Colloidal surface roughness, shear thickening, and the glass transition

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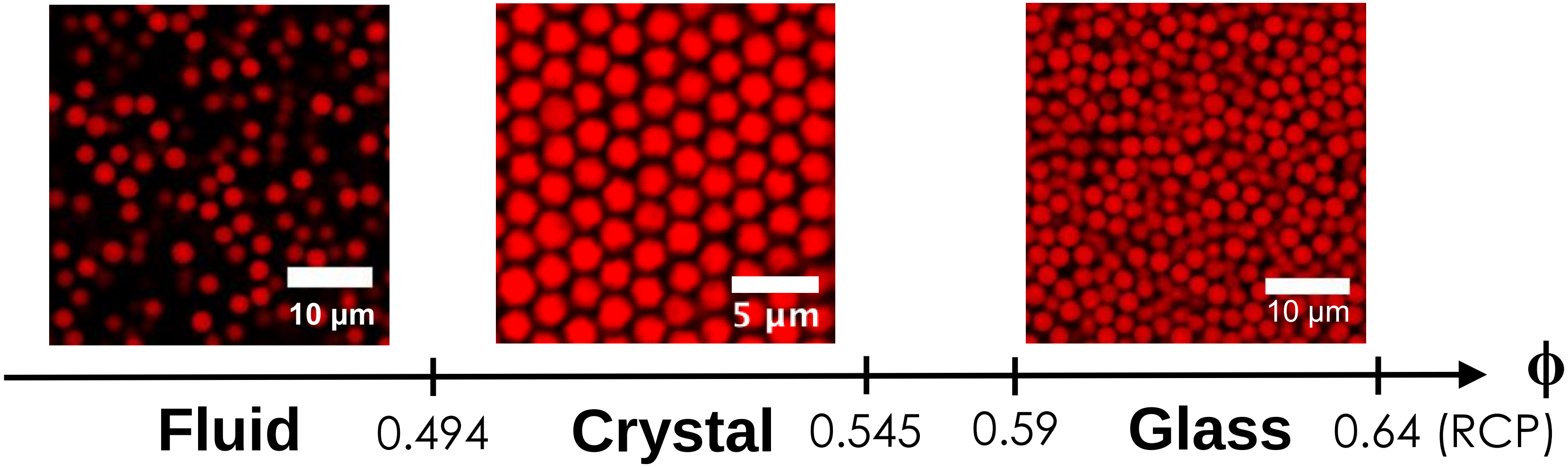
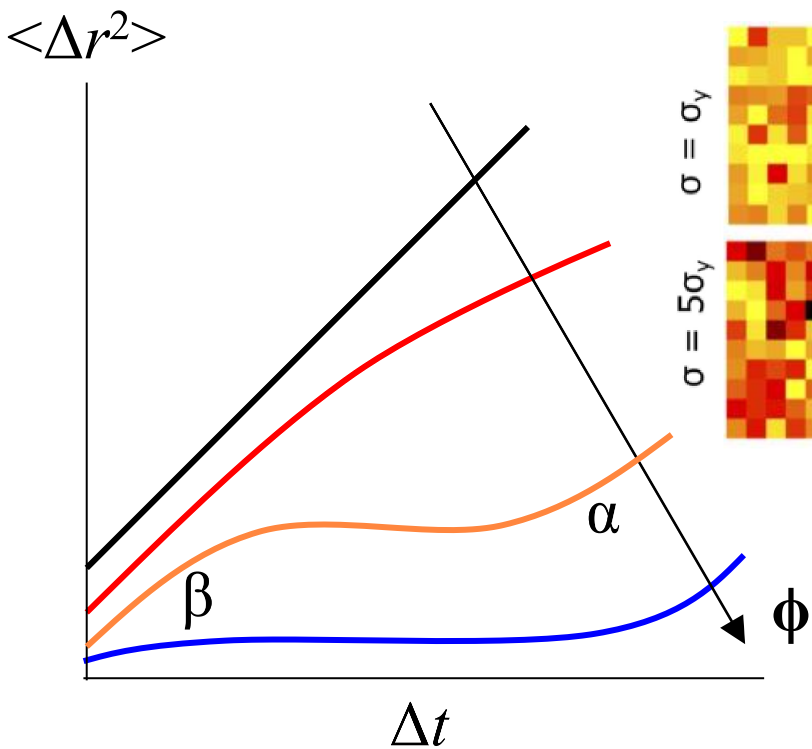
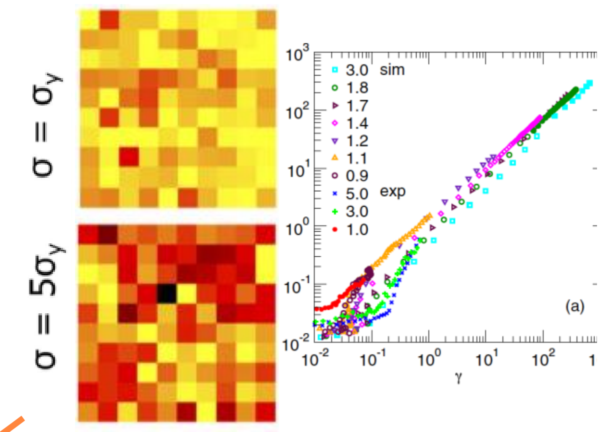
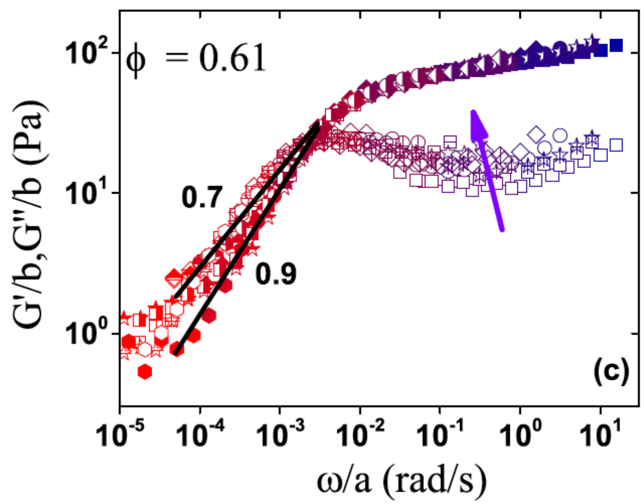
Rheology and dynamics of colloidal glasses







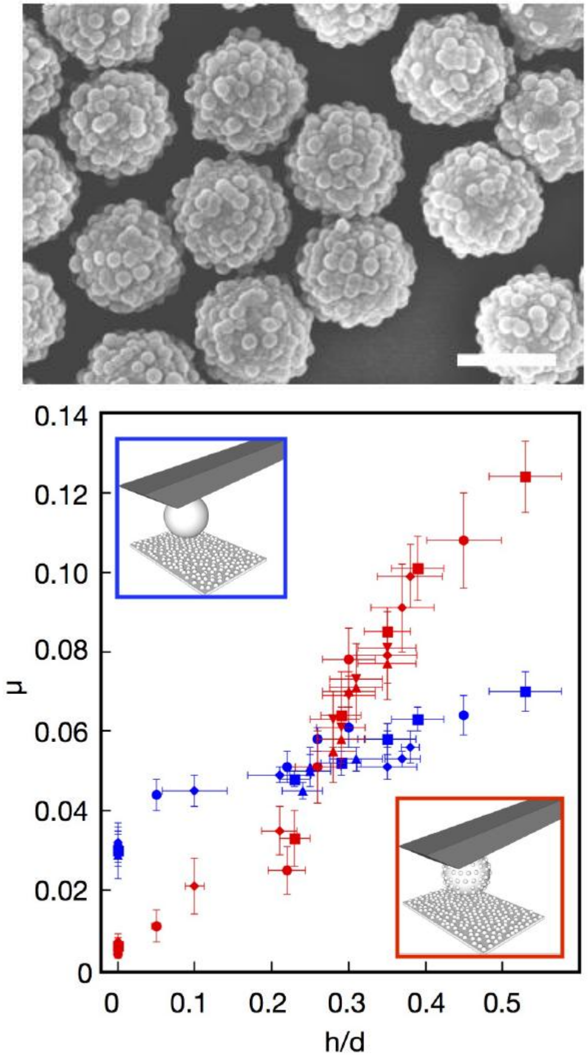
Sentjabrskaja et al. Sci Rep (2015).



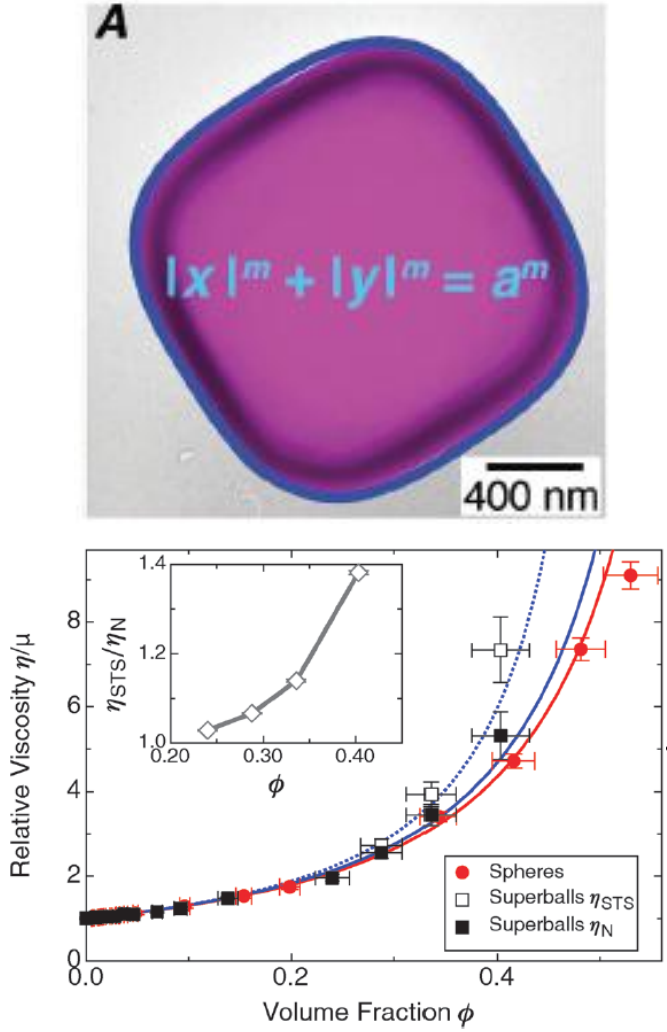
Jacob et al. PRL (2015)

