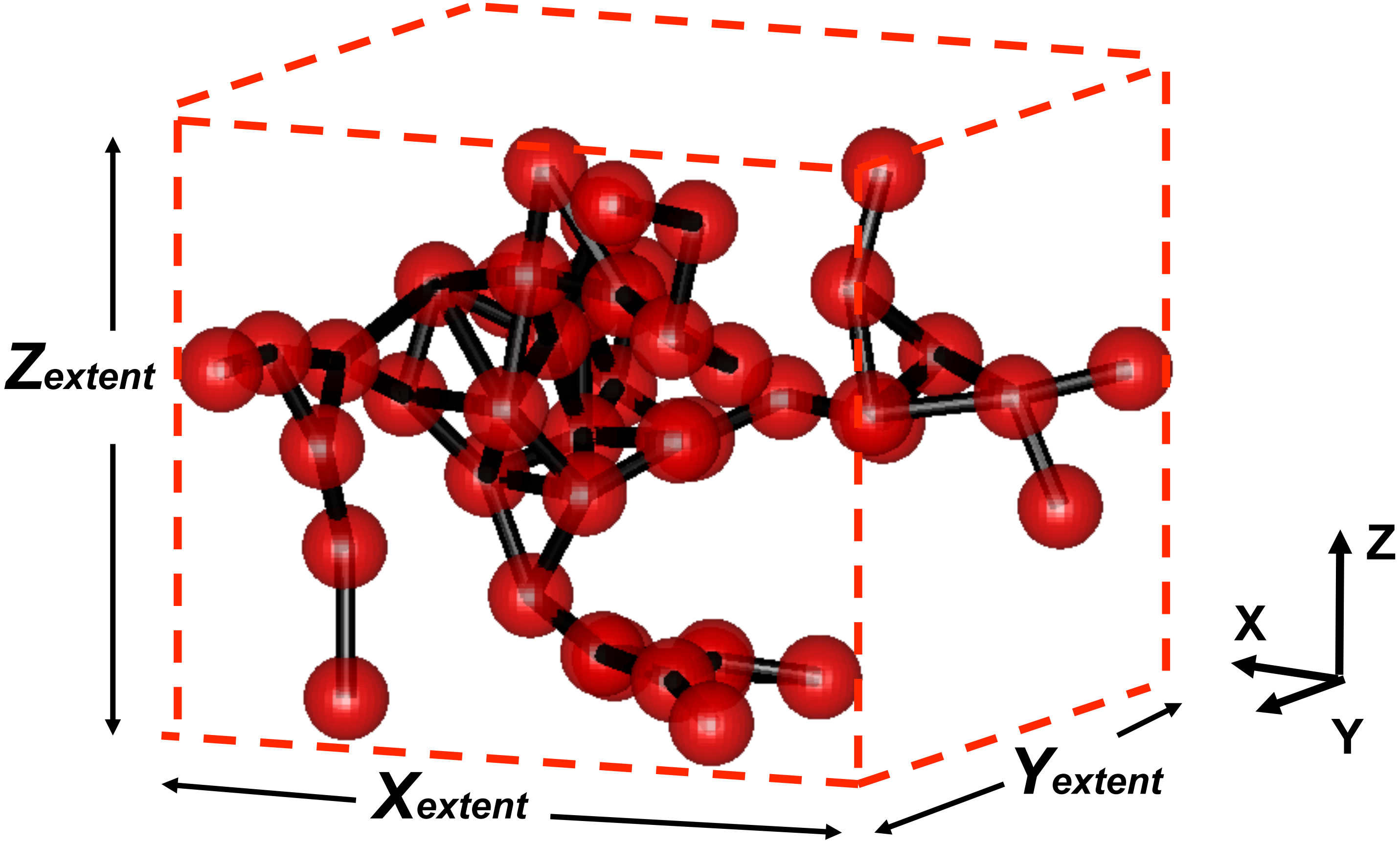
Mobile particles cluster together
Do they spread in a particular direction?

Examine extent of largest highly mobile region



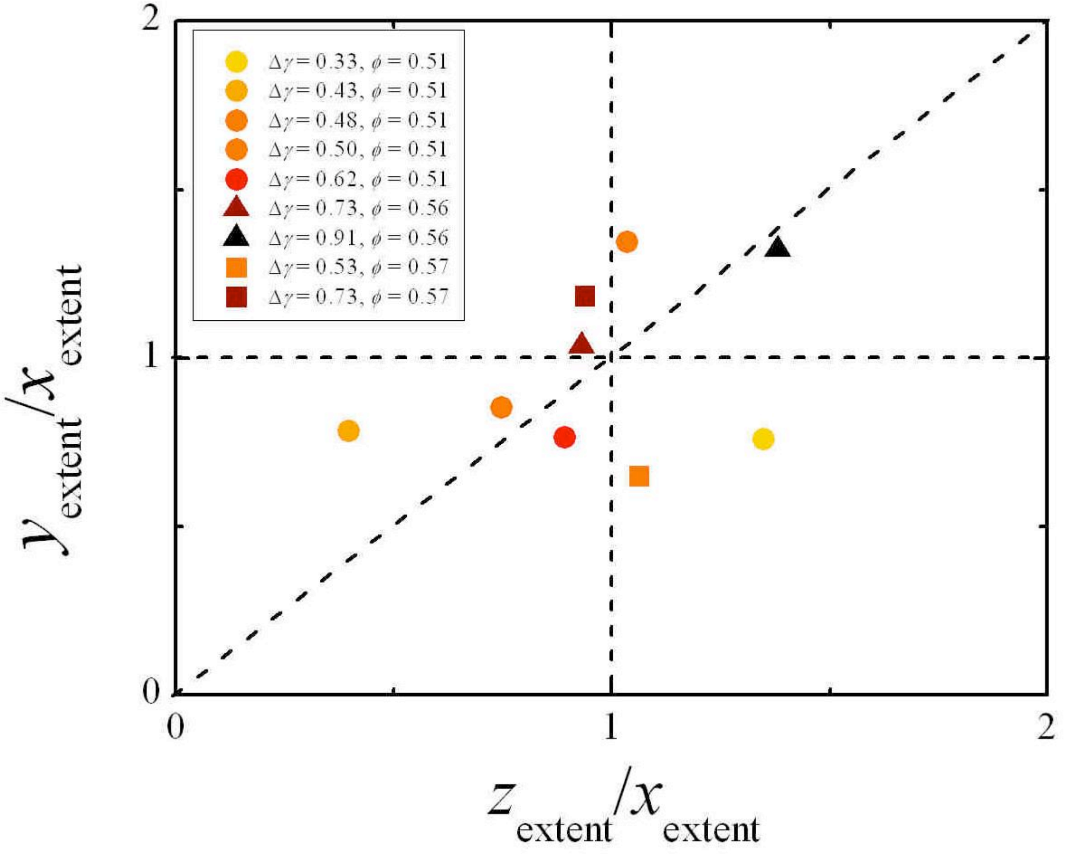
A non-affine mobile cluster: a network of neighboring particles with large non-affine mobility ($\Delta\tilde{r} > 1$ μm).

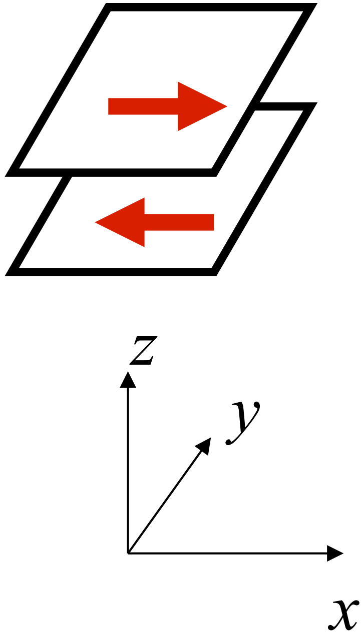
Examine extent of largest highly mobile region



A non-affine mobile cluster: a network of neighboring particles with large non-affine mobility ($\Delta\tilde{r} > 1$ μm).

Mobile clusters have no preferential orientation



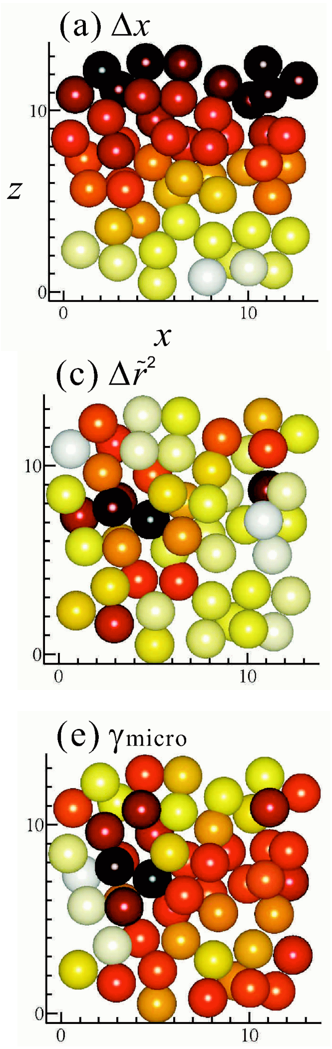


Note 1: Plausible that more subtle analysis would show anisotropy
[4p correlations: A Furukawa, K Kim, S Saito, and H Tanaka, PRL (2009)]

Note 2: We checked other measures of deformation: similar results

Summary part 1 of talk:

- Examined shear of dense monodisperse colloidal suspensions
- Shear results in deformations which are isotropic in several senses
- Length scales of ~ 2 particle diameters



For more details:

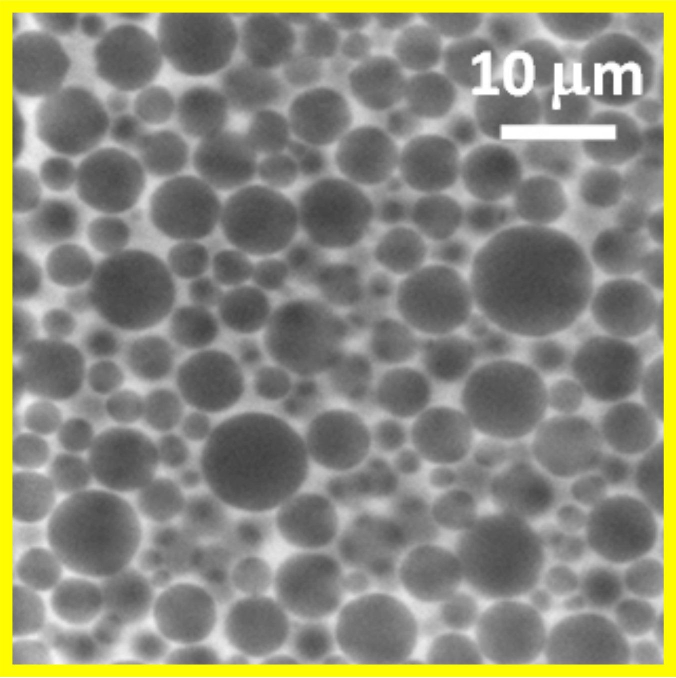
D Chen *et al.*, Phys Rev E **81**, 011403 (2010)

