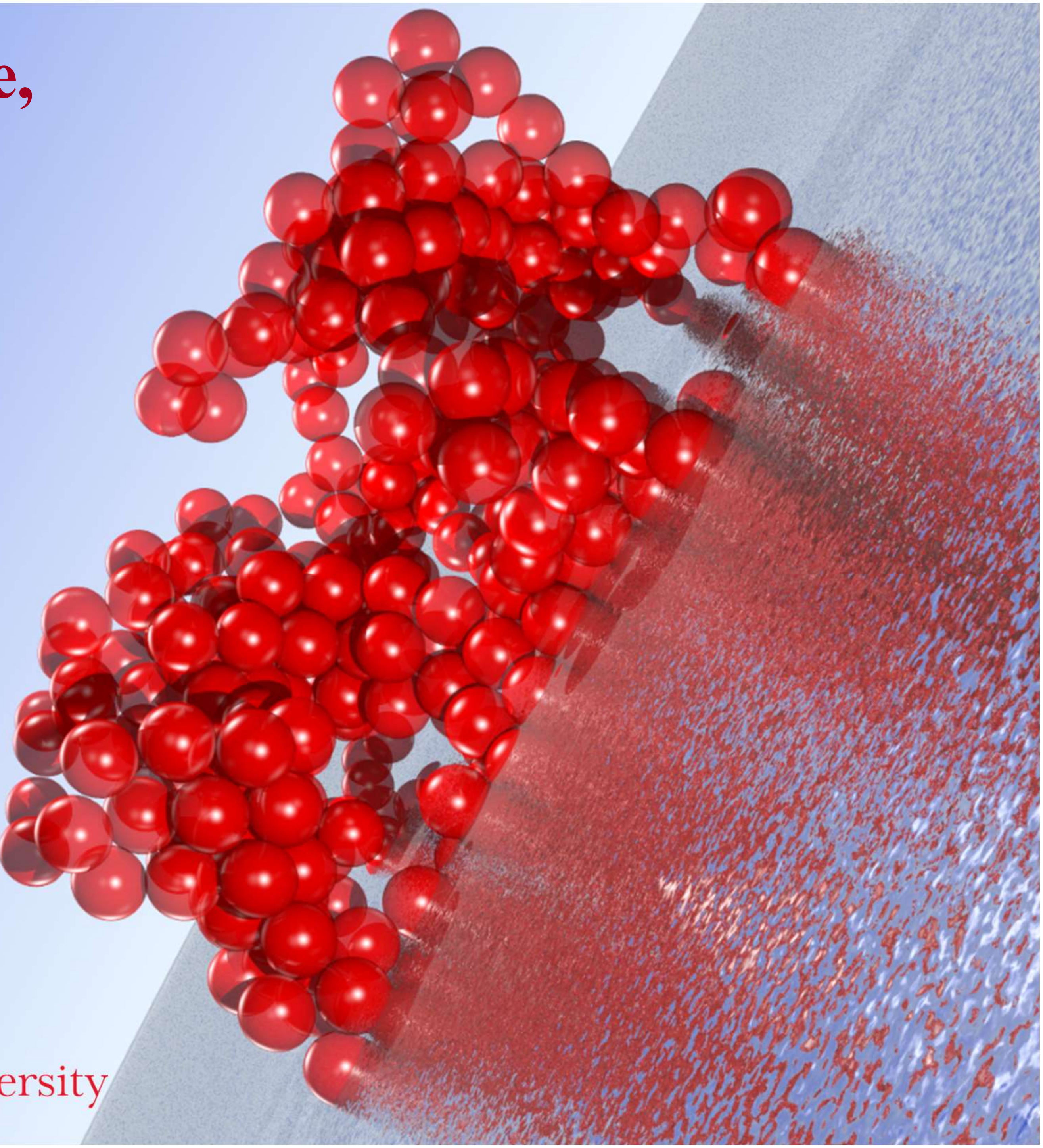
Microstructure, Heterogeneity and Rheology in Colloidal Gels