Two dynamic relaxation modes responsible for cage + bond release under shear

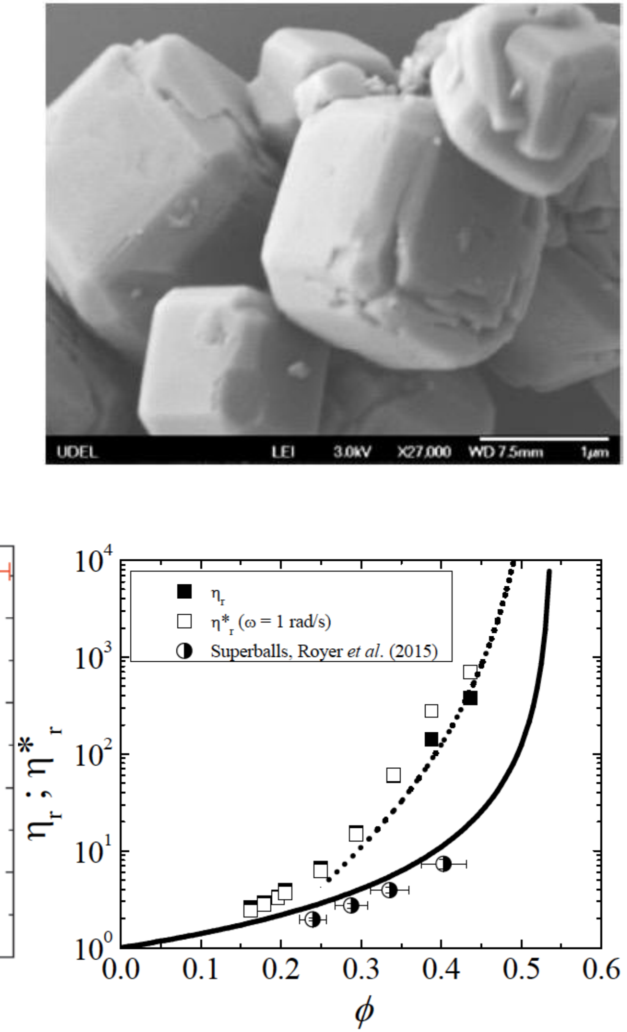
Effect of surface anisotropy on dense suspensions



Hsu & Isa et al. arXiv (2018).



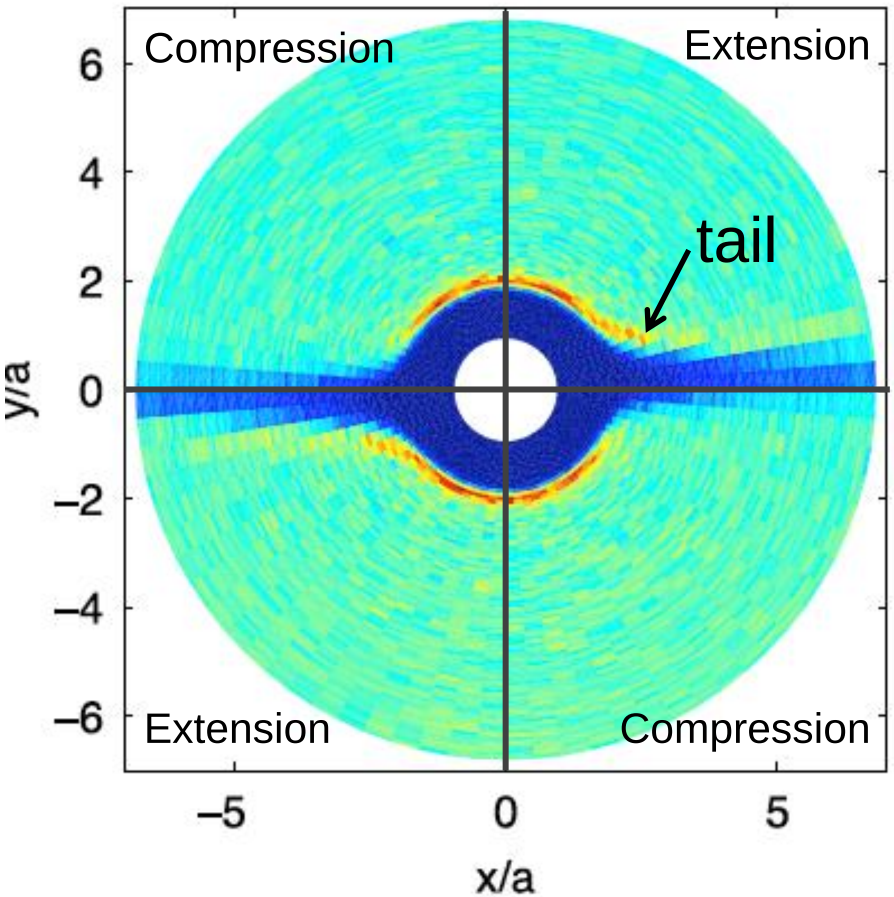
Royer, Blair, Hudson et al. Soft Matter (2015).



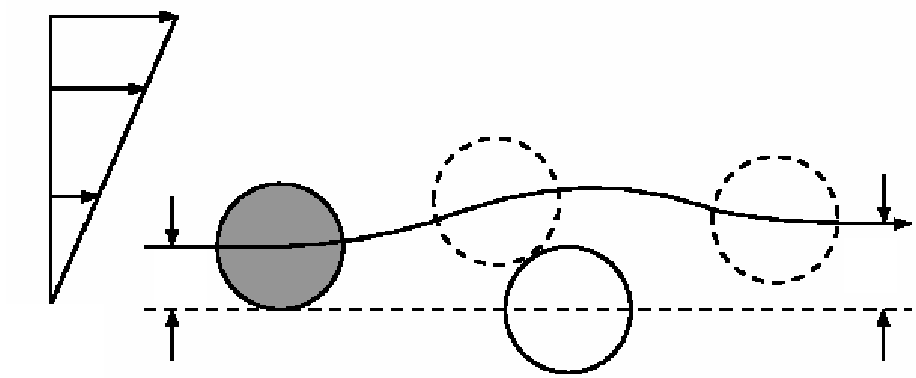
Cwalina & Wagner. Soft Matter (2016).

Roughness breaks fore-aft symmetry in simple shear

Experiments with non-Brownian PMMA (ϕ = 0.05) in Couette cell



Blanc et al. Phys Rev Lett (2011)



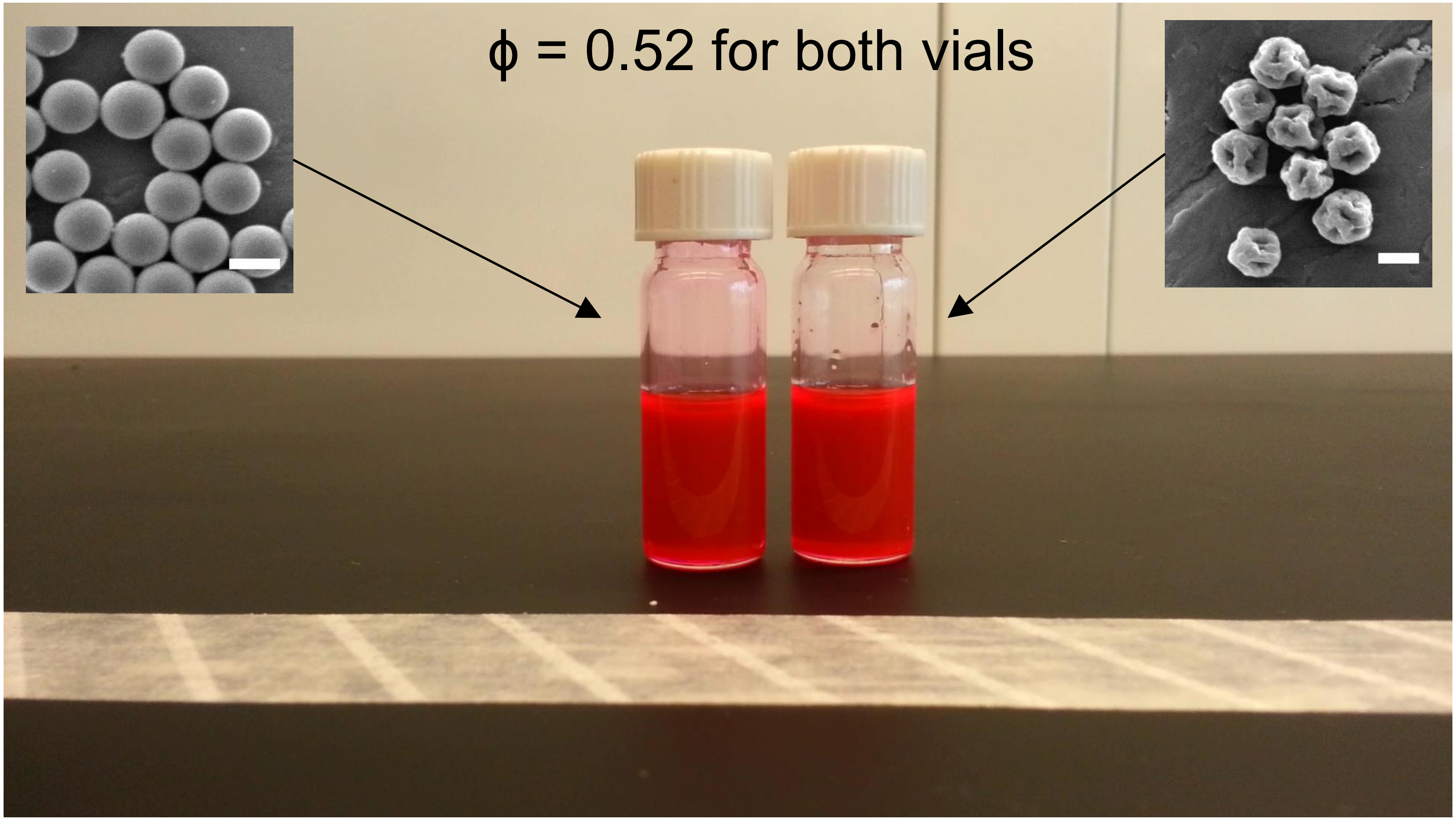
Davis et al. Phil Trans R Soc Lond A (2003)

Theoretical developments:

- Asperity prevents particle contact
- 2 limiting cases: slide (frictionless) and interlocking rigid bodies (frictional)
- Roughness brings Stokes flow irreversibility and symmetry breaking