Part 2: Shear of polydisperse emulsions

*Joaquim Clara Rahola, K Feitosa, JC Crocker, ER Weeks

Compared to first part of the talk:

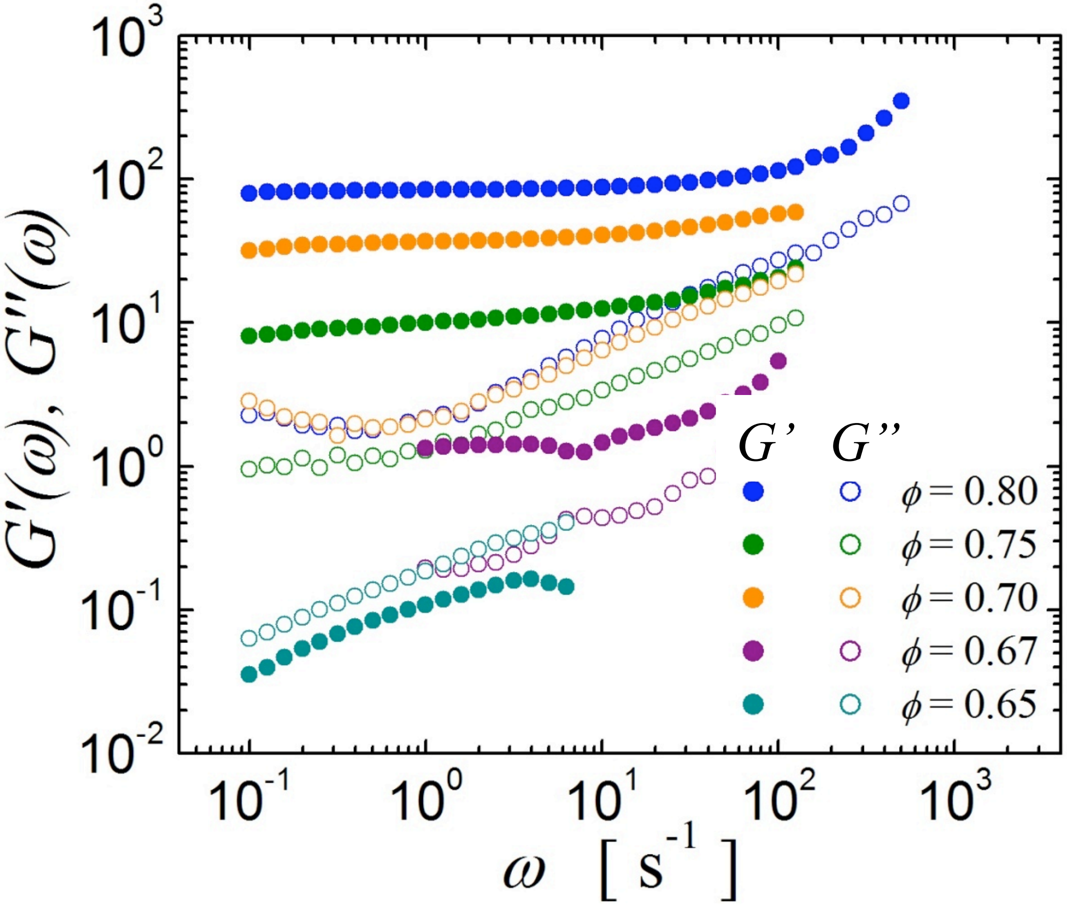
- **Highly polydisperse**
- Droplets are soft
- High volume fractions (jammed samples)
- Smaller strains
- Elastic deformations rather than plastic rearrangements
- Sinusoidal driving rather than triangle wave
- Mostly 2D analysis of 3D samples

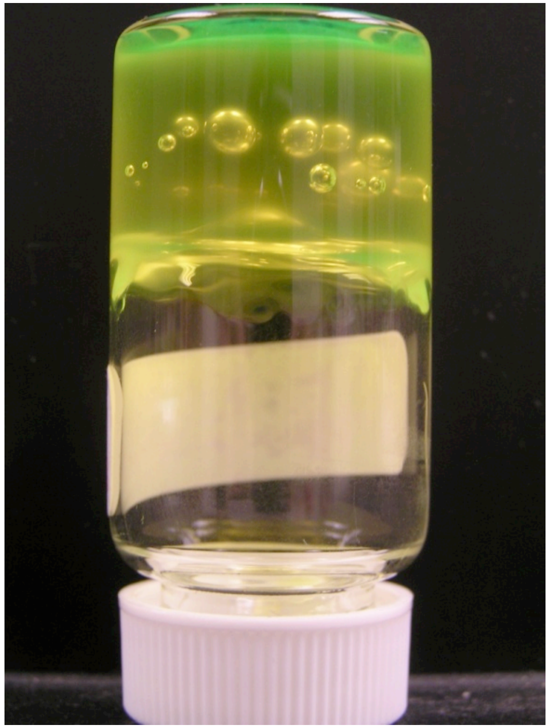


Decane in water/glycerol emulsions (with SDS)

rheology results in collaboration with Rut Besseling & Wilson Poon

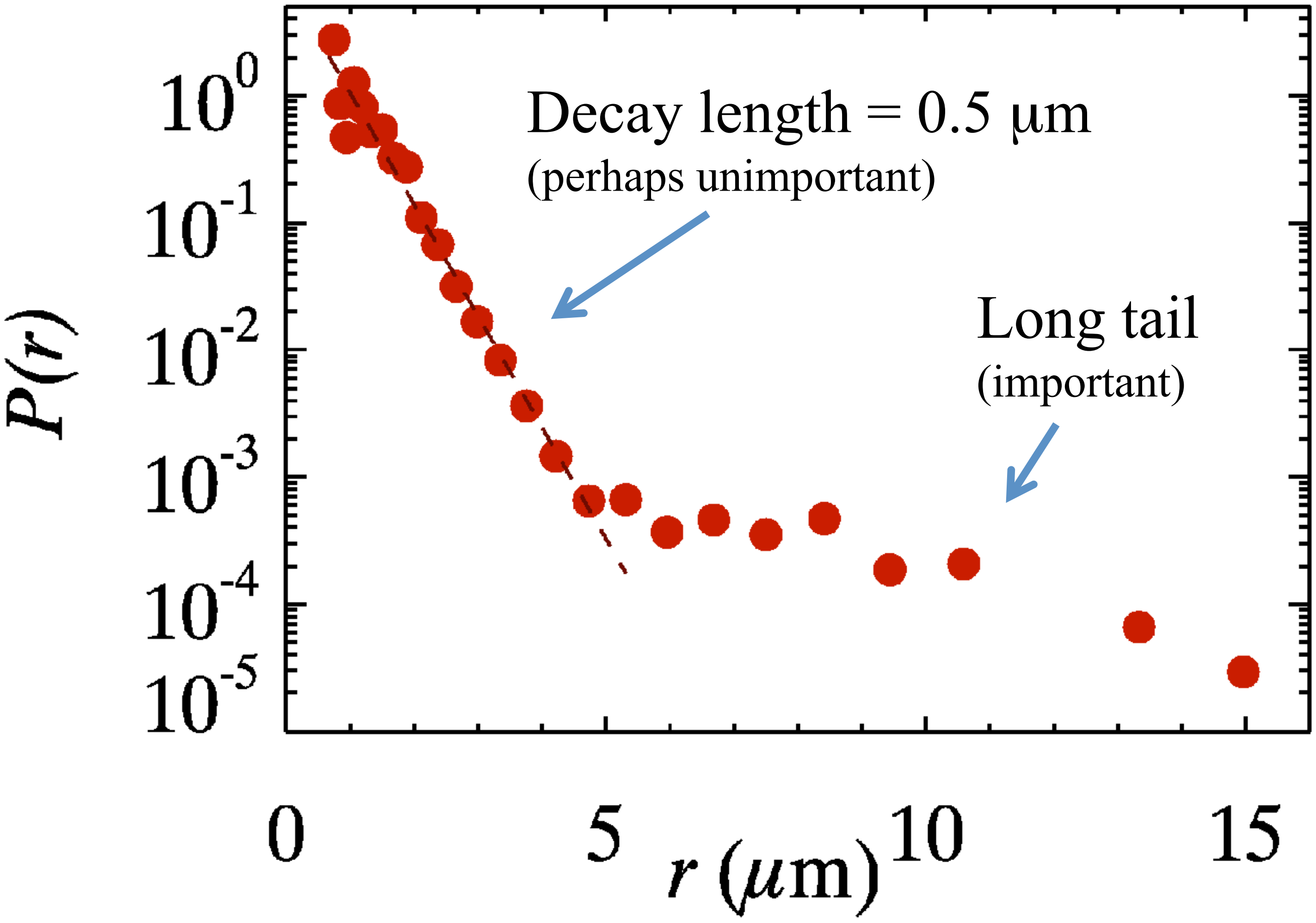
microscopy data to be shown in talk all taken at $f = 1$ Hz, $\omega = 6.3\ \mathrm{s}^{-1}$





Droplet size distribution

(image analysis: K Feitosa, algorithm similar to R Penfold *et al.*, Langmuir 2006)



Decay length = 0.5 μm
(perhaps unimportant)

Long tail
(important)