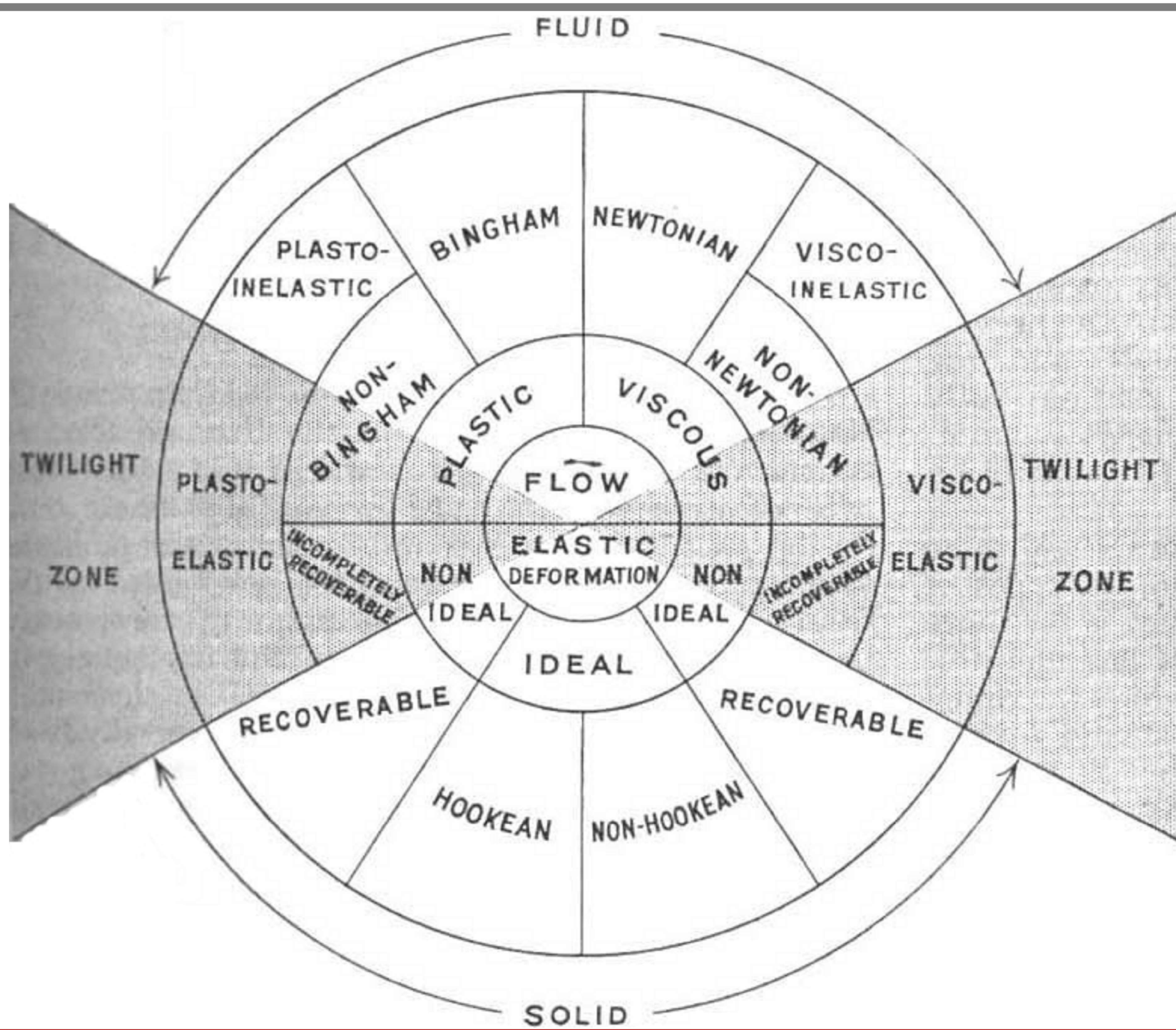


Safa Jamali

MIT: *Gareth H. McKinley, Robert C. Armstrong*
CWRU: *Arman Boromand, Joao Maia*

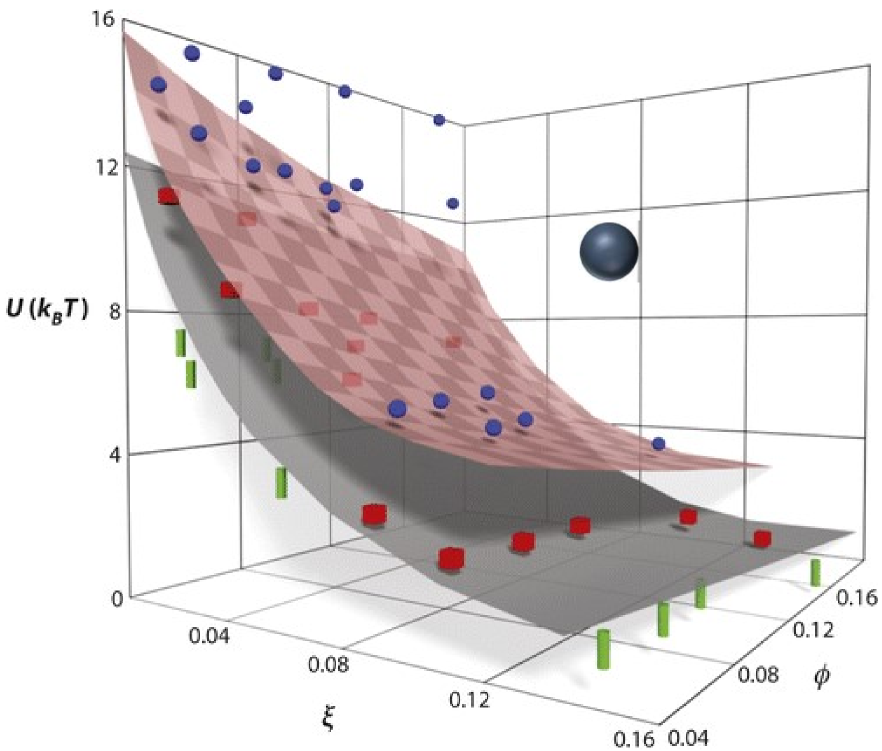
Northeastern University
College *of* Engineering

A Mechanical Perspective

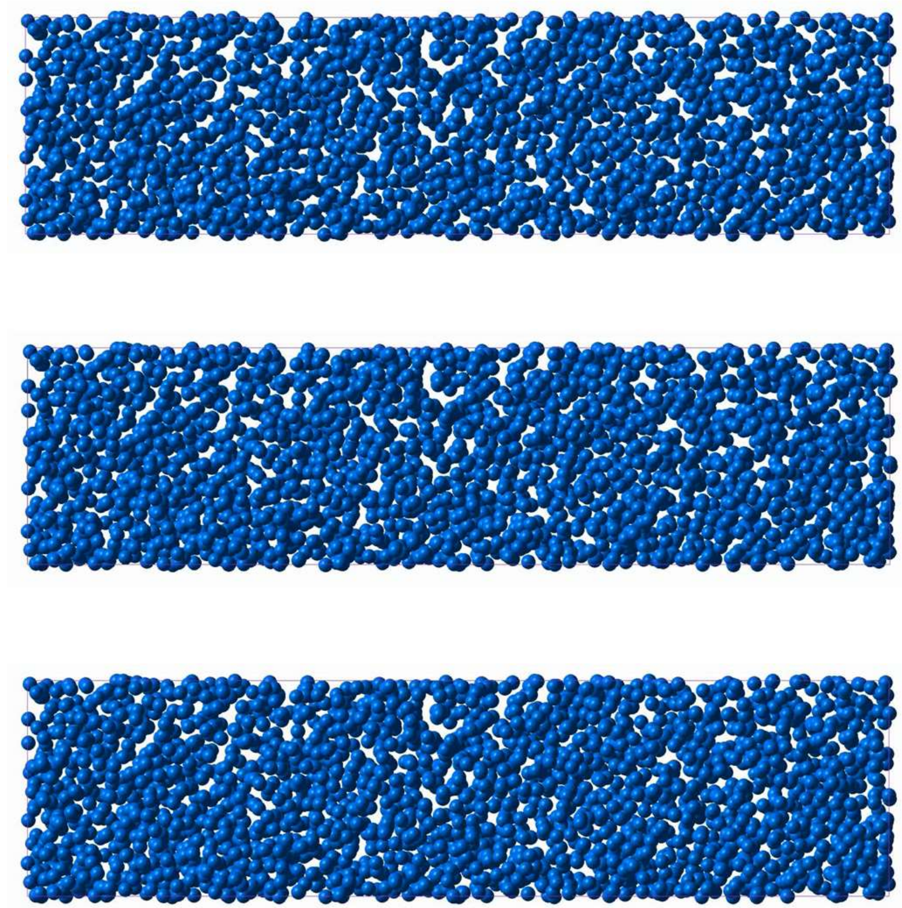


L. Bilmes, *Nature*, 150, p.432, 1942

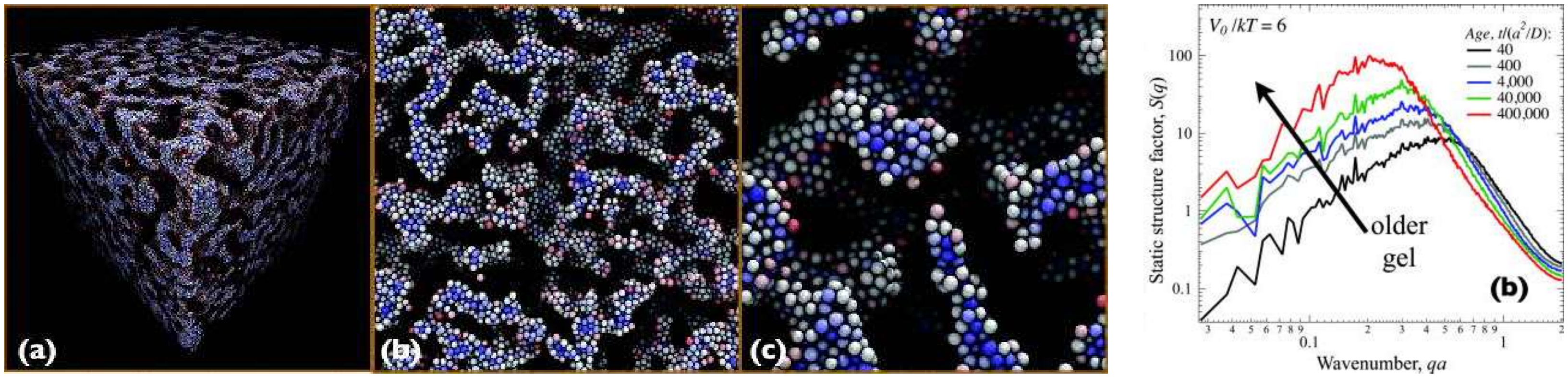
The State Diagram: Does it work?