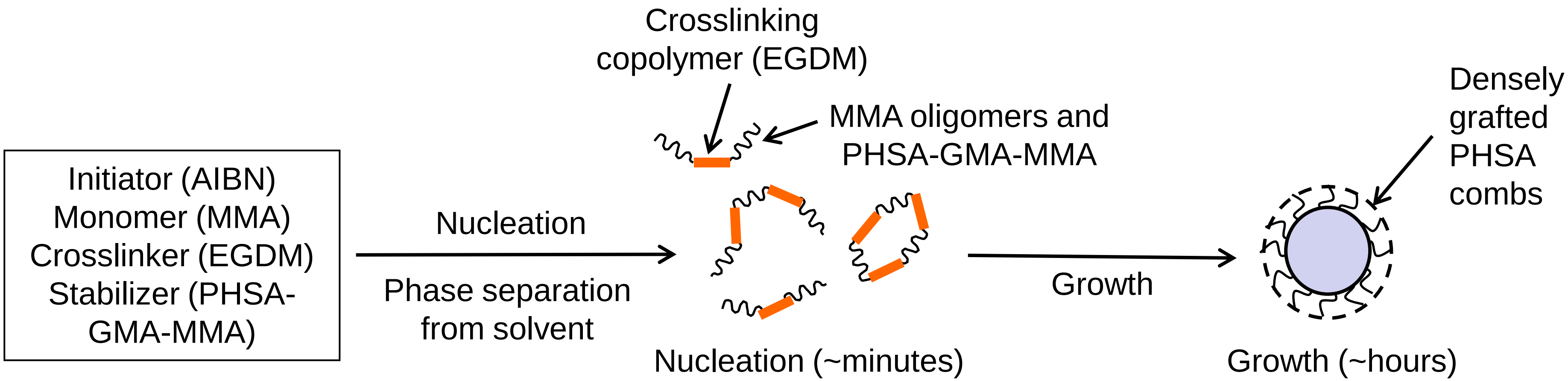
Smart & Leighton (1993), Gallier et al. (2014), Wilson & Davis (2002), Da Cunha & Hinch (1996), Sierou & Brady (2002)

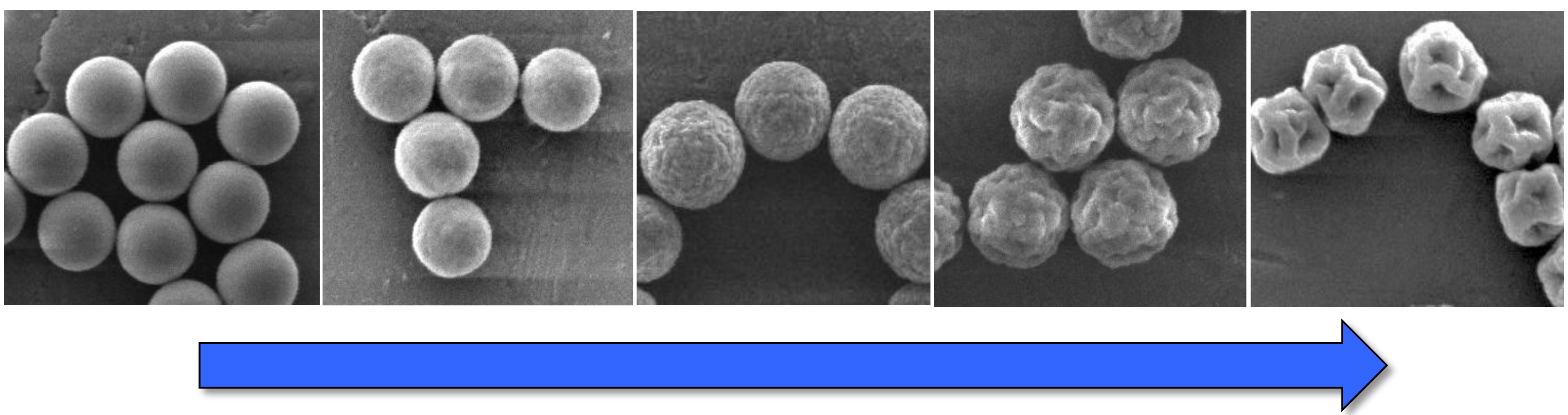
Particle roughness and shear thickening



Synthesis of PHSA-PMMA rough colloids

Free-radical dispersion polymerization

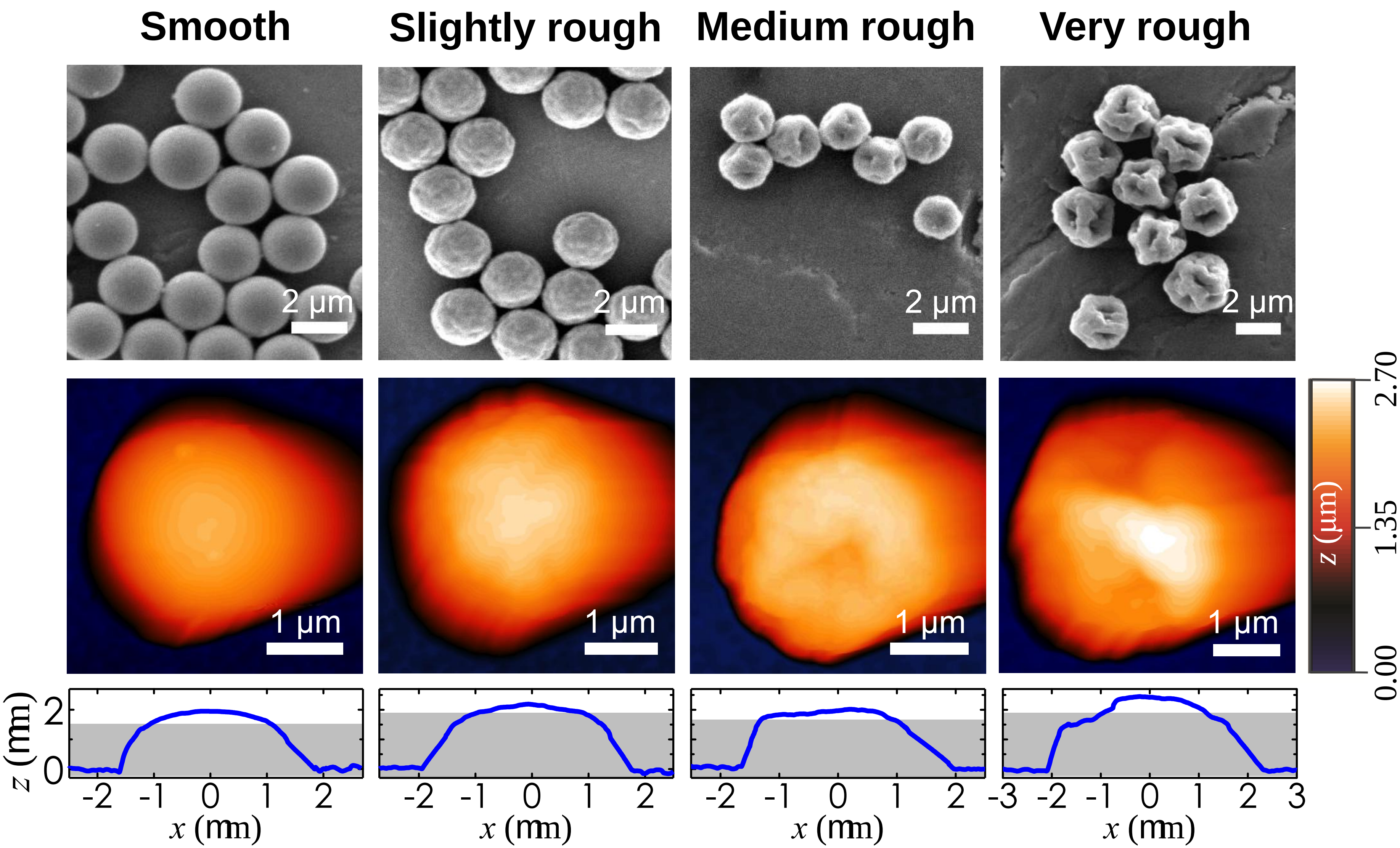




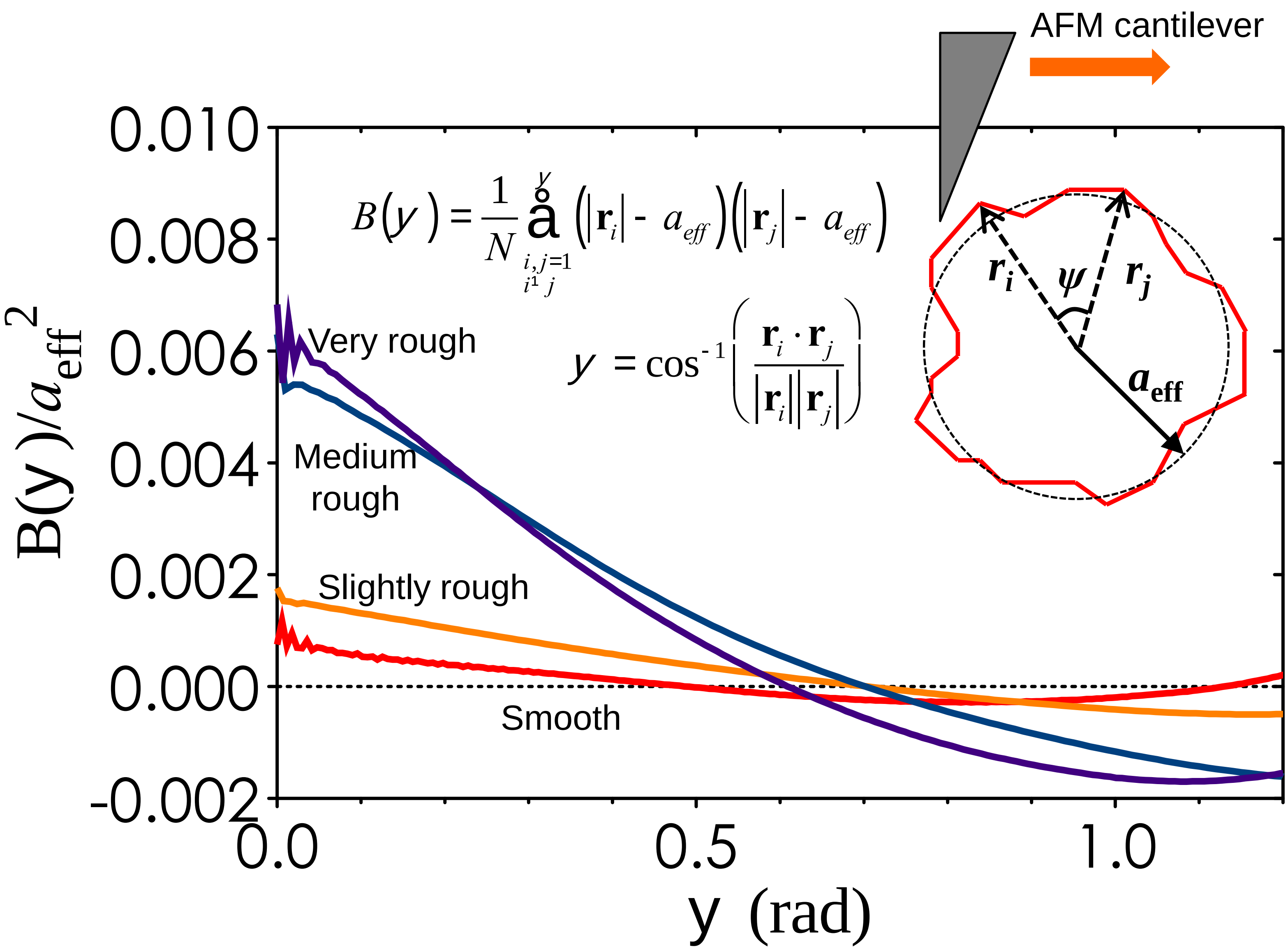
Increasing crosslinker concentration, Increasing roughness