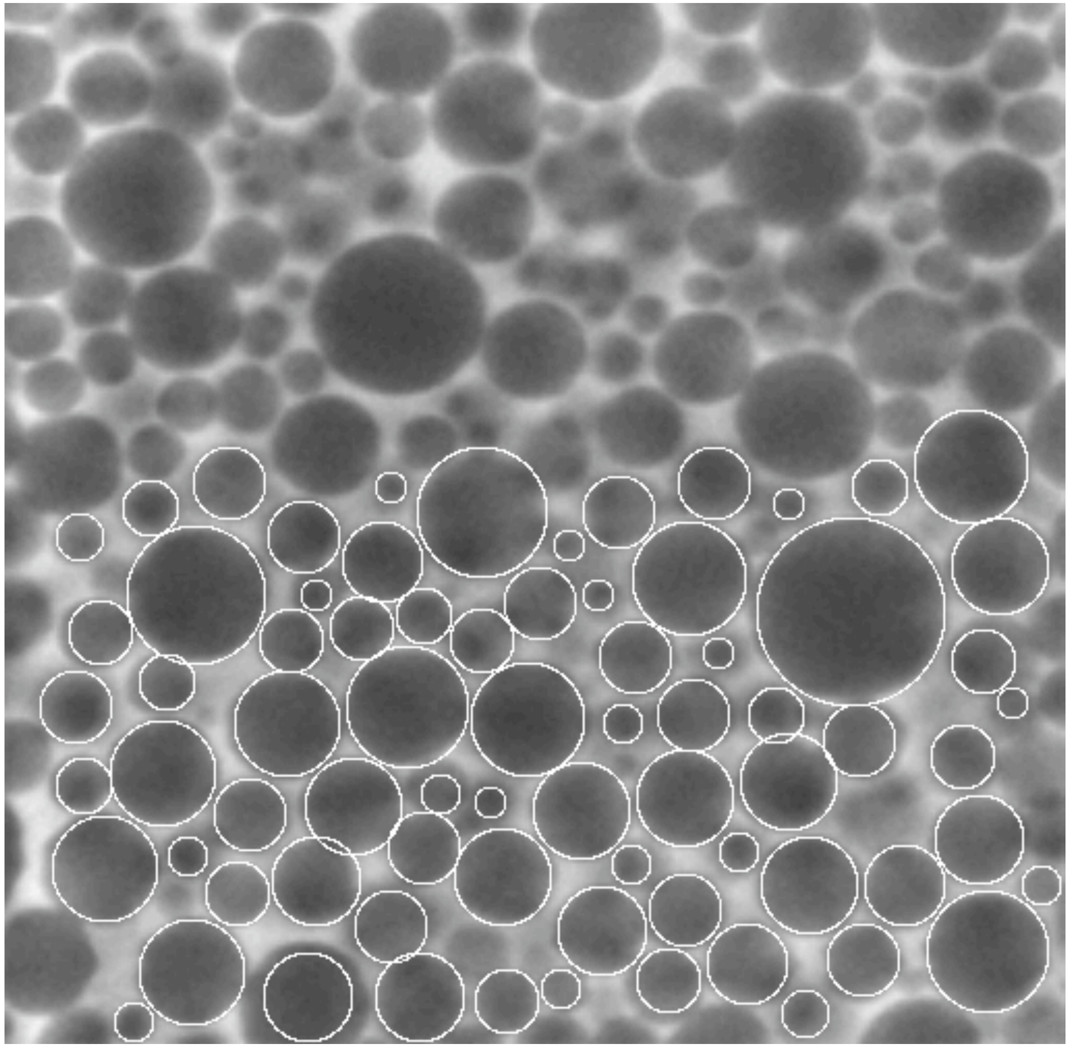
ϕ = 0.80

look at movie…

- 2D movie in real-time
- $\phi = 0.65$
- Driving frequency $f = 1$ Hz
- depth = 12 μm, $\gamma \approx 0.12$

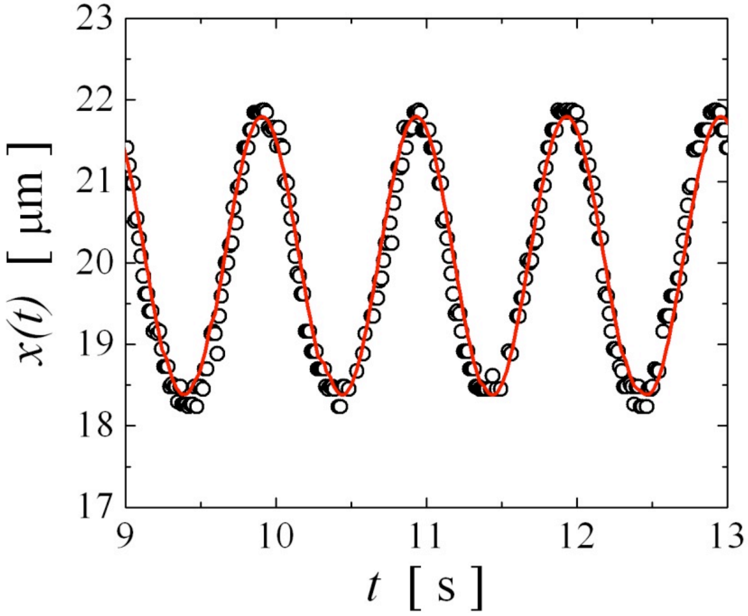
movie “2Vpp-A.tif”, show with ImageJ, \ to start animation

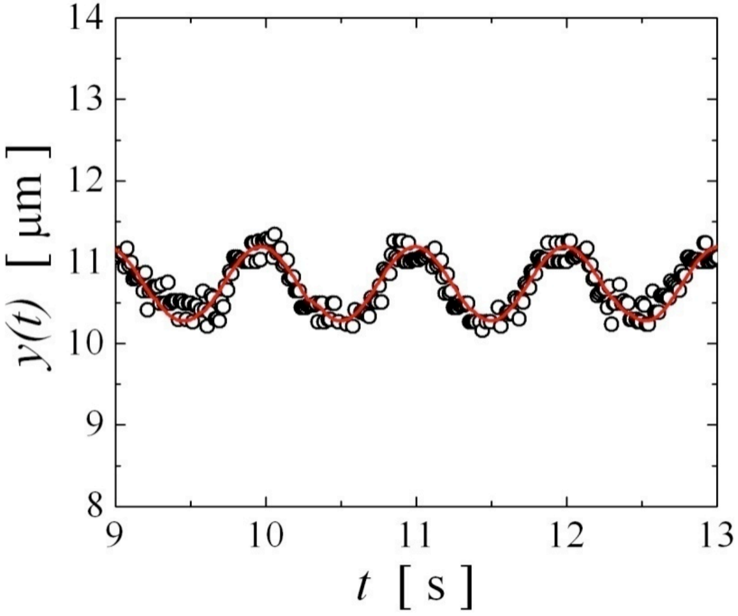
Use Hough transform to identify droplets



Caveat: true radius of droplet might be larger than we observe

Droplet trajectories are sinusoidal



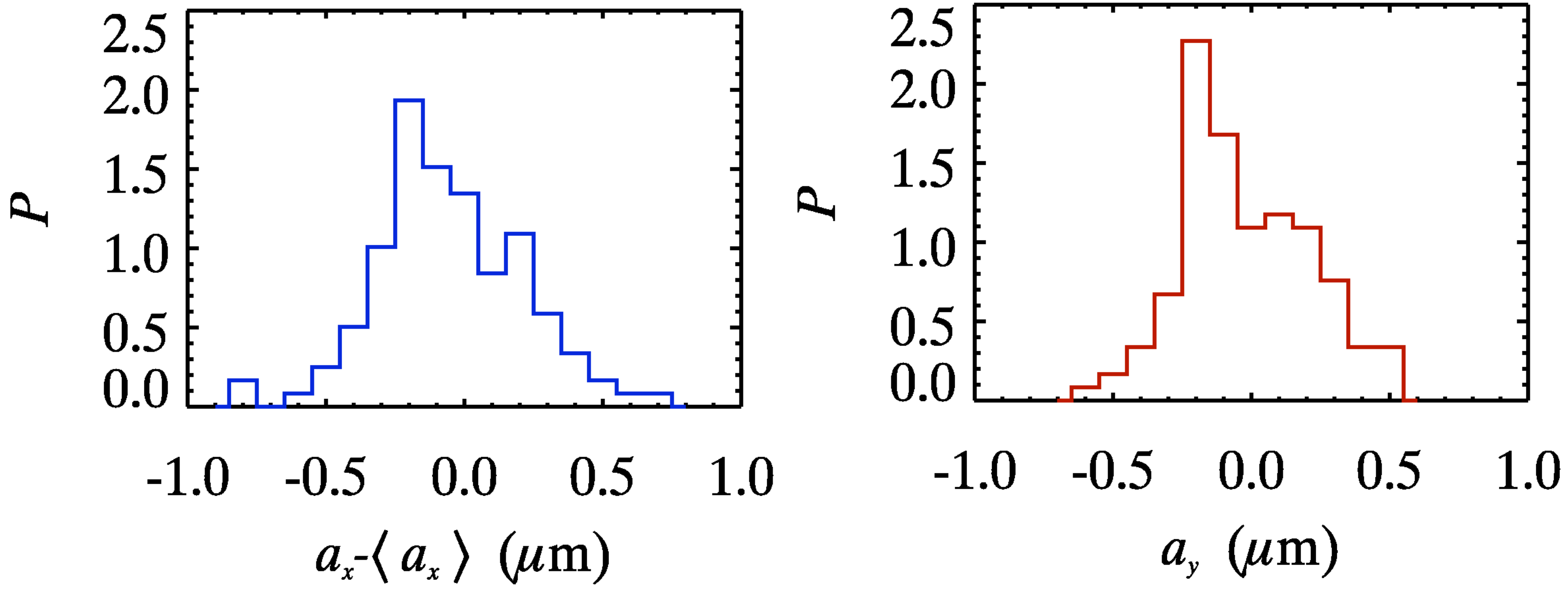


$$x(t) = a_x \sin(\omega t + \theta_x) \qquad y(t) = a_y \sin(\omega t + \theta_y)$$

$\phi = 0.65, \gamma = 0.07$

Amplitude distributions

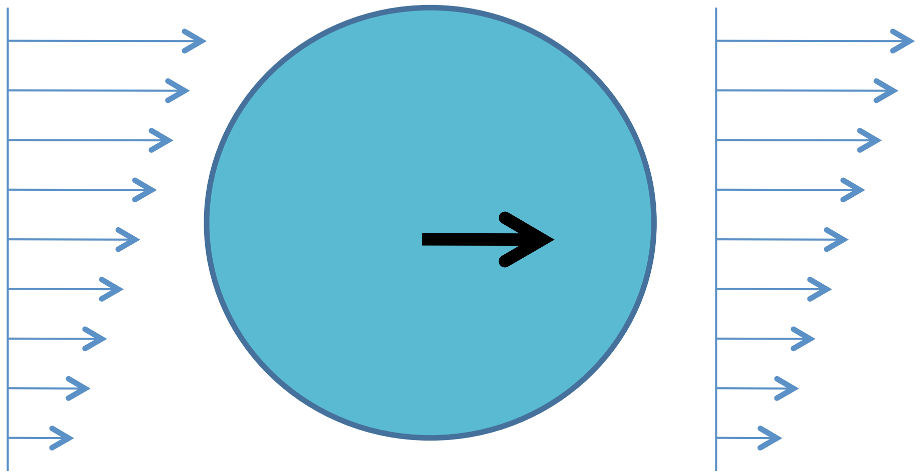
to be shown: small droplets are the outliers