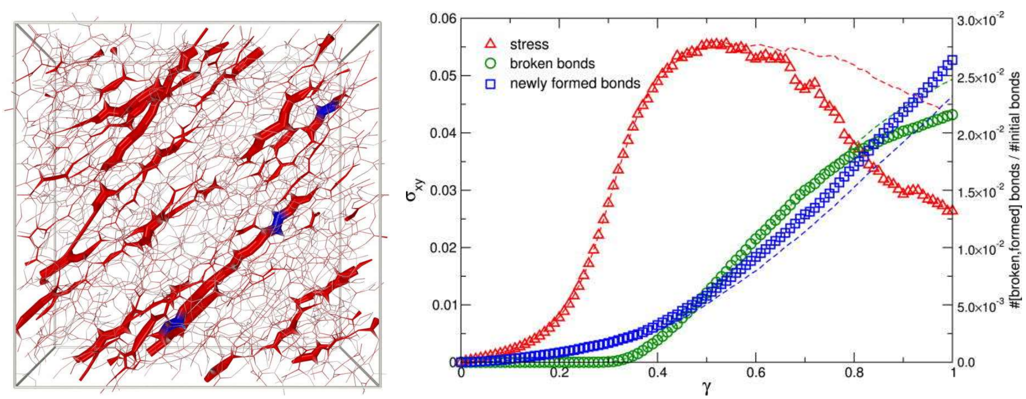
Lu (陸述義) PJ and Weitz DA. 2013.
Annu. Rev. Condens. Matter Phys. 4:217–233



Gels: Formation and Yielding



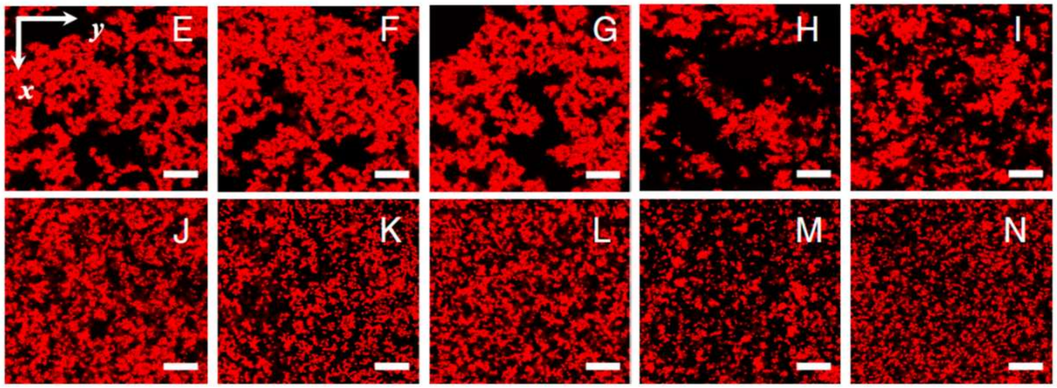
Zia, Landrum and Russel, *Journal of Rheology*, 2014

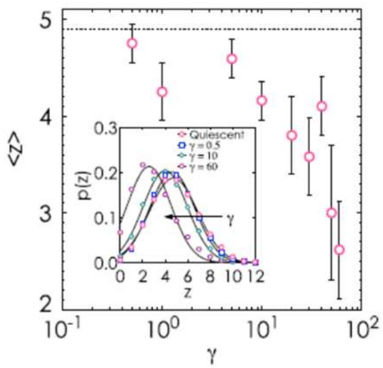


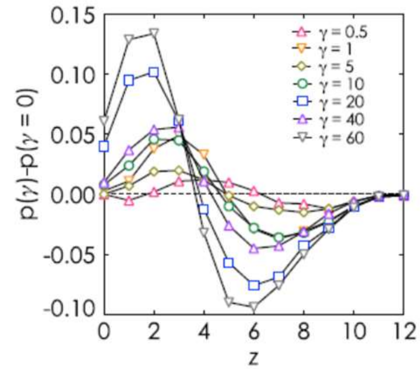
Colombo and Del Gado, *Journal of Rheology*, 2014

Others:
Poon, Petekidis,
Vlassopoulos, Cipelletti,
Zaccarelli, Weitz, Helgeson,
Wagner, Manneville, Divoux,
Fielding, Cates, Solomon…