Colloidal particles of varying roughness

PHSA-stabilized PMMA colloids, $2a_{eff}$ = 1.9 to 2.5 μm, Size polydispersity = 3-4%



Extracting roughness from AFM measurements

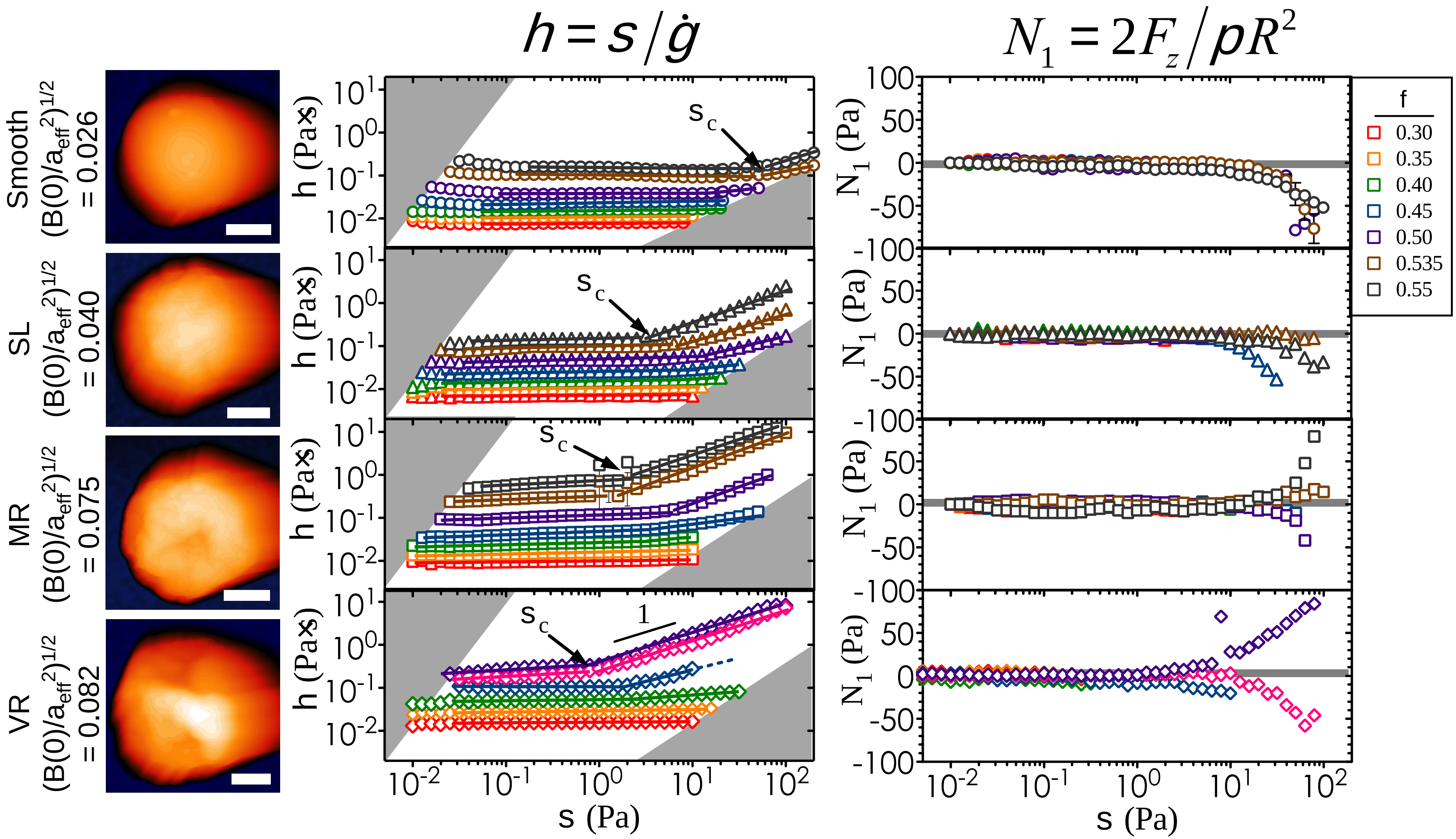


$$B(\psi) = \frac{1}{N}\sum_{\substack{i,j=1 \\ i \neq j}}^{\psi} \left(|\mathbf{r}_i| - a_{eff}\right)\left(|\mathbf{r}_j| - a_{eff}\right)$$

$$\psi = \cos^{-1}\left(\frac{\mathbf{r}_i \cdot \mathbf{r}_j}{|\mathbf{r}_i||\mathbf{r}_j|}\right)$$

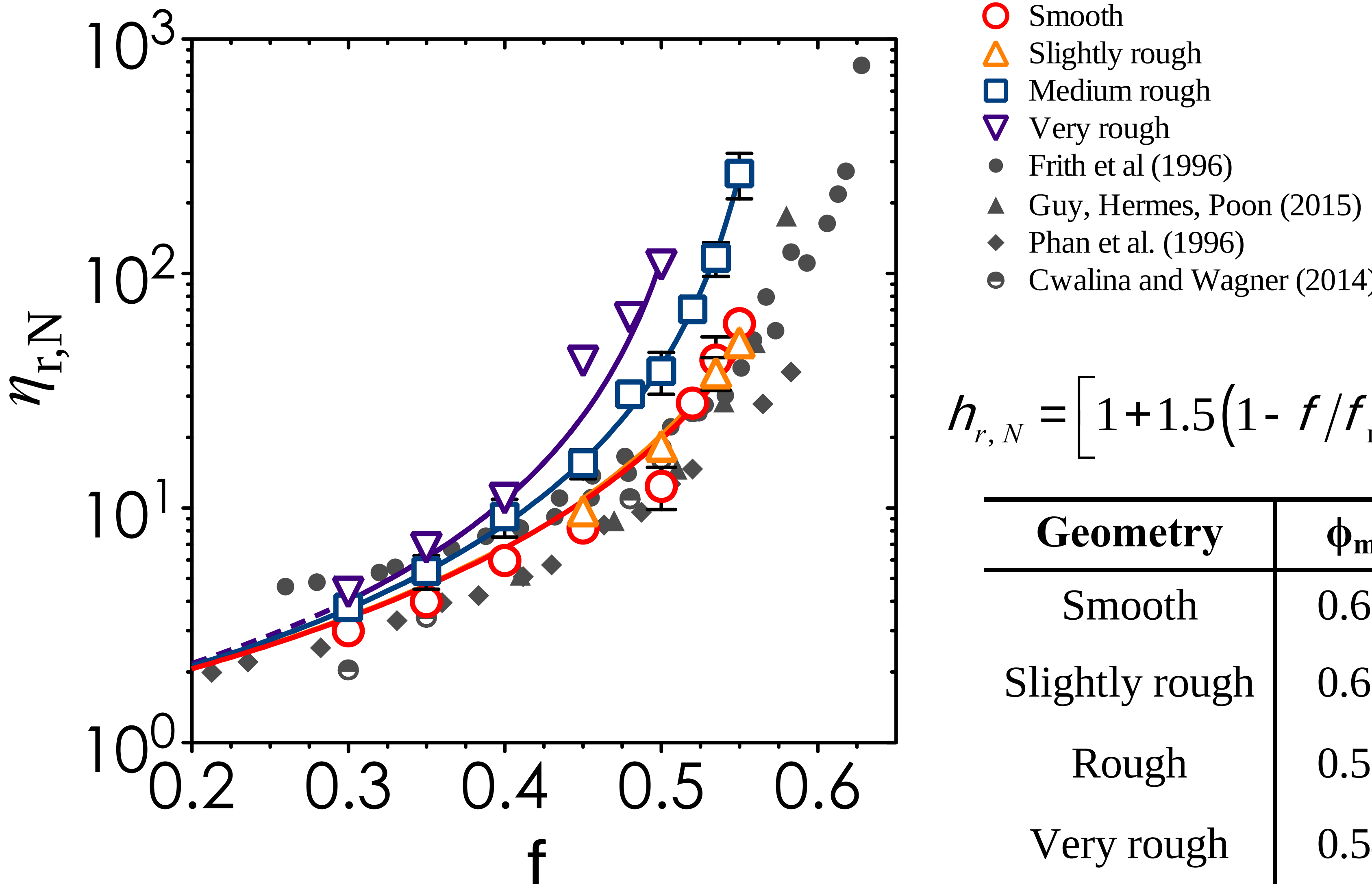
Hsiao, Jamali et al. Phys Rev Lett (2017)

Steady state viscosity and first normal stresses



Hsiao, Jamali et al. Phys Rev Lett (2017)