$\langle a_x \rangle = 2.4\ \mu\text{m}$

$\phi = 0.65$, $\gamma = 0.07$, $z = 24\ \mu\text{m}$

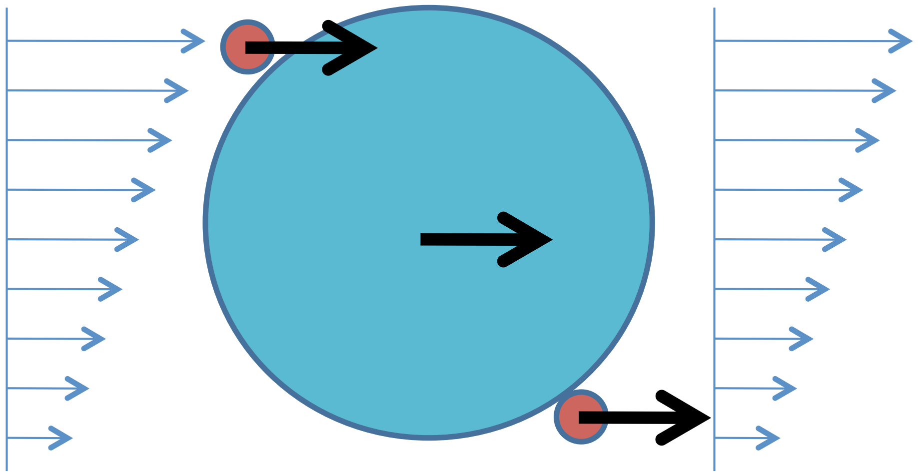
What we think is happening:



Large droplet in otherwise
homogeneous strain field

droplet velocity = mean flow velocity

What we think is happening:

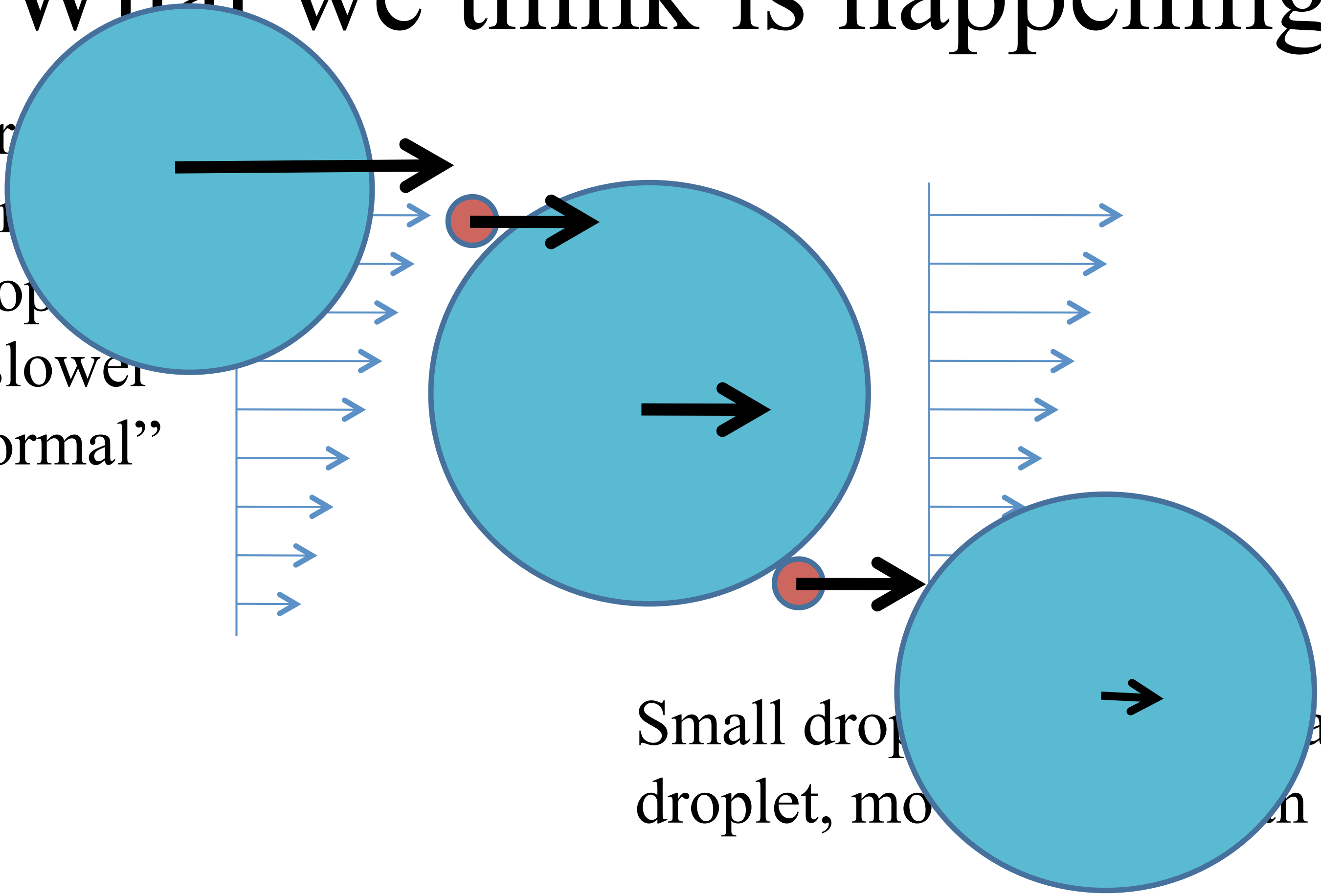


Small droplet constrained by large droplet, moves slower than "normal"

Small droplet pushed by large droplet, moves faster than "normal"

Summary: Smaller droplets pushed around by larger droplets; their "anomalous" motion results in "correct" <u>average</u> flow field around largest droplets

What we think is happening:

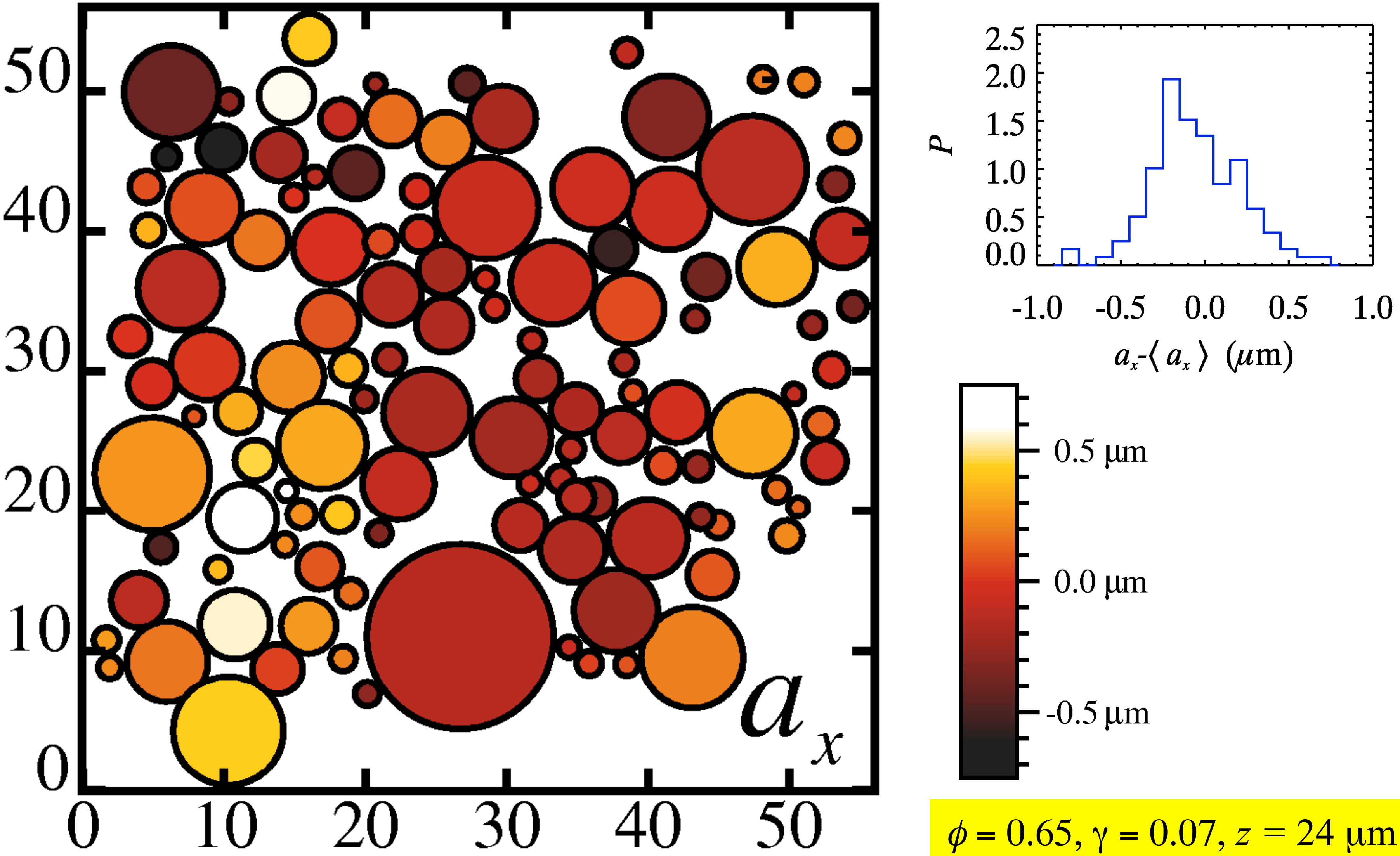


Small dr
constrai
large drop
moves slower
than “normal”

Small drop
arge
droplet, mo
n “normal”