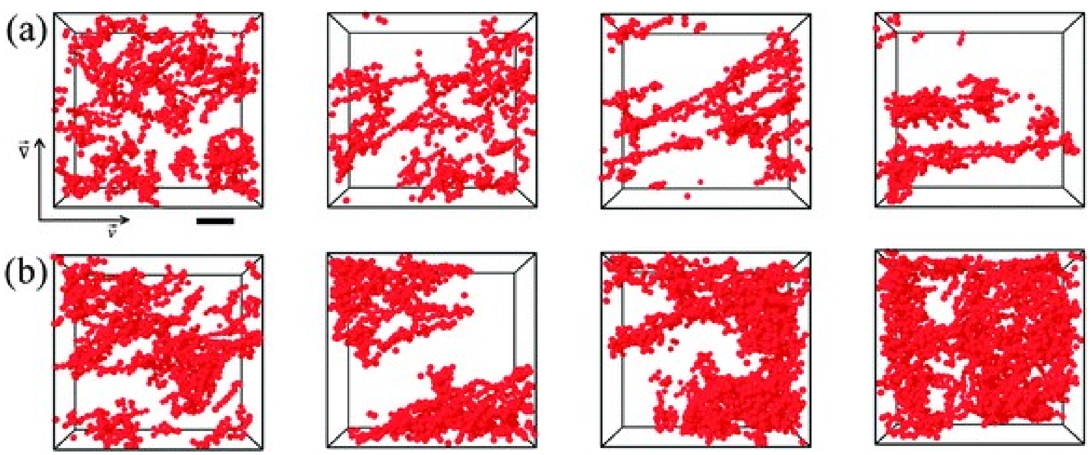
Attractive Colloids under Flow

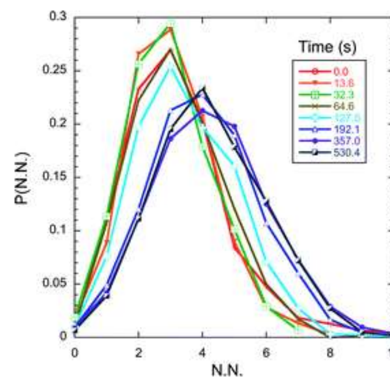


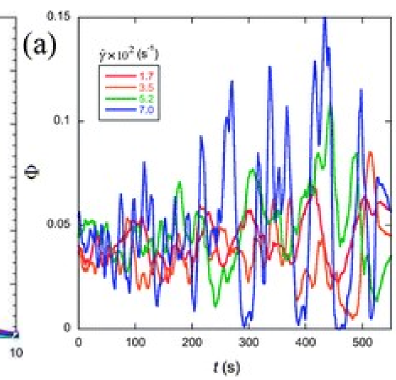




Hsiao et al., PNAS, 109 (40), 2012

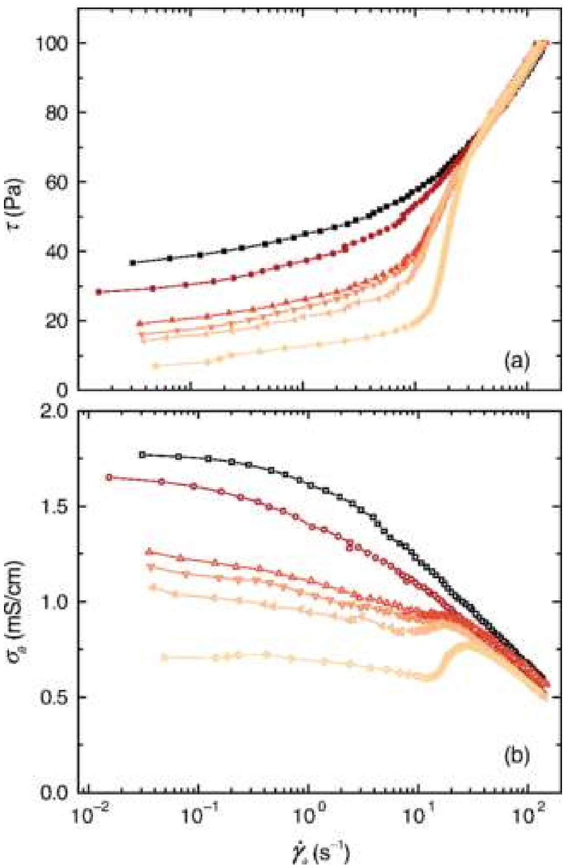




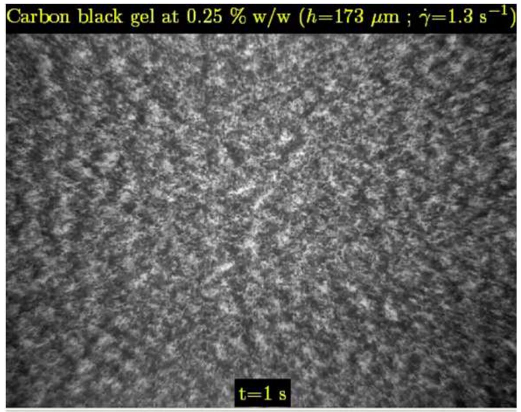


Rajaram and Mohraz, Soft Matter, 6, 2010

Macroscopic Features under Flow

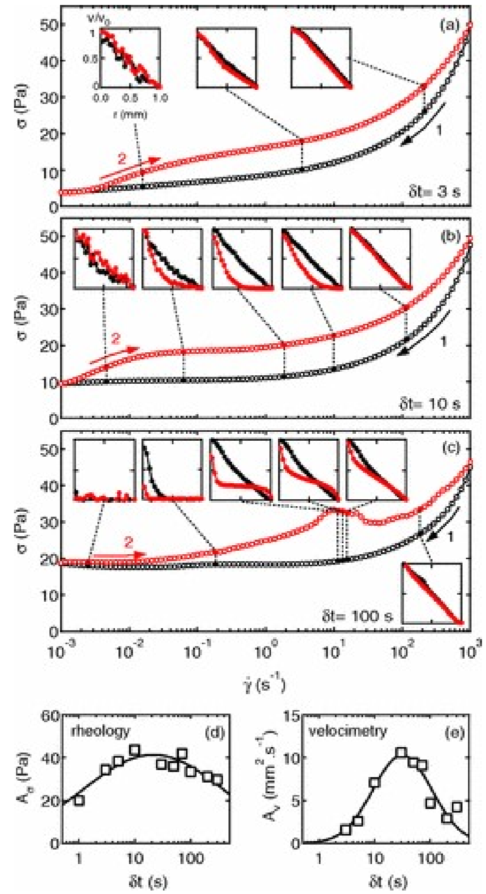


Helal, Divoux and McKinley, *PRApplied*, 2017



Grenard, Taberlet and Manneville, *Soft Matter*, 2011

Osuji and Weitz, *Soft Matter*, 2008



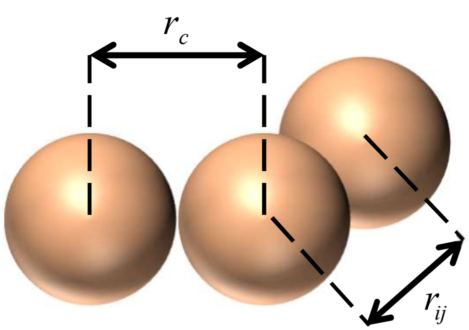
Divoux, Grenard and Manneville, *PRL*, 2013

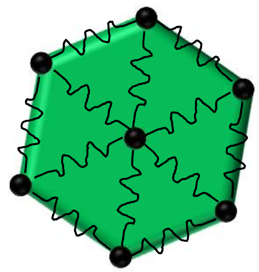
Radhakrishnan et al., *Soft Matter*, 2017

Modified DPD: Attractive Particles

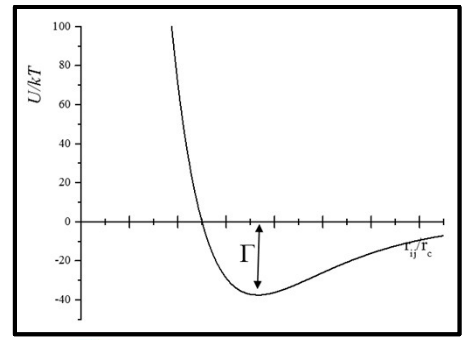
- Equation of motion:

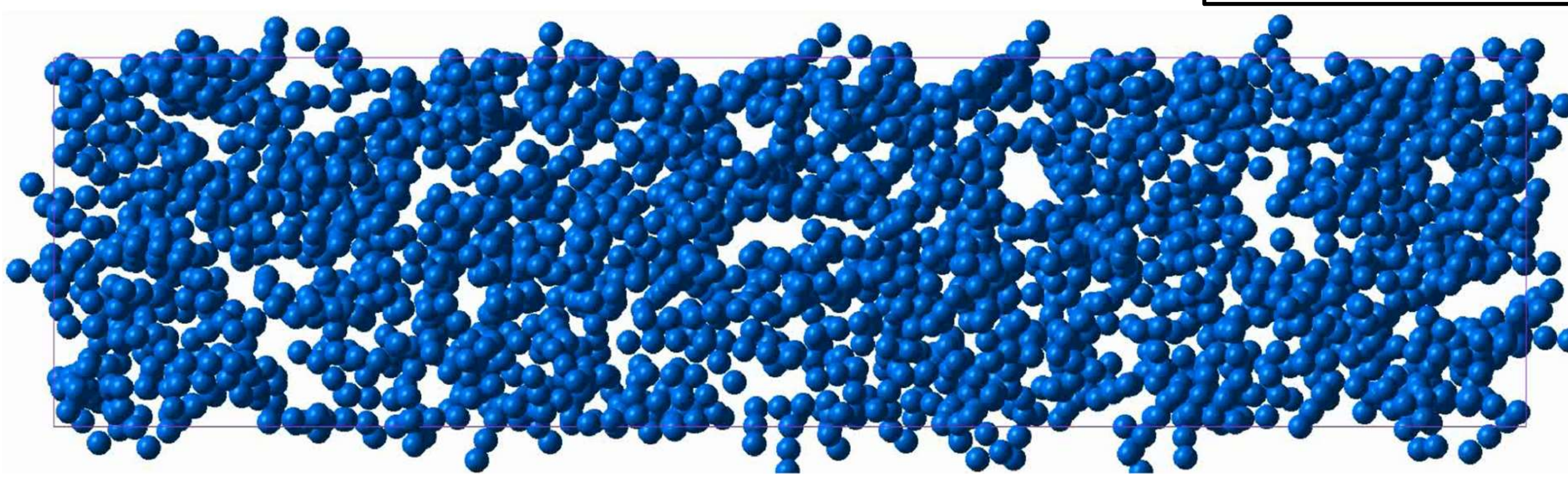
$$m_i \frac{d\mathbf{v}_i}{dt} = \sum \mathbf{F}_{ij}^{Att.} + \mathbf{F}_{ij}^{D} + \mathbf{F}_{ij}^{R}$$





$$\mathbf{F}_{ij}^{Att.} = \Gamma\kappa(e^{-\kappa r_{ij}} - e^{-2\kappa r_{ij}}).\mathbf{e}_{ij}$$

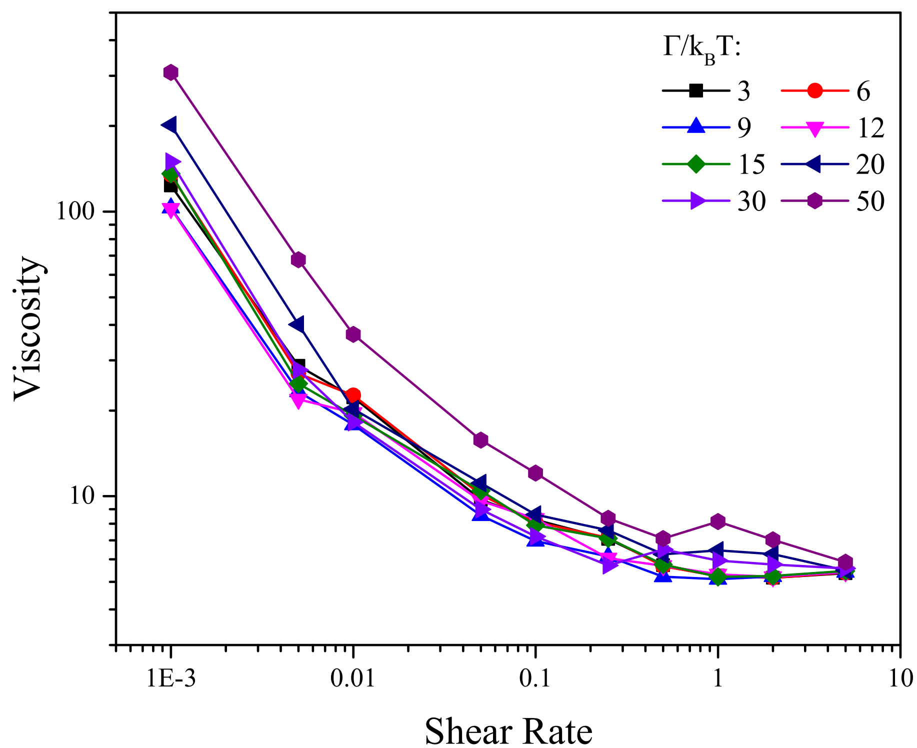


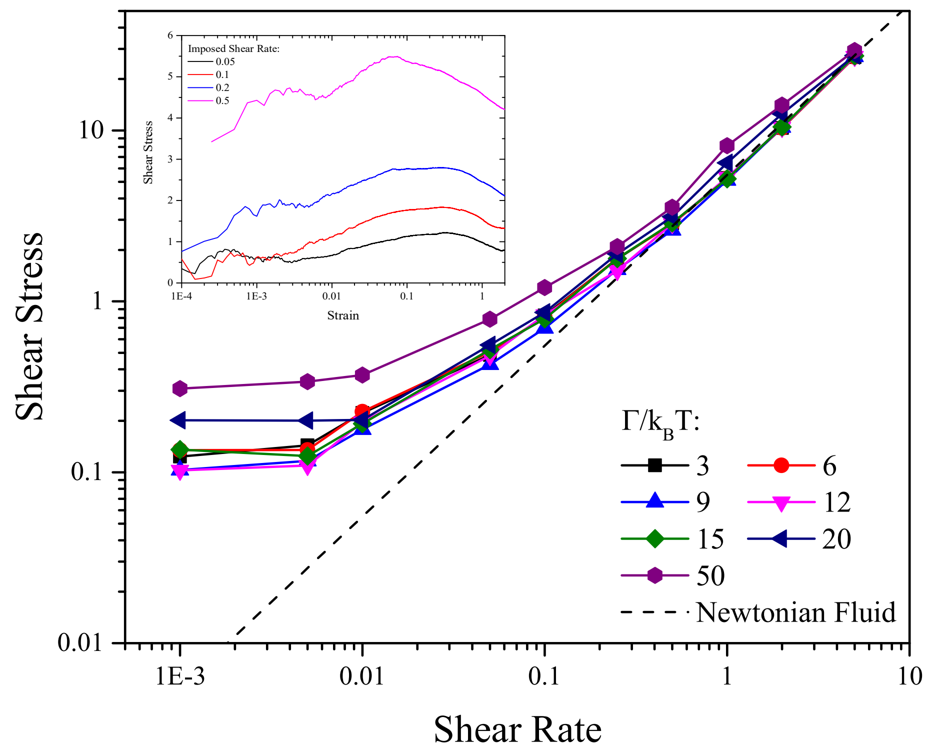


Jamali, McKinley and Armstrong, *Physical Review Letters*, 2017
Boromand, **Jamali** and Maia, *Soft Matter*, 2017
Jamali, McKinley and Armstrong, *Soft Matter*, submitted