Fitting the high-shear viscosity to the Eilers model

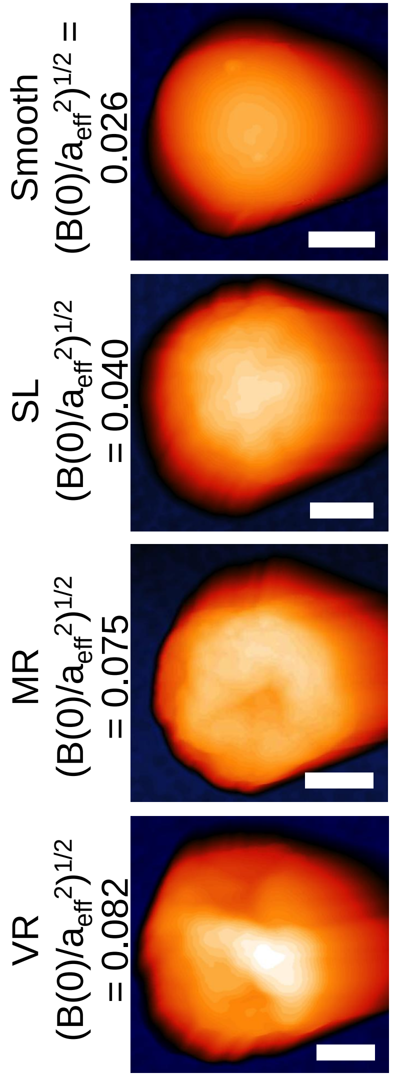


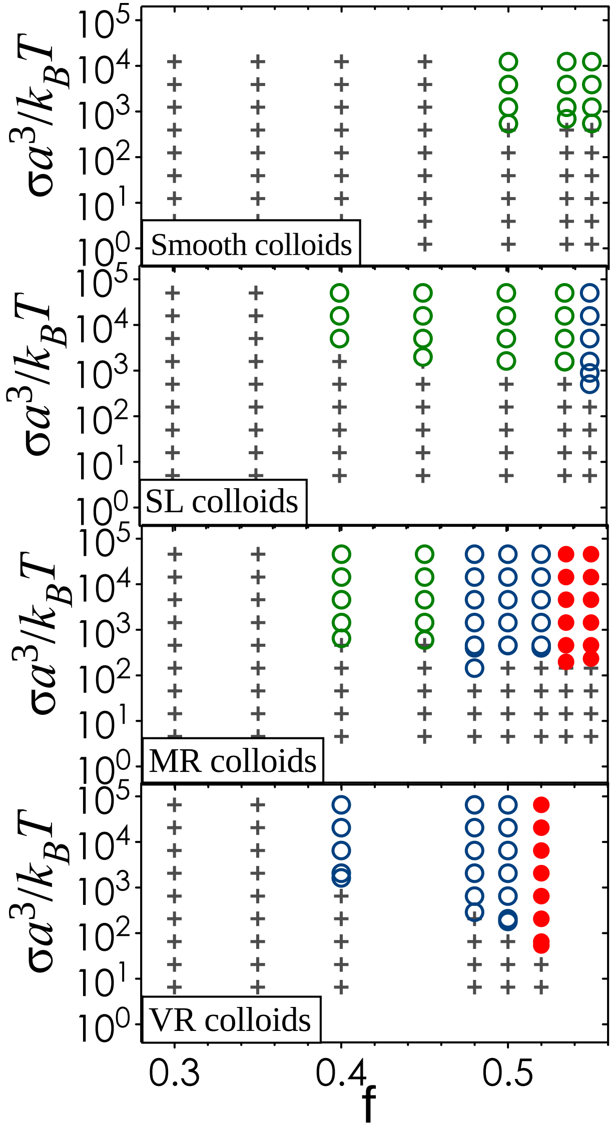
$$\eta_{r,N} = \left[1 + 1.5\left(1 - \phi/\phi_{max}\right)^{-1}\right]^2$$

Geometry	ϕ_{max}
Smooth	0.635
Slightly rough	0.636
Rough	0.581
Very rough	0.542

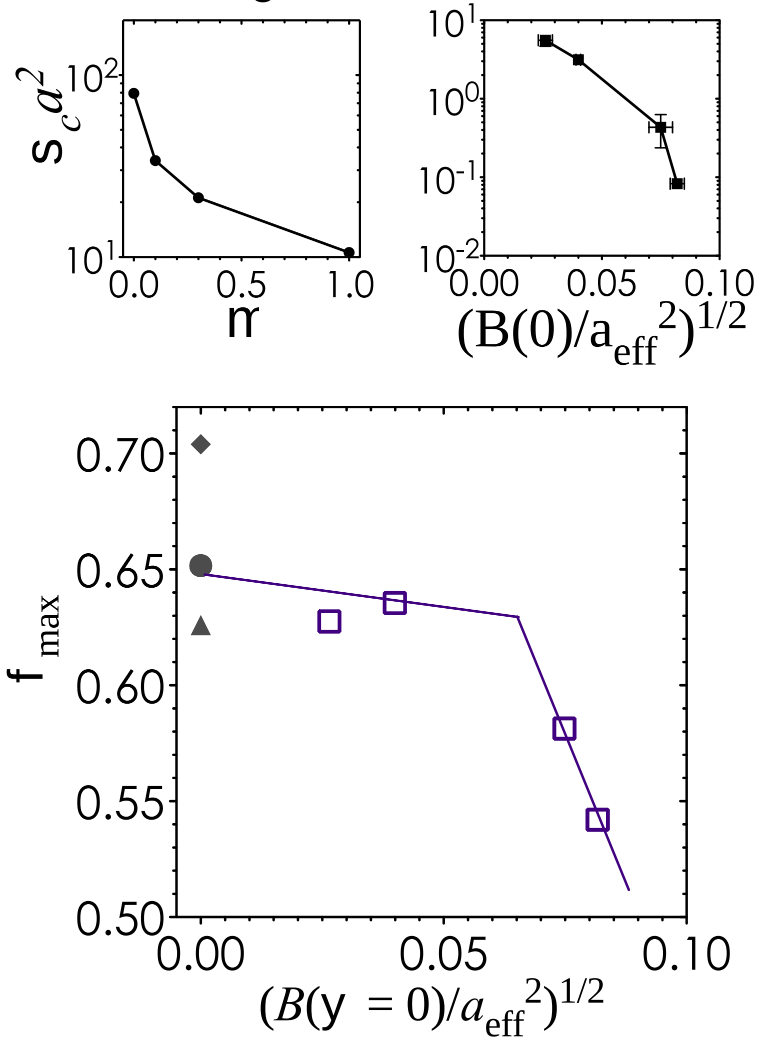
Hsiao, Jamali et al. Phys Rev Lett (2017)

State diagrams for rough colloids in shear flow



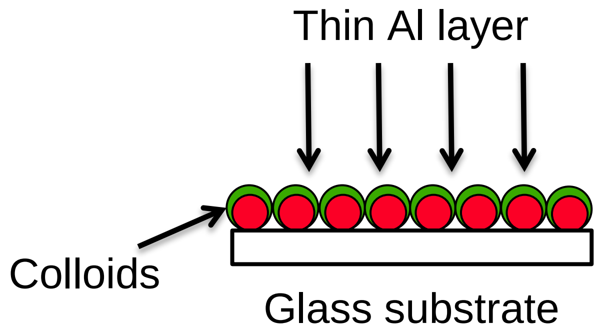


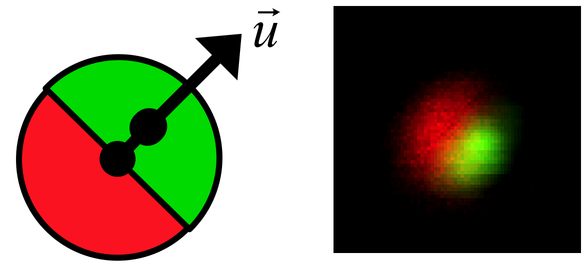
Onset stress depends on roughness and "friction"



Hsiao, Jamali et al. Phys Rev Lett (2017)

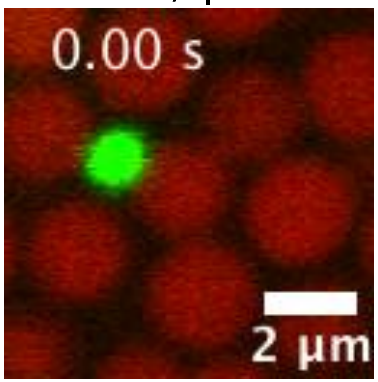
Roughness slows down rotational relaxation



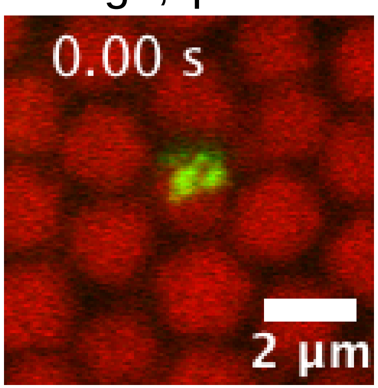


$$\langle \Delta \vec{\varphi}^{\,2}(\Delta t) \rangle = \langle [\vec{\varphi}(t+\Delta t) - \vec{\varphi}(t)]^2 \rangle$$