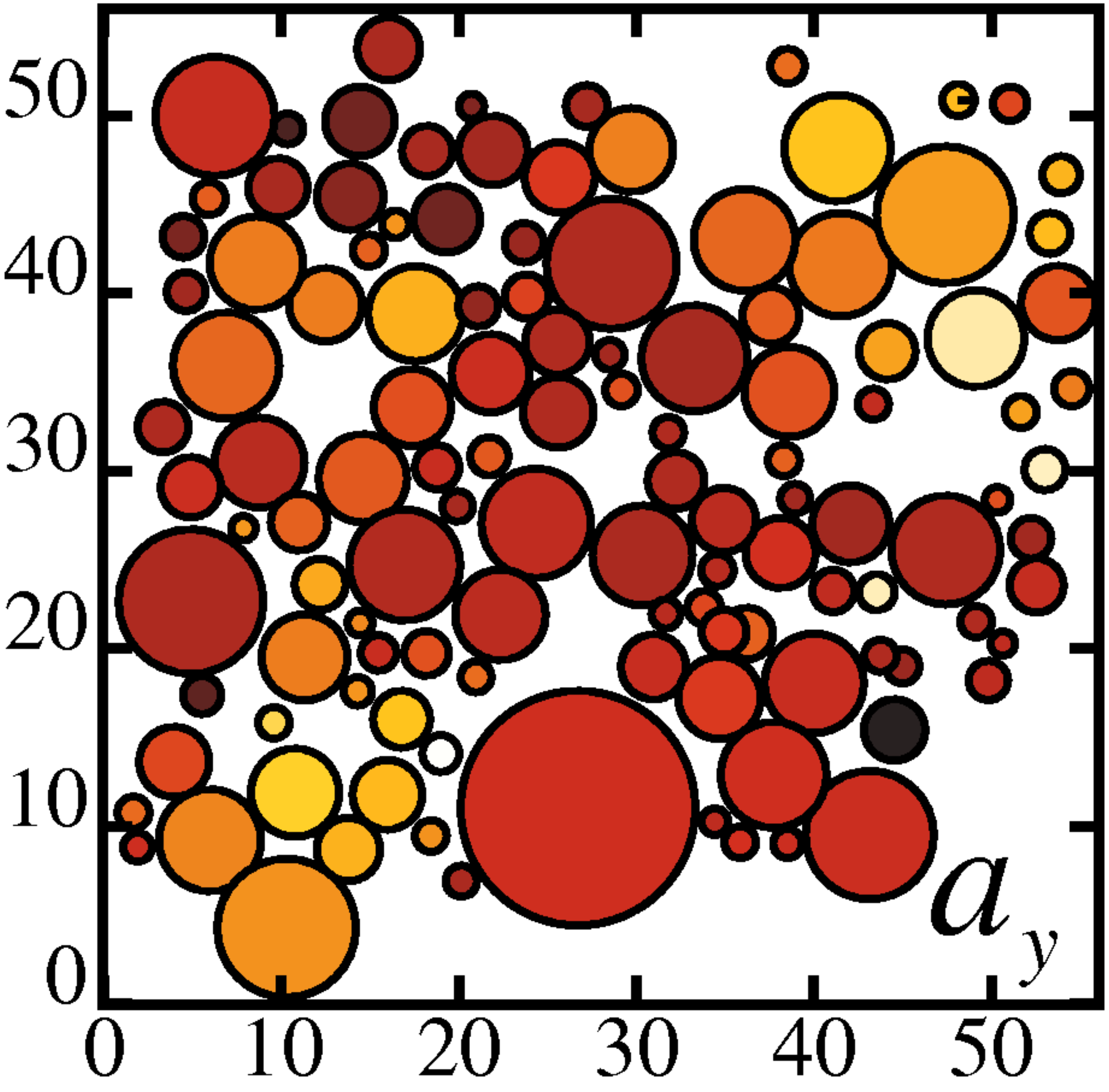
Summary: Smaller droplets pushed around by larger droplets; their “anomalous” motion results in “correct” <u>average</u> flow field around largest droplets