Flow Curve of the Gelled Networks

- Our gel shows yielding behavior with increasing yield stress as a function of attraction between the primary particles
- Stress overshoot at the inception of shear flow is observed





Jamali, McKinley and Armstrong, *Physical Review Letters*, 2017

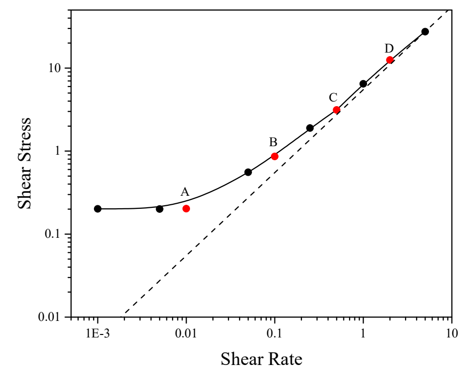
Morphology (Simulation vs. Experiment)

A

$\dot{\gamma} = 0.01$

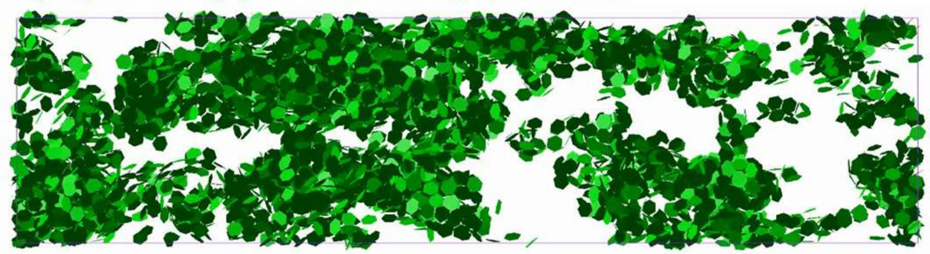
B

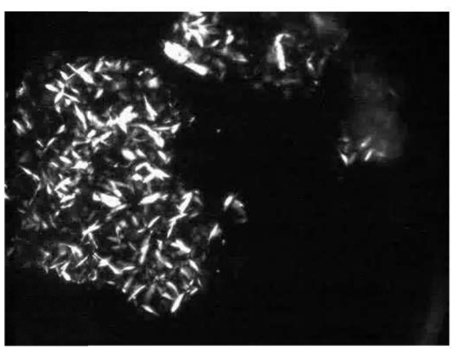
$\dot{\gamma} = 0.1$



C

$\dot{\gamma} = 0.5$





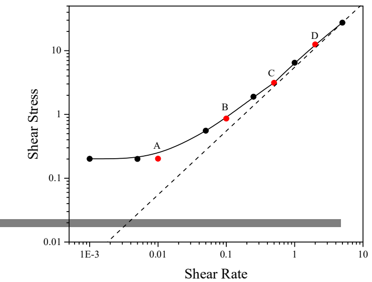
D

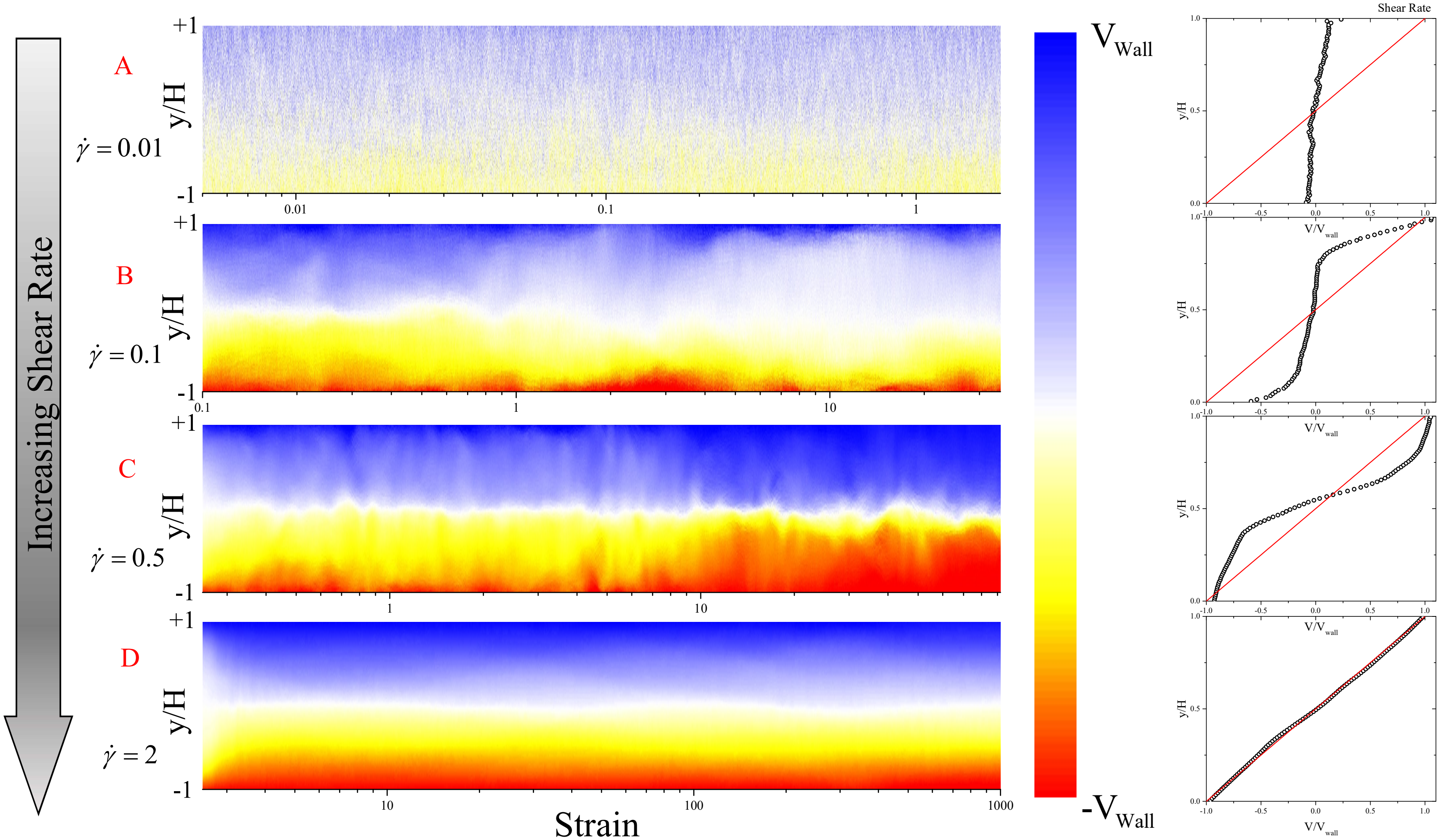
$\dot{\gamma} = 2$

Christopher Dimitriou, PhD Thesis, Massachusetts Institute of Technology, 2013

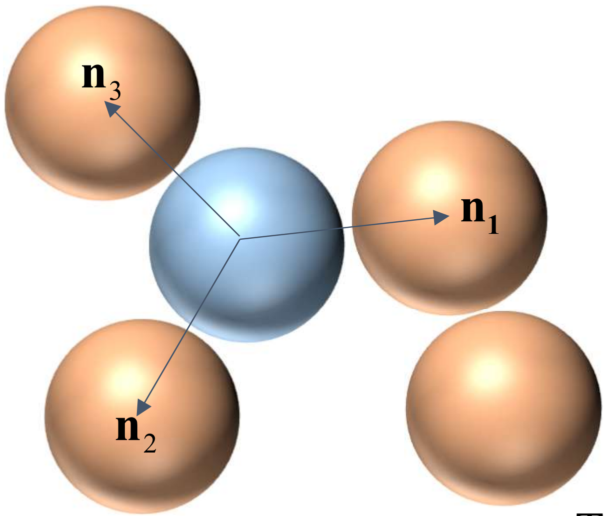
Jamali, McKinley and Armstrong, *Physical Review Letters*, 2017