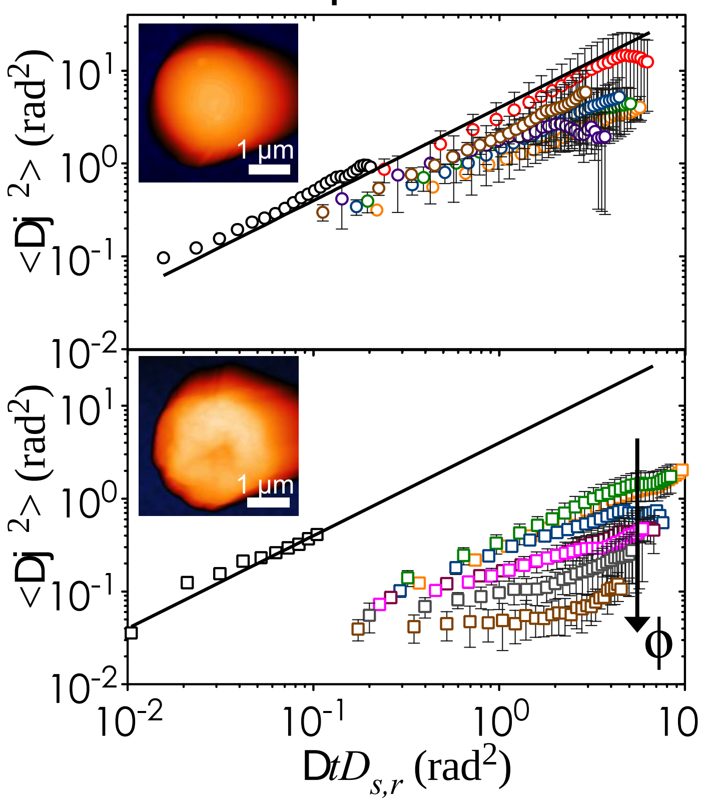
Smooth, ϕ = 0.50



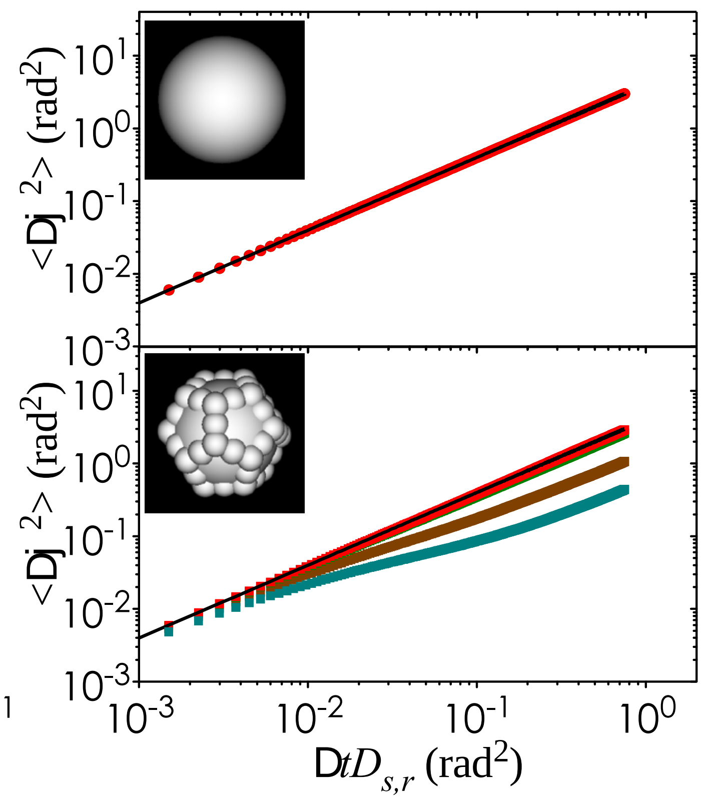
Rough, ϕ = 0.50



Experiments

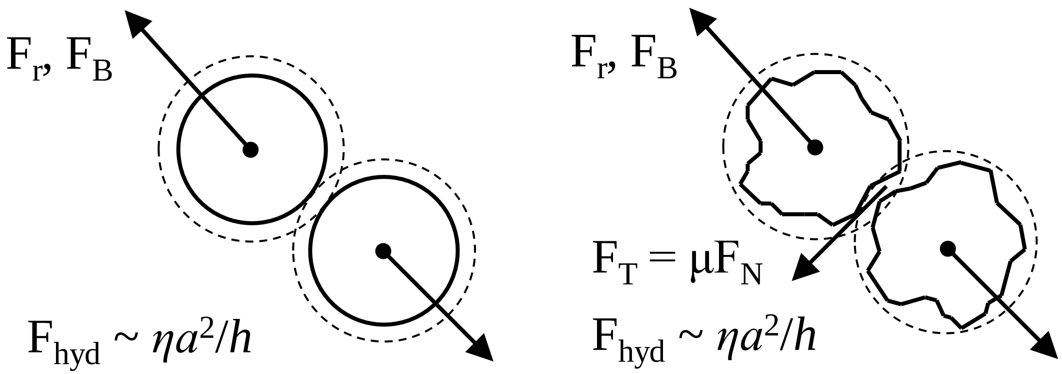


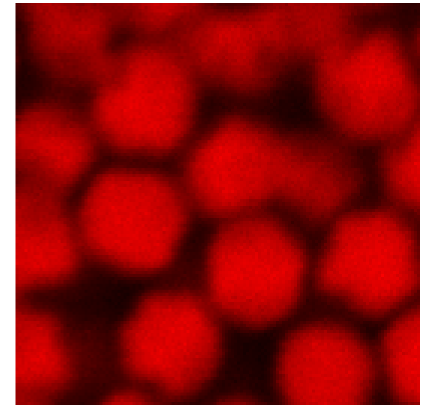
BD simulations



Hsiao et al. Soft Matter (2017)

Resistance to rotations and the glass transition





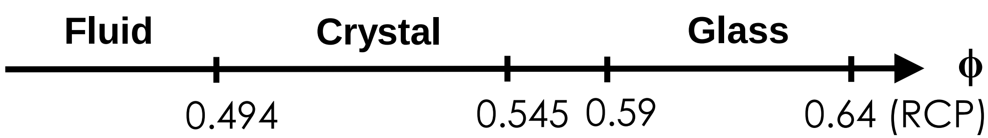
$$m = \frac{F_t}{F_N}$$

1) Besides viscosity and N_1, what are other rheological signatures for smooth sphere and rough sphere suspensions of similar φ?

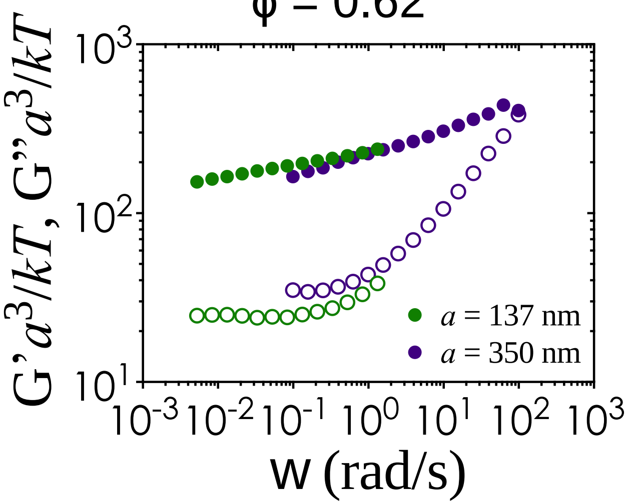
2) Does roughness shift the glass transition to lower φ? Why?

3) How does roughness affect the avalanche-like yielding of dense suspensions?

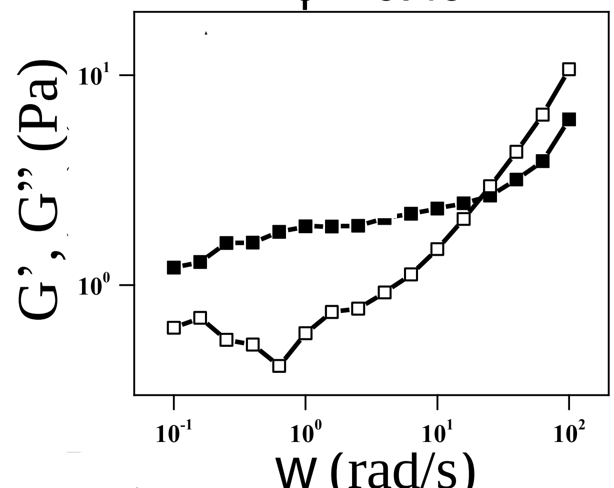
Linear viscoelasticity of rough colloids

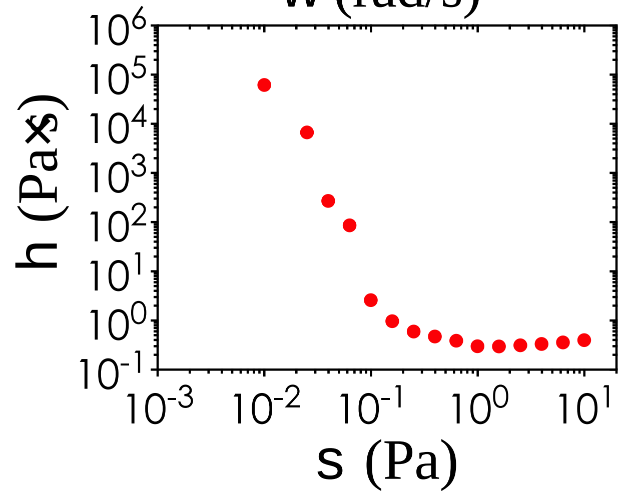


Smooth PMMA colloids
ϕ = 0.62



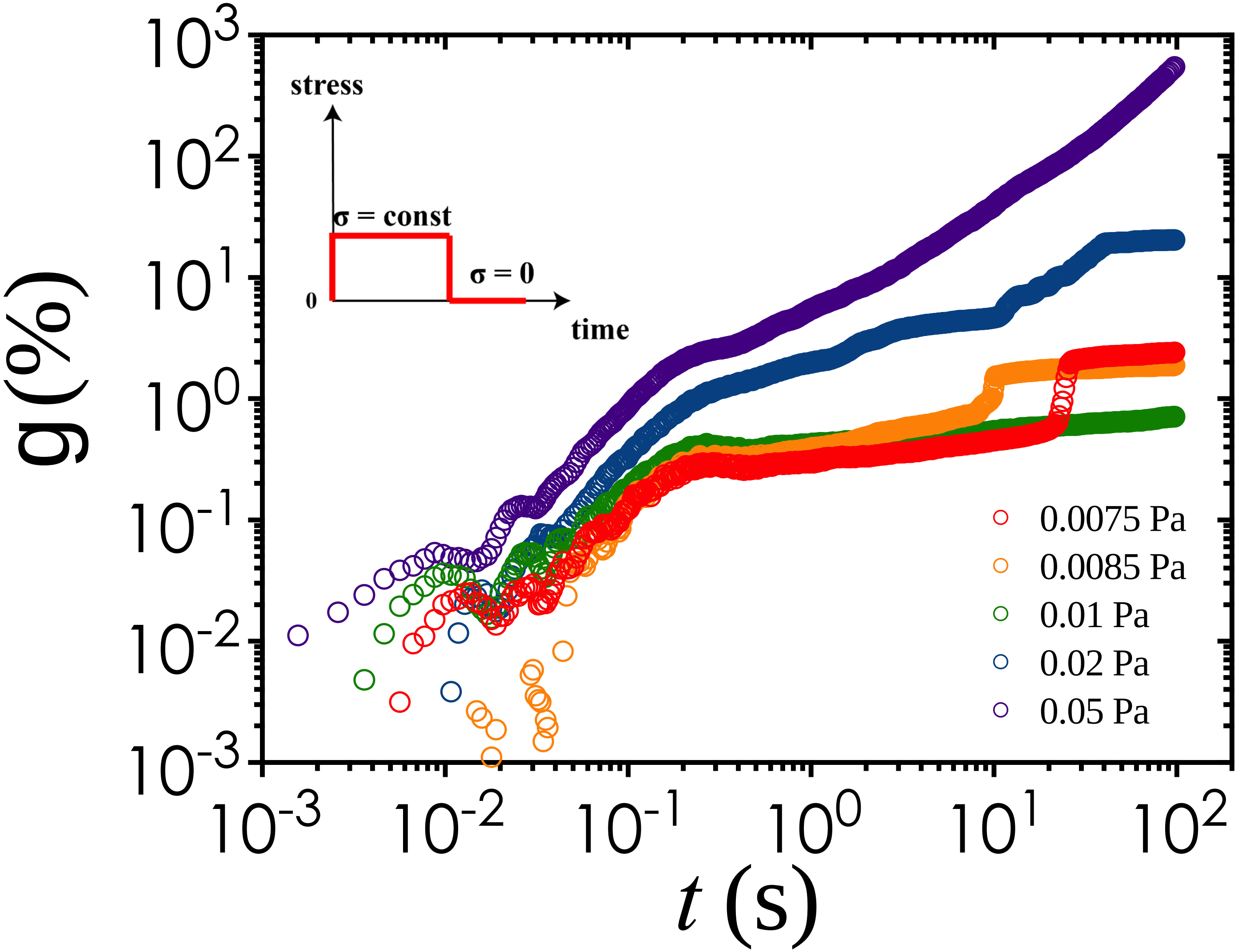
Rough PMMA colloids
ϕ = 0.49





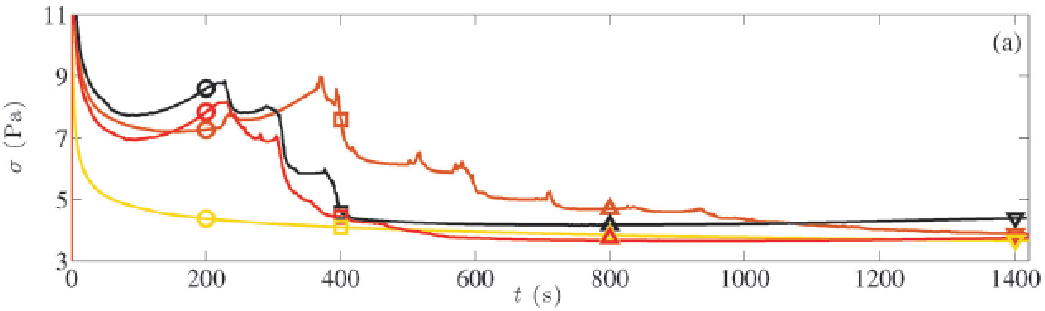
Glass-like viscoelasticity
Yield stress fluid