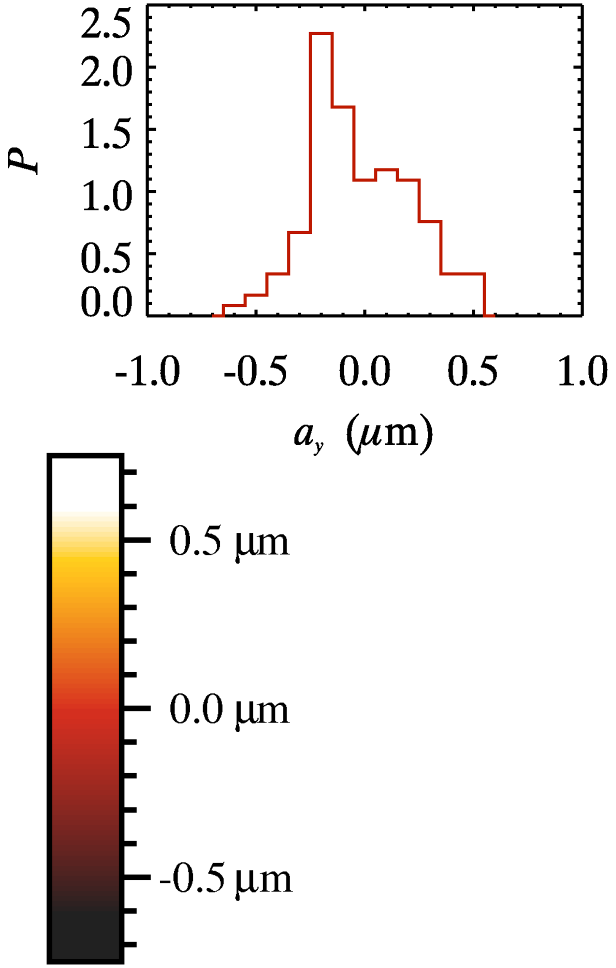
Small droplets are outliers: a_x



$\phi = 0.65$, $\gamma = 0.07$, $z = 24$ μm

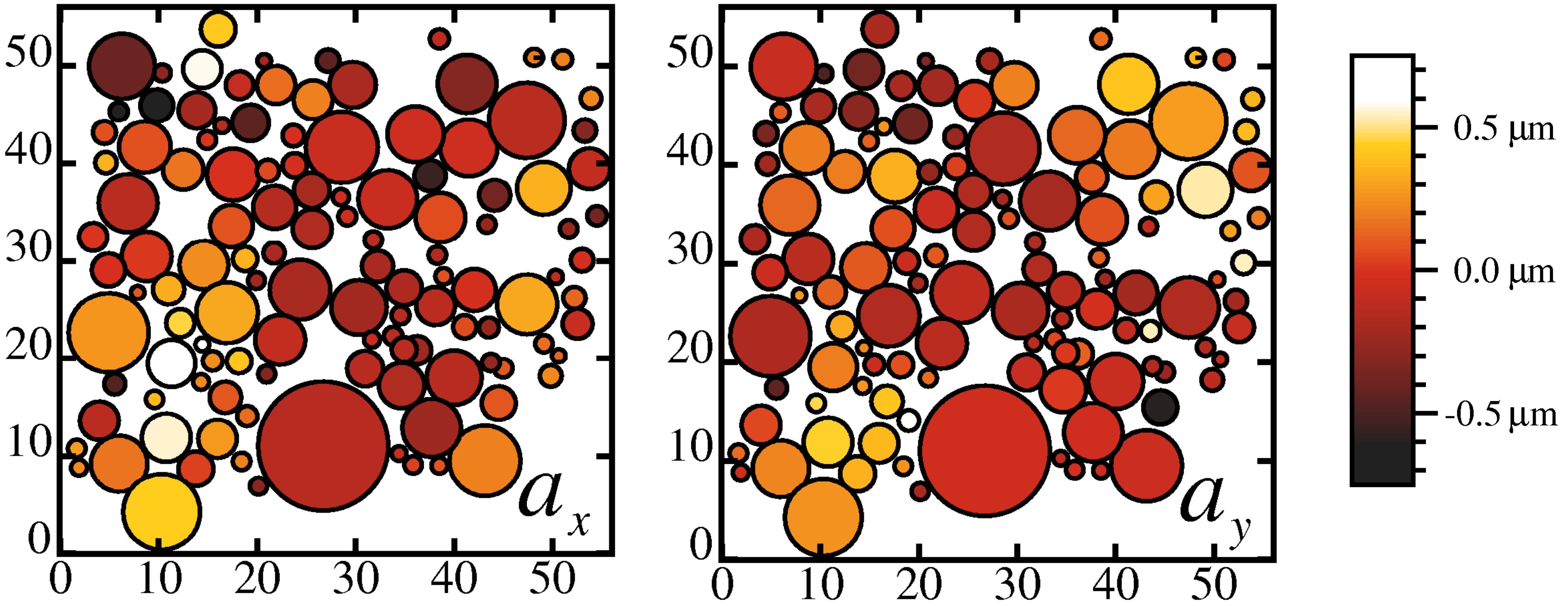
Small droplets are outliers: a_y





$\phi = 0.65$, $\gamma = 0.07$, $z = 24$ μm

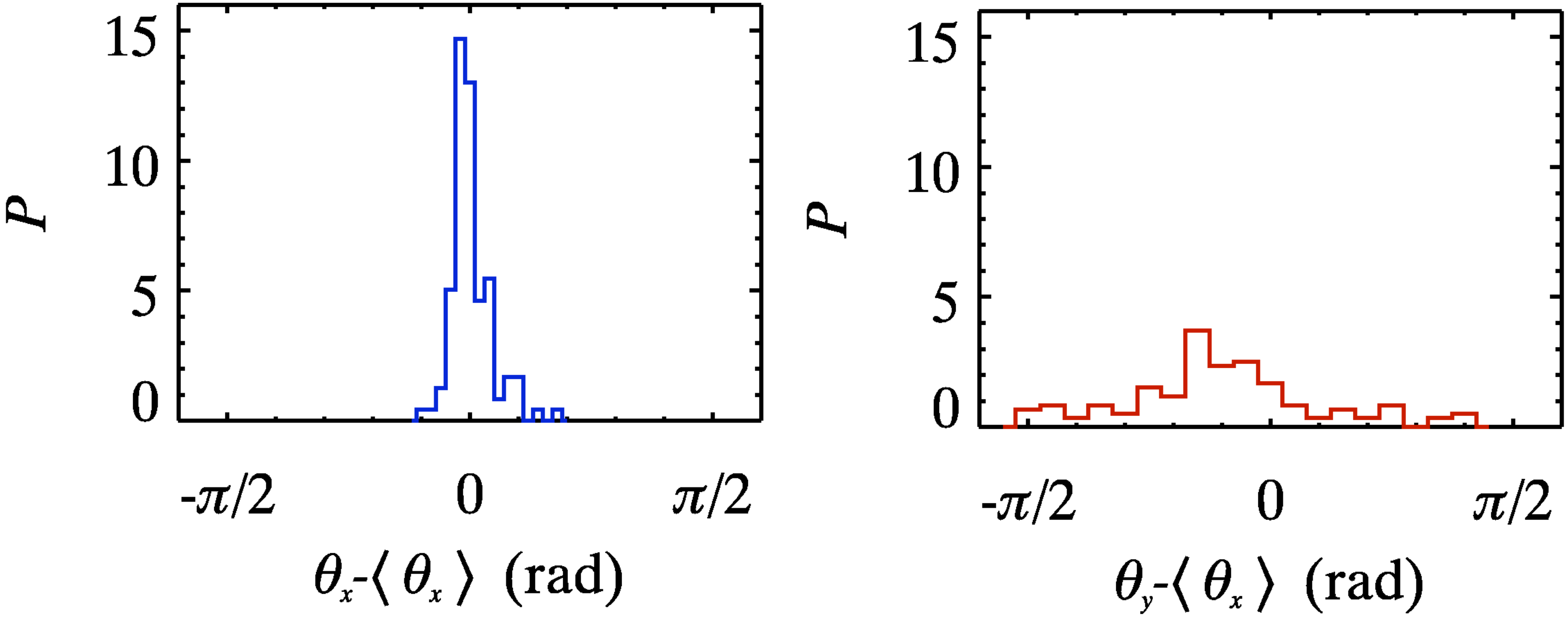
Small droplets are outliers



$\phi = 0.65$, $\gamma = 0.07$, $z = 24$ µm

Phase angle distributions

$$x(t) = a_x \sin(\omega t + \theta_x)$$



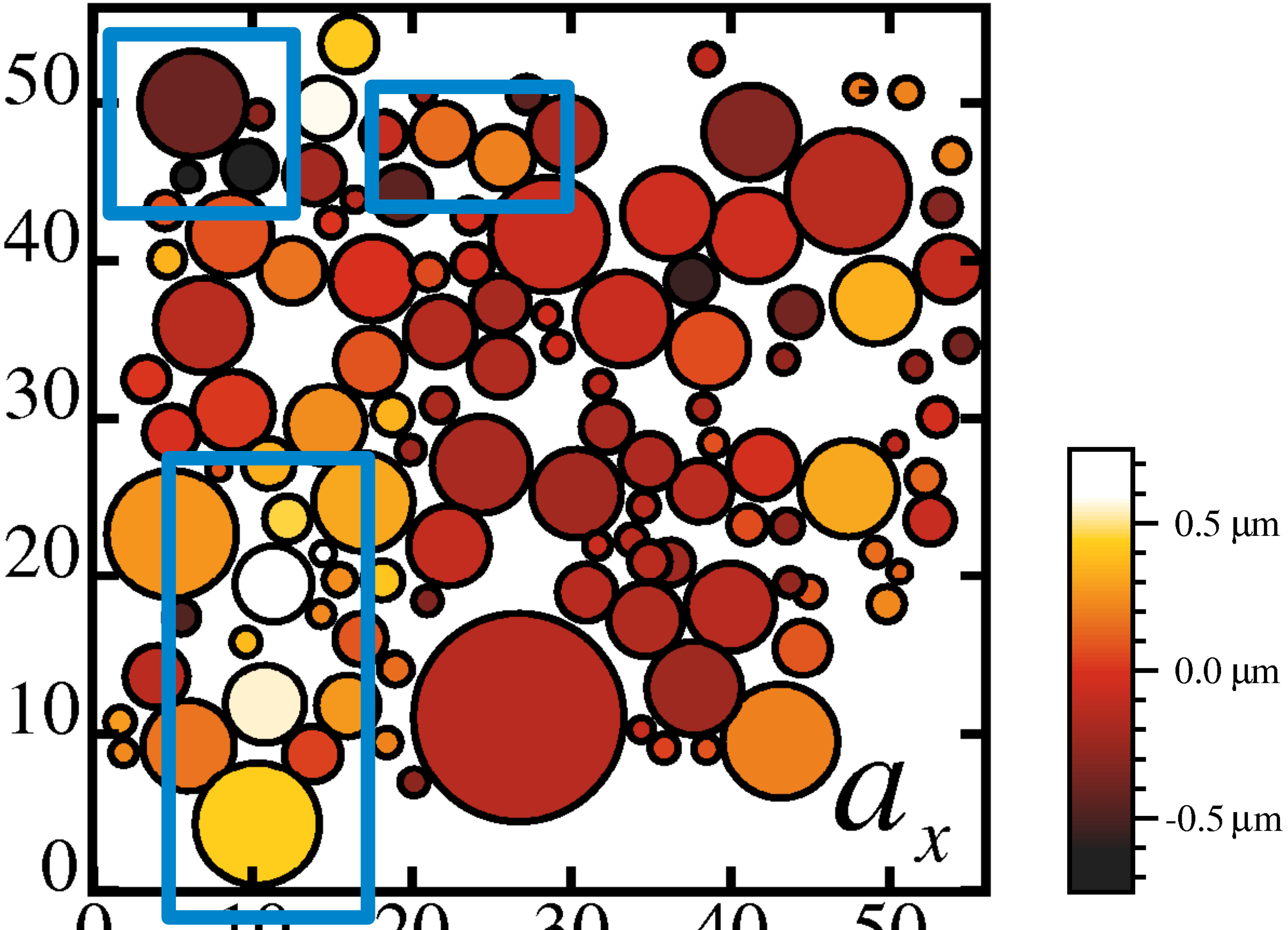
Again, the small droplets are the outliers

Use $\langle \theta_x \rangle$ as reference

$\phi = 0.65$, $\gamma = 0.07$, $z = 24\ \mu\text{m}$

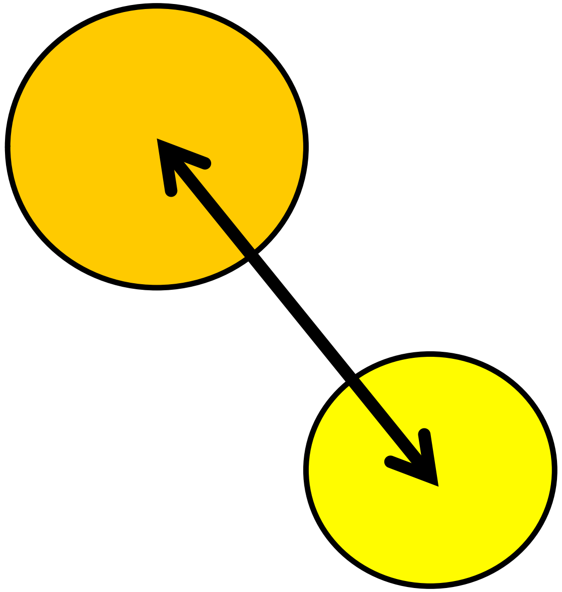
Do neighbors move in similar ways?

answer will be yes…

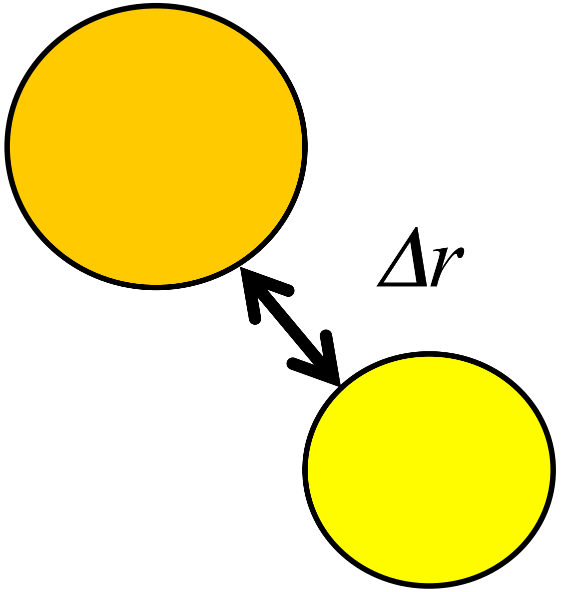


Calculate correlation function

$$C(\Delta r) = \frac{1}{N(\Delta r)} \sum_{\langle i,j \rangle} \frac{(a_{x_i} - \langle a_x \rangle)(a_{x_j} - \langle a_x \rangle)}{\sigma_{a_{x_i}} \sigma_{a_{x_j}}}$$

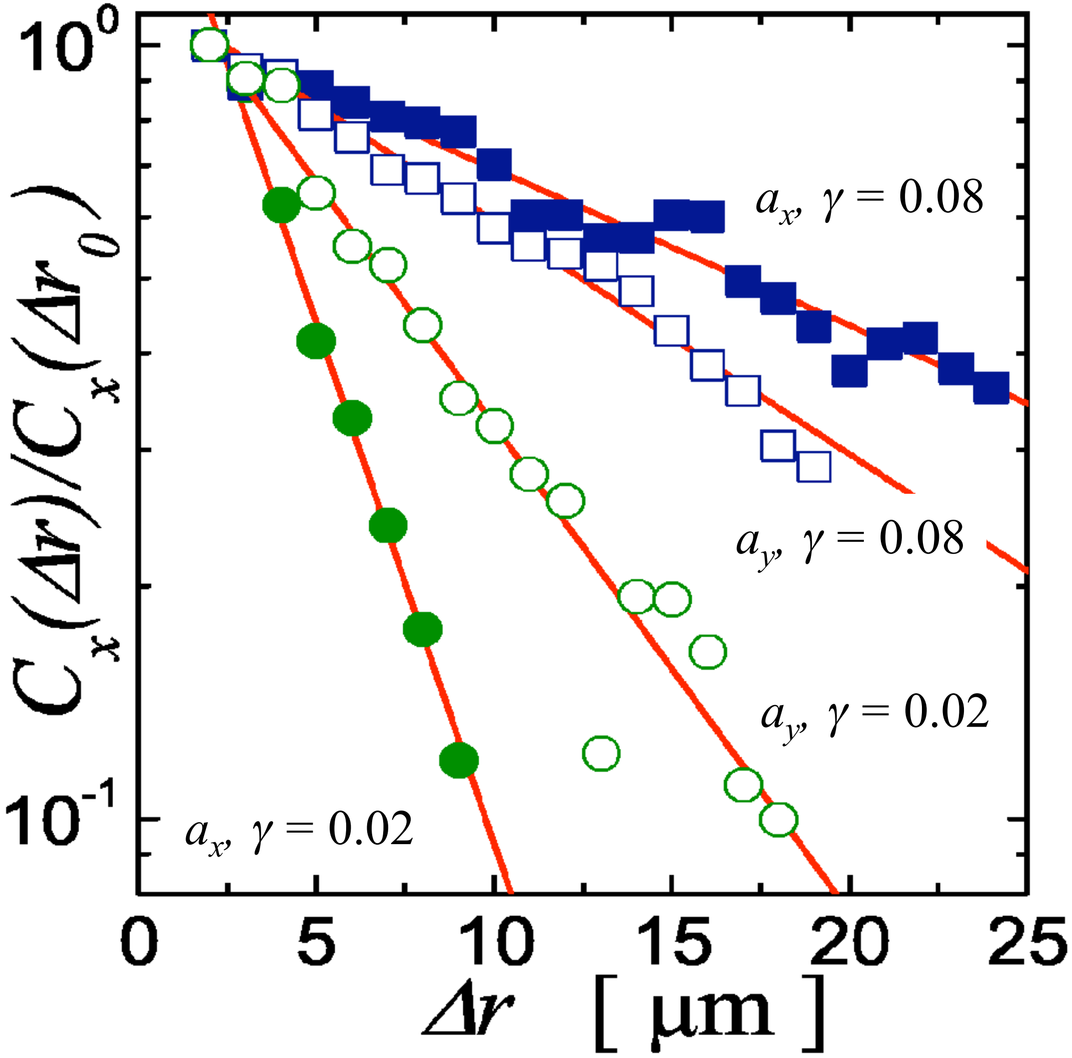


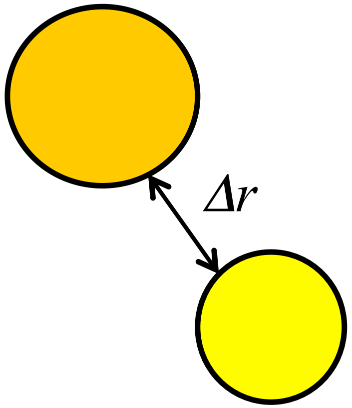
Δr center-to-center: NO!



Δr surface-to-surface: YES!