Flow Heterogeneity from STD





Fabric Tensor: a Mathematical Representation of the Microstructural



Spatial arrangement of a particle's neighbors

$$\mathbf{Z}^p = \sum_i \mathbf{n}_i \otimes \mathbf{n}_i$$

Microstructure and anisotropy of the network

$$\mathbf{Z} = \frac{1}{N}\sum_{p=1}^{N} \mathbf{Z}^p$$

Trace of **Z** gives the number of links (coordination number)

$$\mathbf{Z} = \begin{pmatrix} Z_{xx} & Z_{xy} & Z_{xz} \\ Z_{xy} & Z_{yy} & Z_{yz} \\ Z_{xz} & Z_{yz} & Z_{zz} \end{pmatrix}$$

$$\mathrm{tr}\mathbf{Z} = Z_{xx} + Z_{yy} + Z_{zz}$$

Deviator tensor gives information about the anisotropy

$$\zeta = \mathbf{Z} - \frac{tr\mathbf{Z}}{3}\mathbf{I}$$

"Rheology without Morphology is Theology, and Morphology without Rheology is just Zoology"

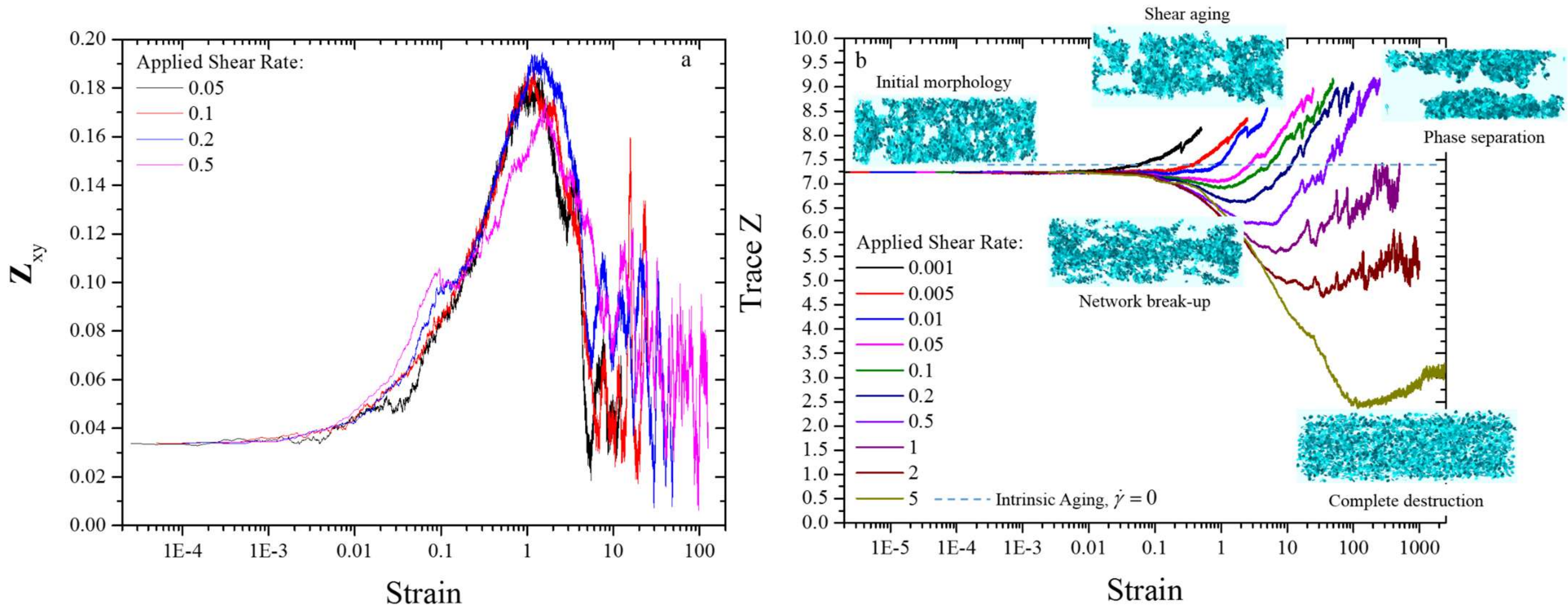
R. Stein (U. Mass-Amherst)