Creep under step stress



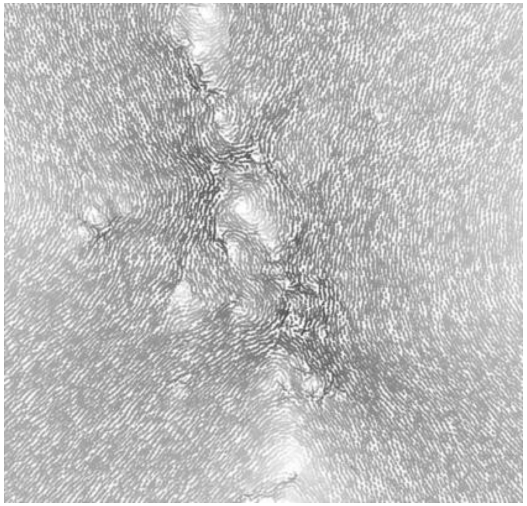
Avalanche-like fluidization in particulate systems

Non-Brownian gels



Kurokawa, Divoux & Manneville et al. Soft Matter (2015).

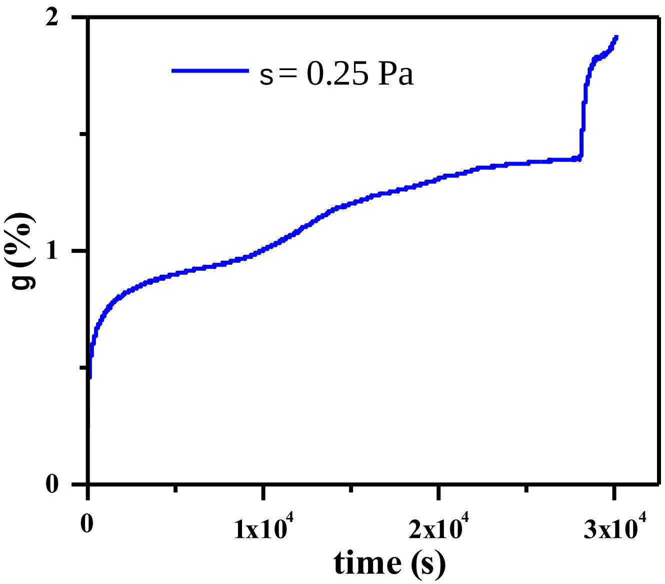
2D athermal systems



Maloney & Lemaitre. PRE (2006).

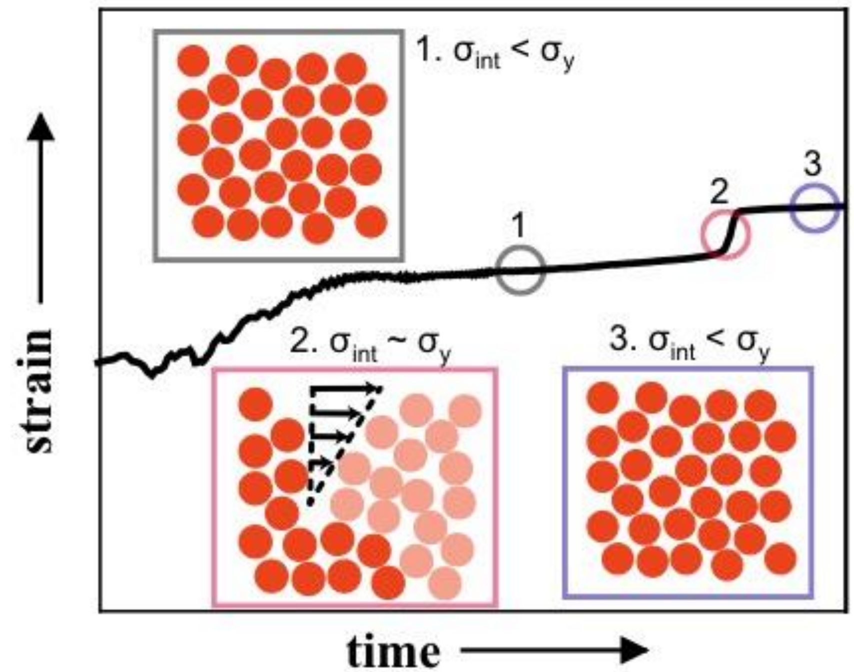
Smooth colloids

(a = 107 nm, ϕ = 0.59)



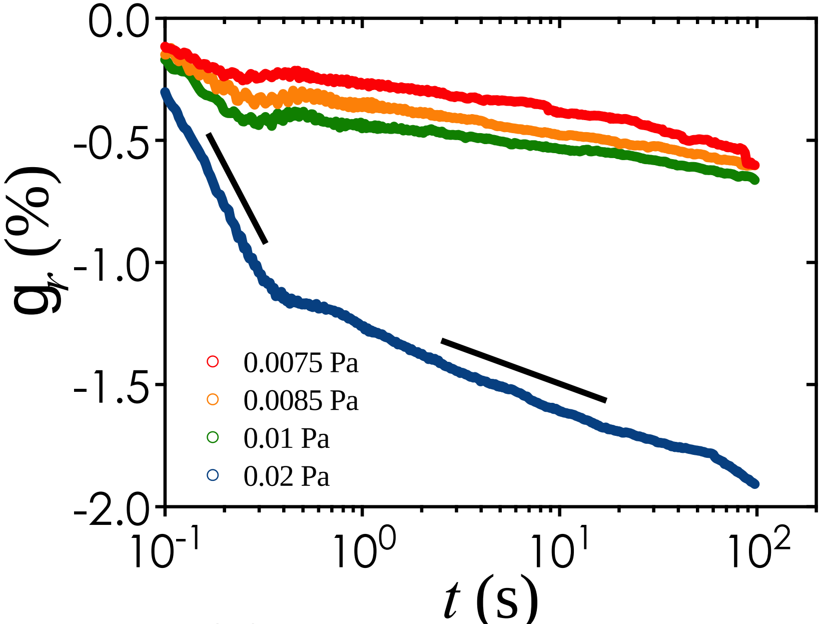
Courtesy Alan Jacob

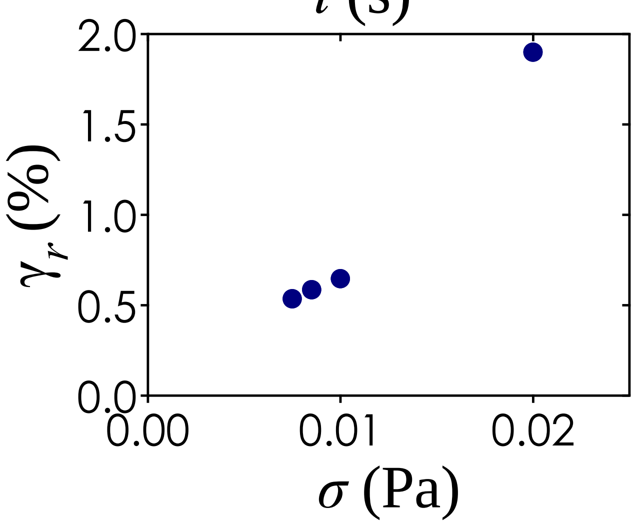
Rough colloids



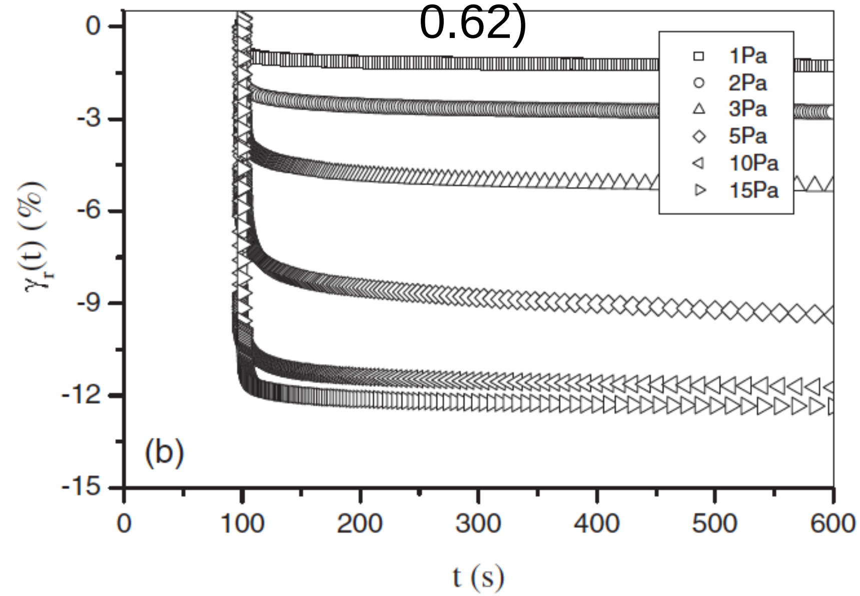
Strain recovery after creep cessation

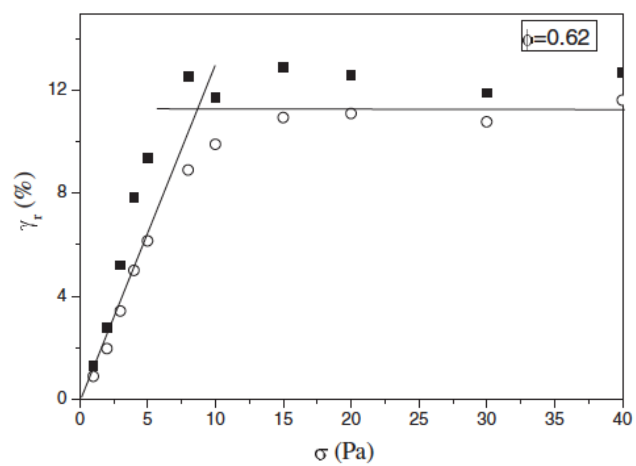
PHSA-PMMA rough colloids
(a = 1.7 μm ± 5%, ϕ = 0.49)