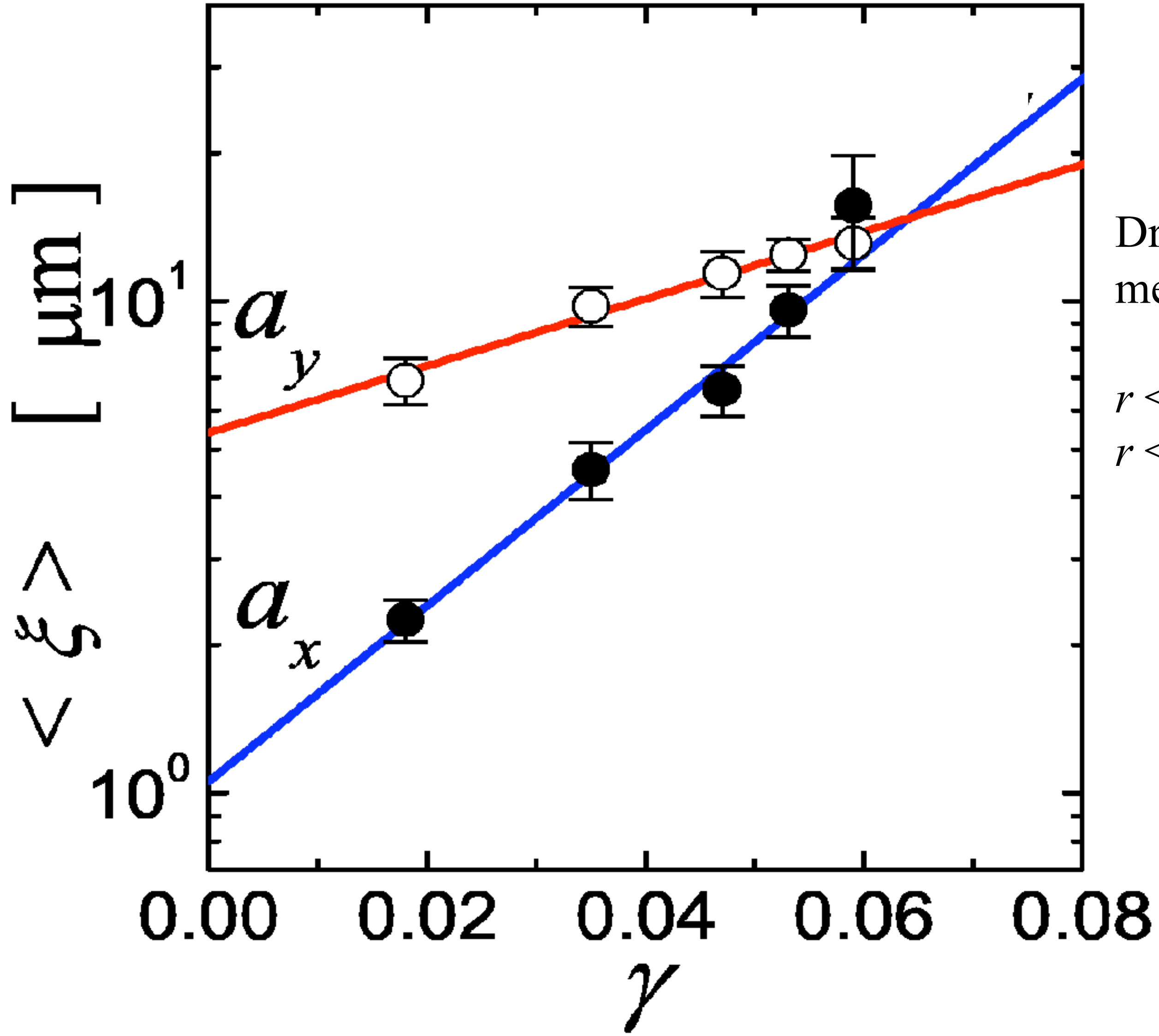
Correlations have exponential decay





$\phi = 0.65$, $z = 24$ μm

Decay length increases with increasing strain



Droplet sizes:
mean r = 1.2 μm

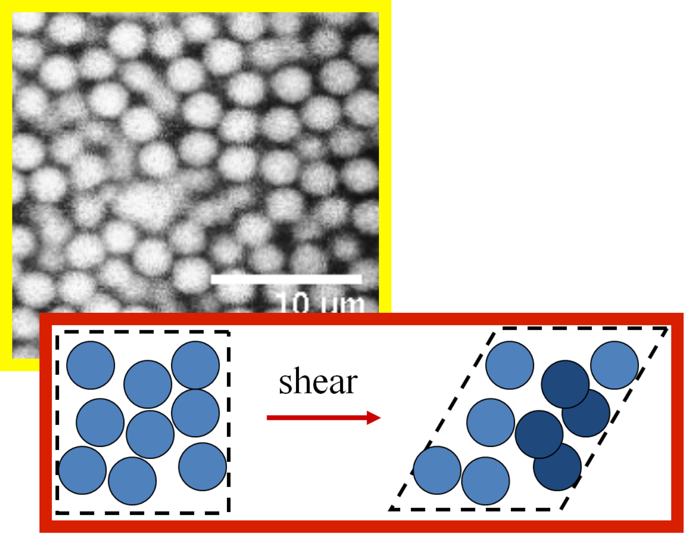
r < 2.6 μm = 50% of volume
r < 10 μm = 85% of volume

$\phi = 0.65$, z = 24 μm

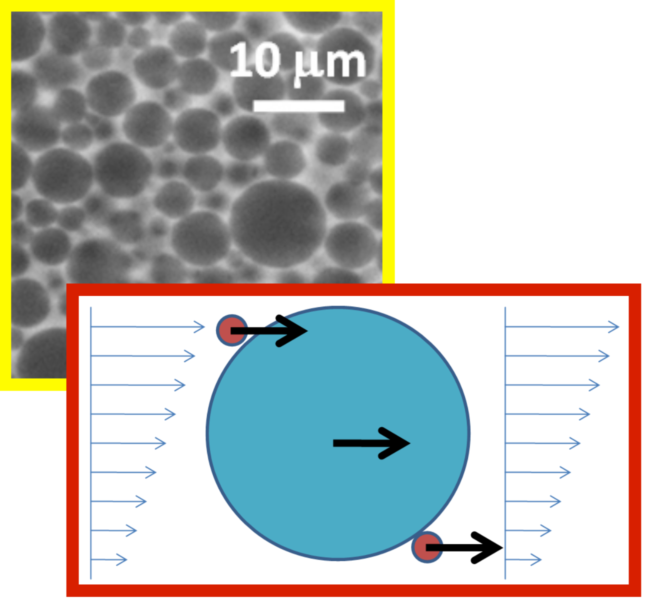
Summary

Shear of jammed soft materials:

- Monodisperse – see locally deforming regions, isotropic, results depend primarily on total accumulated strain



- Polydisperse – see extremely non-affine response of smaller droplets, correlation length that grows with shear rate



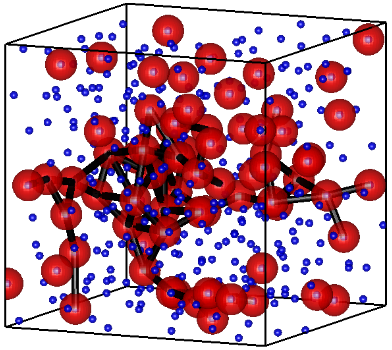
colloids: Chen et al., PRE **81**, 011403 (2010)
emulsions: Clara Rahola et al., hopefully on arXiv soon

Movies, reprints, & free particle tracking software:
www.physics.emory.edu/~weeks/lab/

Extra slides

Find length scale for rearrangements: correlation functions

based on Doliwa & Heuer, PRE (2000);
ER Weeks, JC Crocker, DA Weitz, JP:CM (2007)

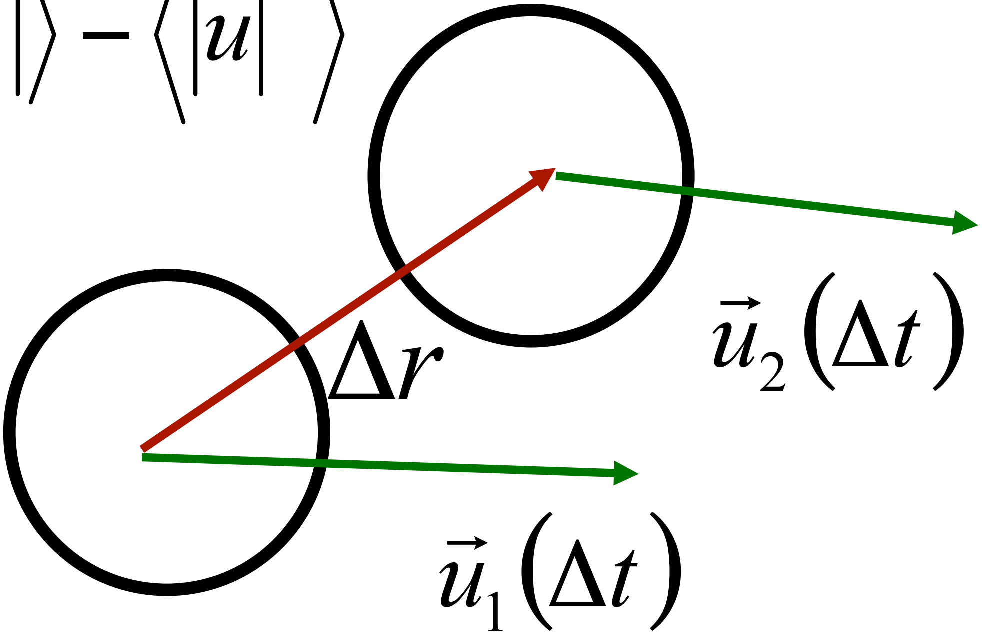


- Vector correlation function:

$$S_{\vec{u}}(\Delta r, \Delta t) \approx \left\langle \vec{u}_1 \cdot \vec{u}_2 \right\rangle$$