PHSA-PMMA spheres
(a = 183 nm ± 12%, ϕ = 0.62)



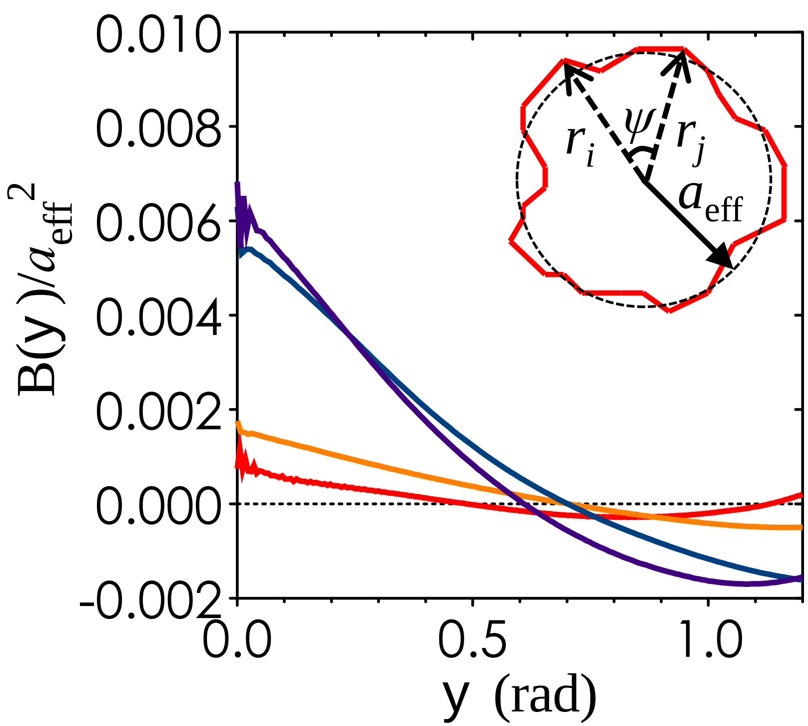


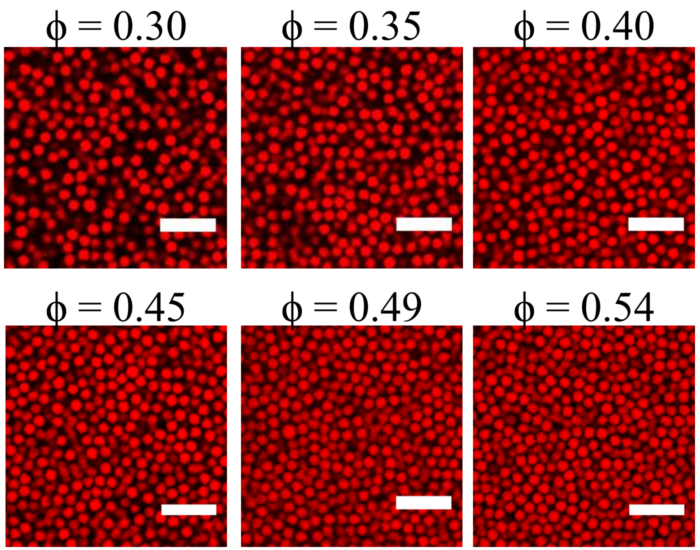
Petekidis, Vlassopoulos & Pusey.
J Phys Condens Matter (2004).

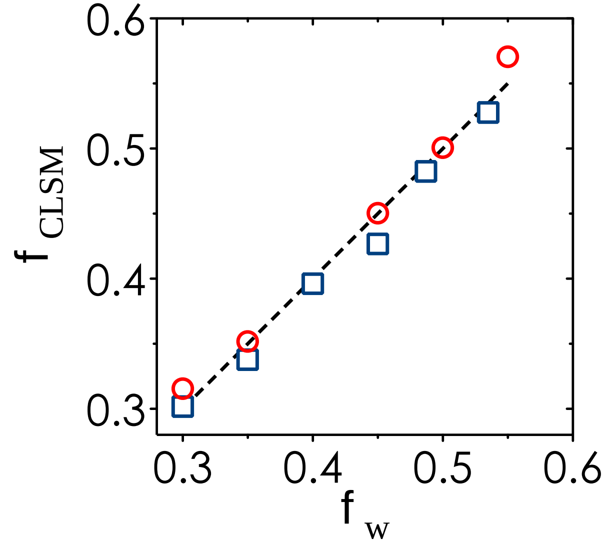
FAQ 1: How did you calculate the volume fraction?

Answer:

Effective size from AFM/SEM, particle counting from 3D confocal microscopy



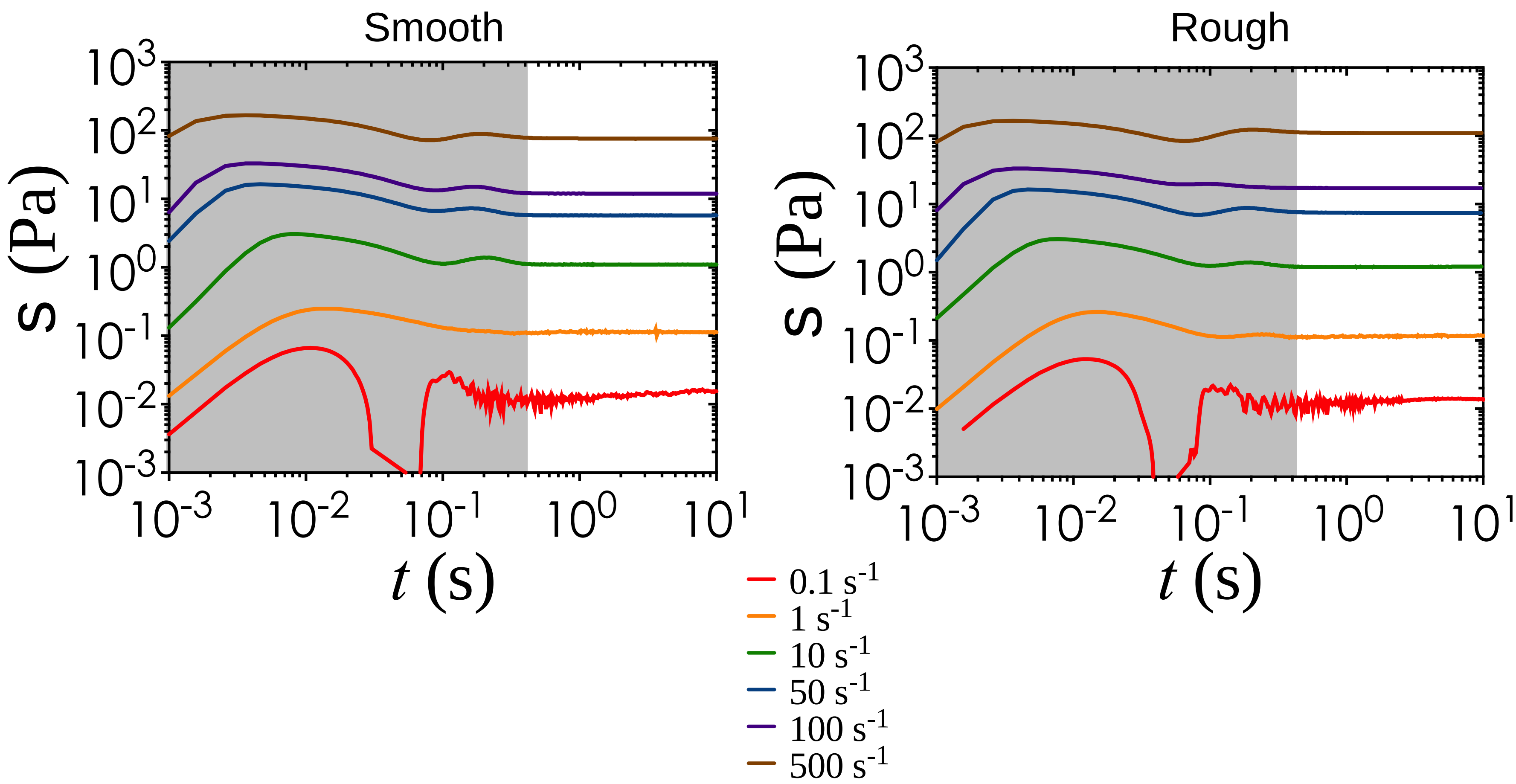




$$f_{CLSM} = \frac{4/3 p a_{eff}^3 N_p}{V_{box}}$$

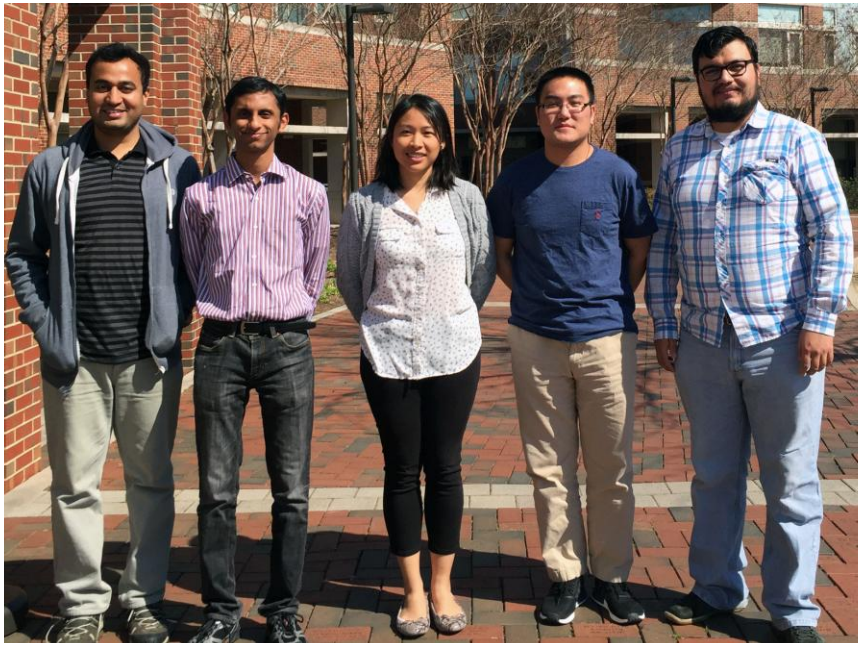
FAQ 2: Is your rheometer sensitive enough?

Answer:
Not below 0.5 s due to motor inertia,
independent of shear rates or strains (DHR-2, TA Instruments)



Summary

- Roughness introduces “friction” in the sense that it is difficult for particles to undergo full rotation
- It shifts maximum packing (based on viscosity divergence) to lower ϕ
- Glassy behavior found at values of ϕ that is normally that of a fluid
  Future work will determine if ϕ/ϕ_{max} can collapse nonlinear rheology data



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