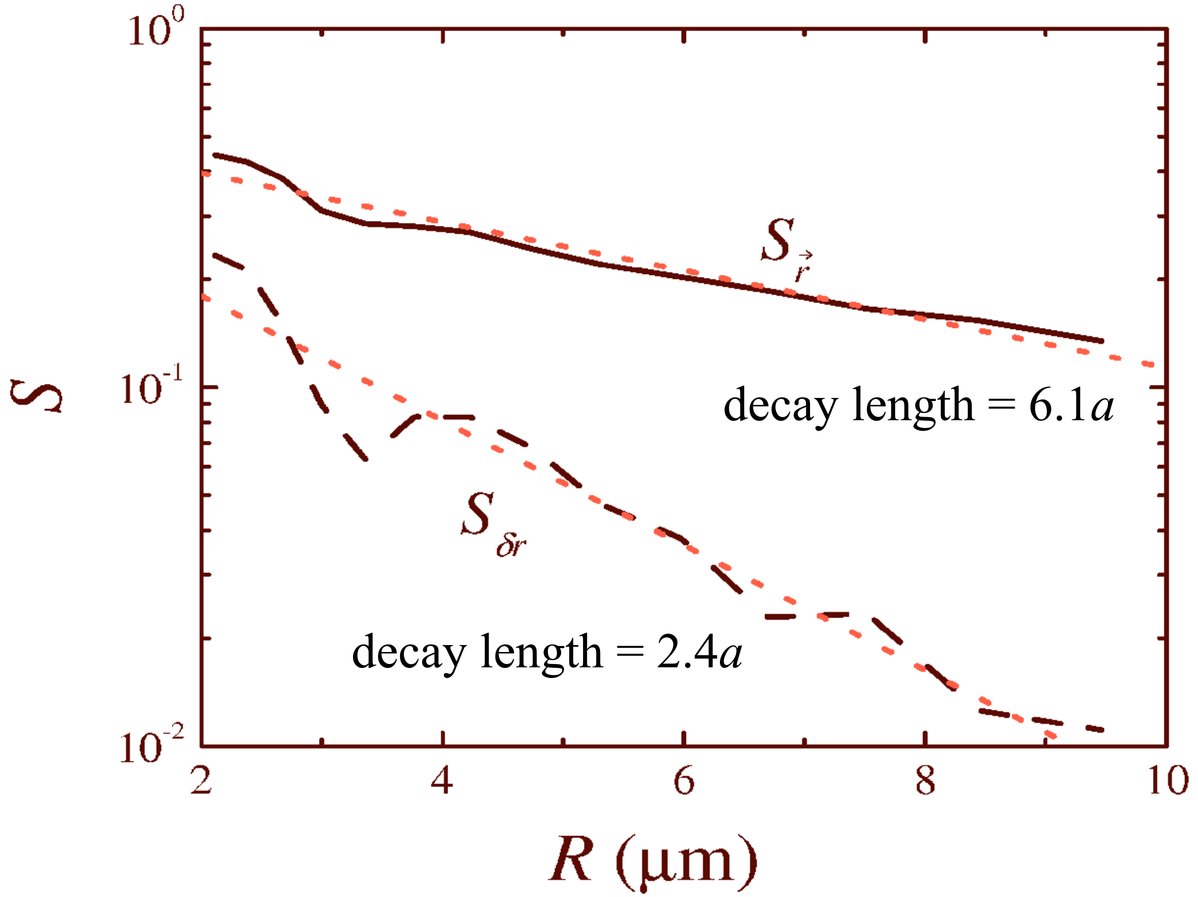
- Scalar correlation function:

$$S_{\delta u}(\Delta r, \Delta t) \approx \left\langle |u_1| \cdot |u_2| \right\rangle - \left\langle |u|^2 \right\rangle$$



- use nonaffine displacements
- use $\Delta t_{\max}$

Correlations decay exponentially in space

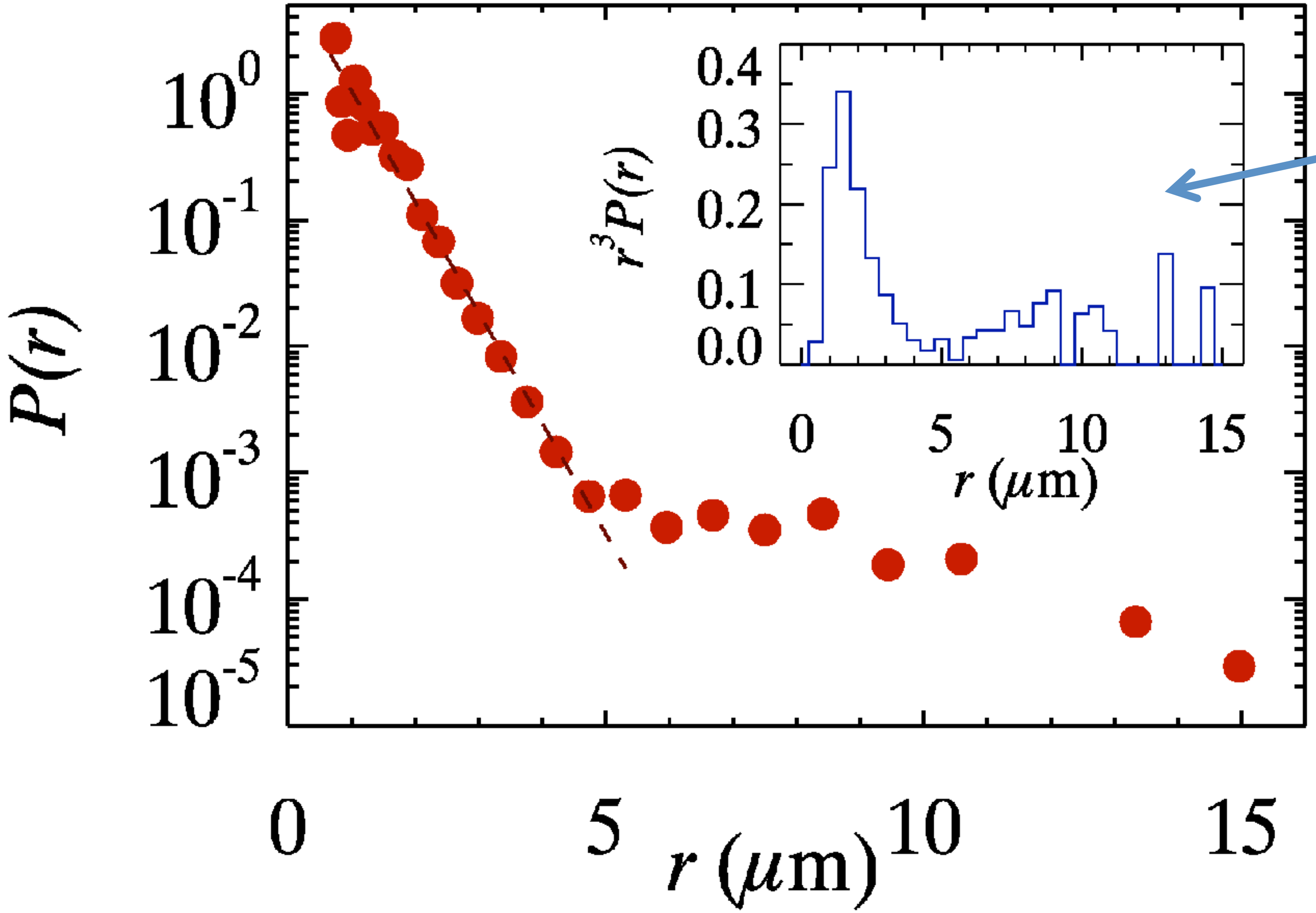


Results shown are typical; no clear dependence on ϕ or $\dot{\gamma}$ seen

$\phi = 0.51$, $\gamma_{\text{meso}} = 0.43$, Pe ≈ 20

Droplet size distribution

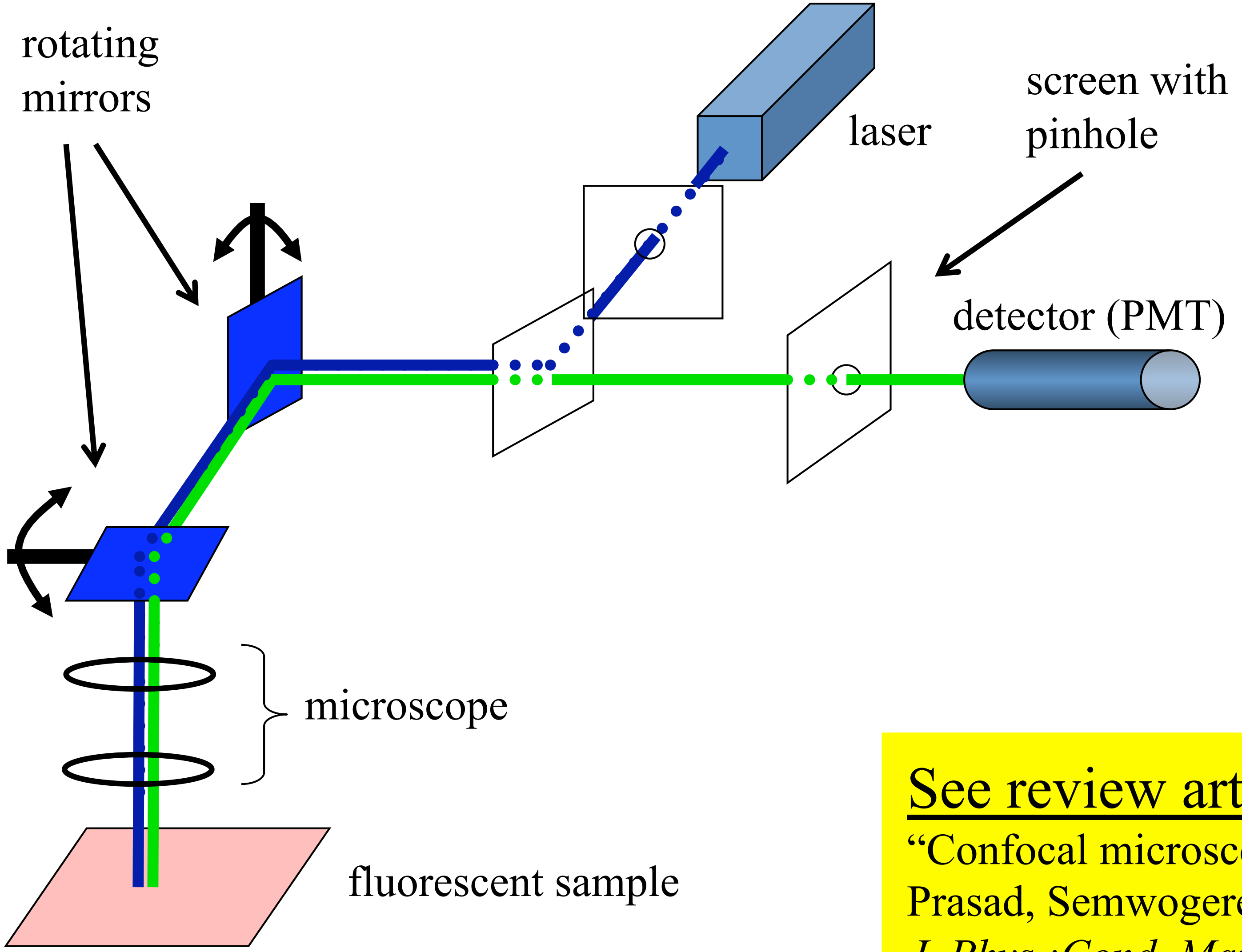
(image analysis: K Feitosa, algorithm similar to R Penfold *et al.*, Langmuir 2006)



Volume weighted distribution

$\phi = 0.80$

Confocal Microscopy



rotating
mirrors

laser

screen with
pinhole

detector (PMT)

microscope

fluorescent sample

See review article:

"Confocal microscopy of colloids"
Prasad, Semwogerere, & Weeks
J. Phys.:Cond. Mat. **19**, 113102 (2007)