Olsen and Kamrin, *Soft Matter*, 2015, pp: 3875-3883

Jamali, McKinley and Armstrong, *Physical Review Letters*, 2017

Microstructural Evolution under Deformation

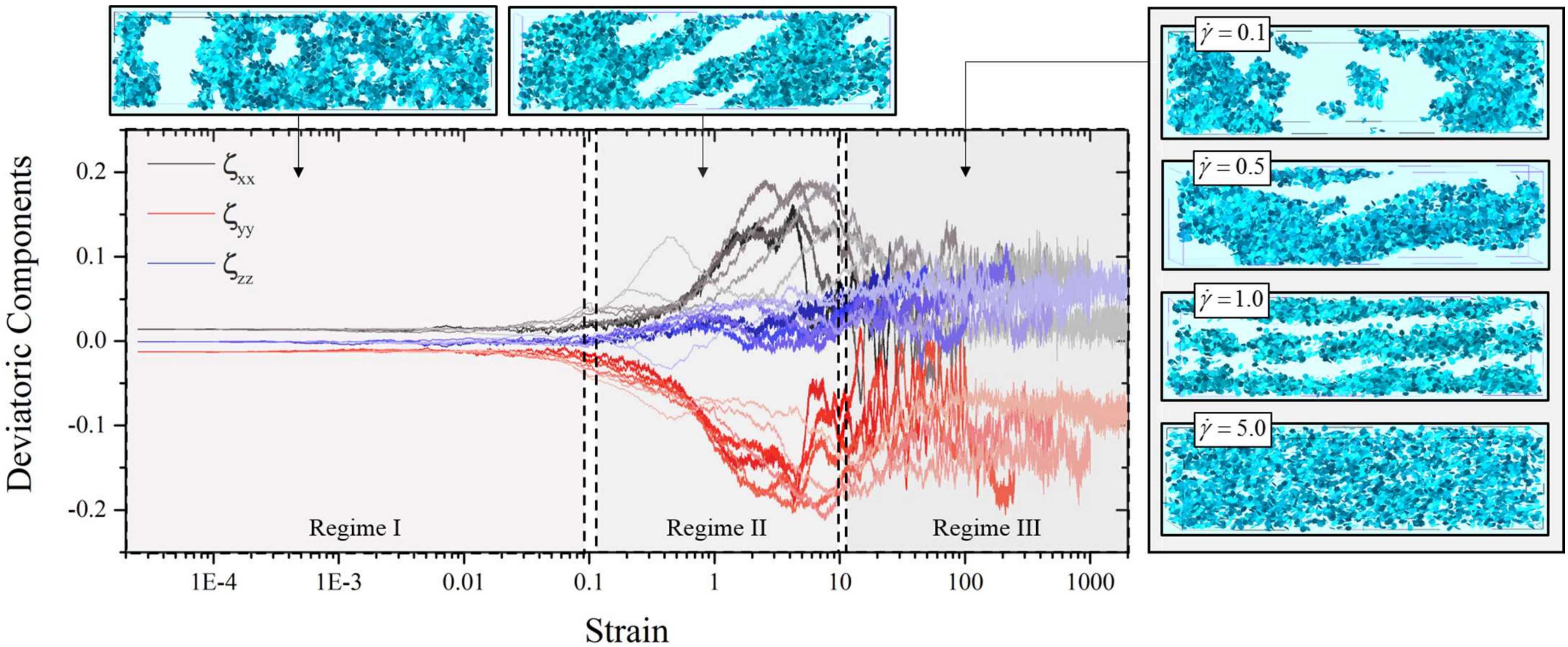
- Shear component of the fabric tensor shows the same transition as the stress overshoot at the flow start up
- Trace of fabric shows 3 distinct regimes: aging, rejuvenation and break-up



Jamali, McKinley and Armstrong, *Physical Review Letters*, 2017

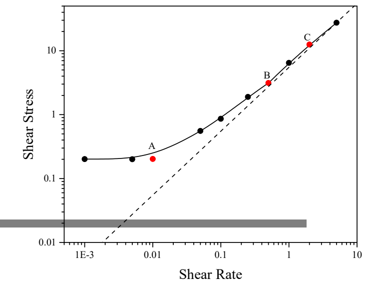
How about Microstructural Anisotropy?

- Deviatoric components of the fabric tensor exhibit a universal behavior regardless of imposed rate with regards to particle network anisotropy



Jamali, McKinley and Armstrong, *Physical Review Letters*, 2017

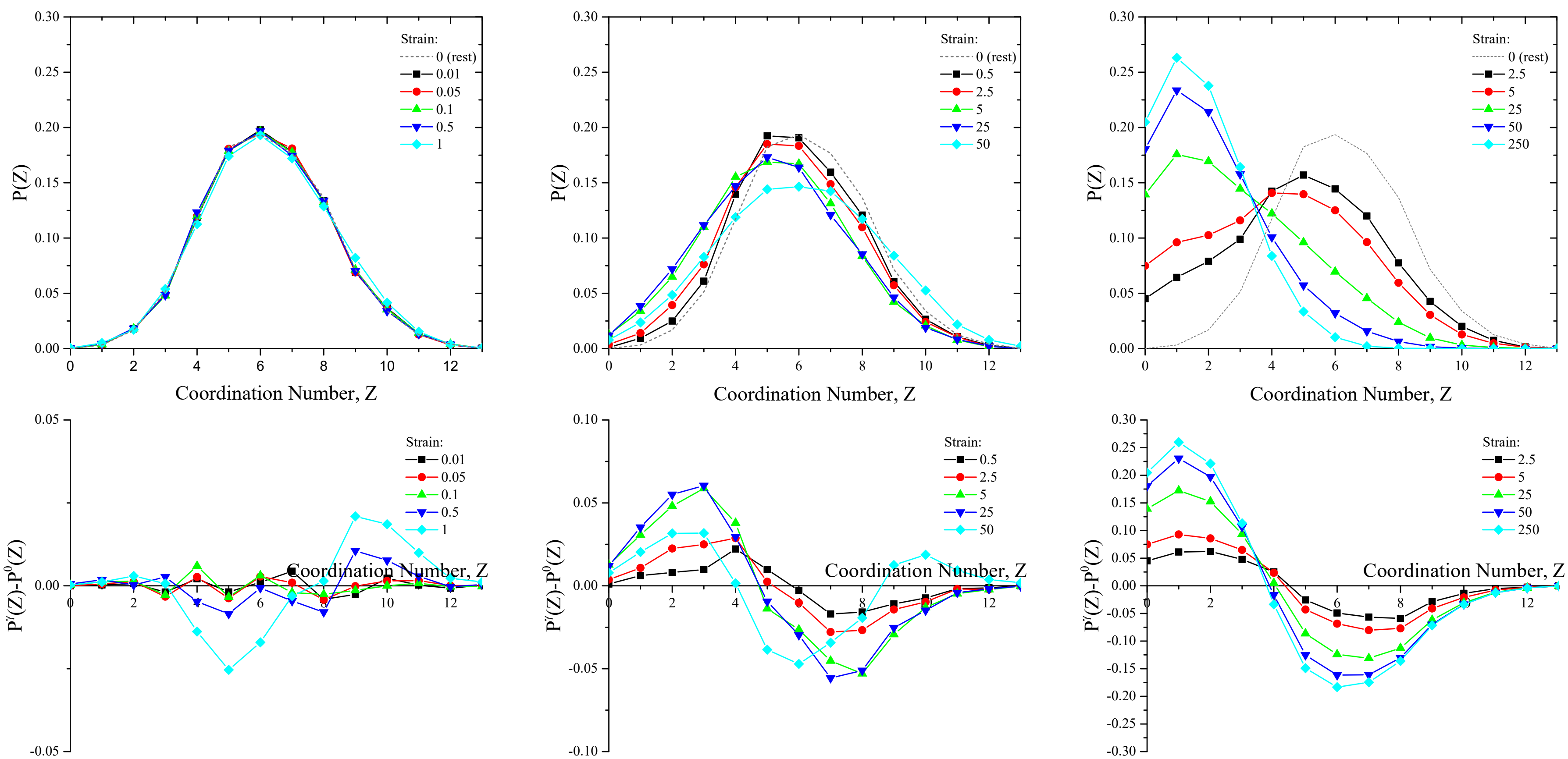
Evolution of Particle-level Fabric



$\dot{\gamma} = 0.01$ Shear Aging Compaction

$\dot{\gamma} = 0.5$ Shear Rejuvenation

$\dot{\gamma} = 2$ Fluidization

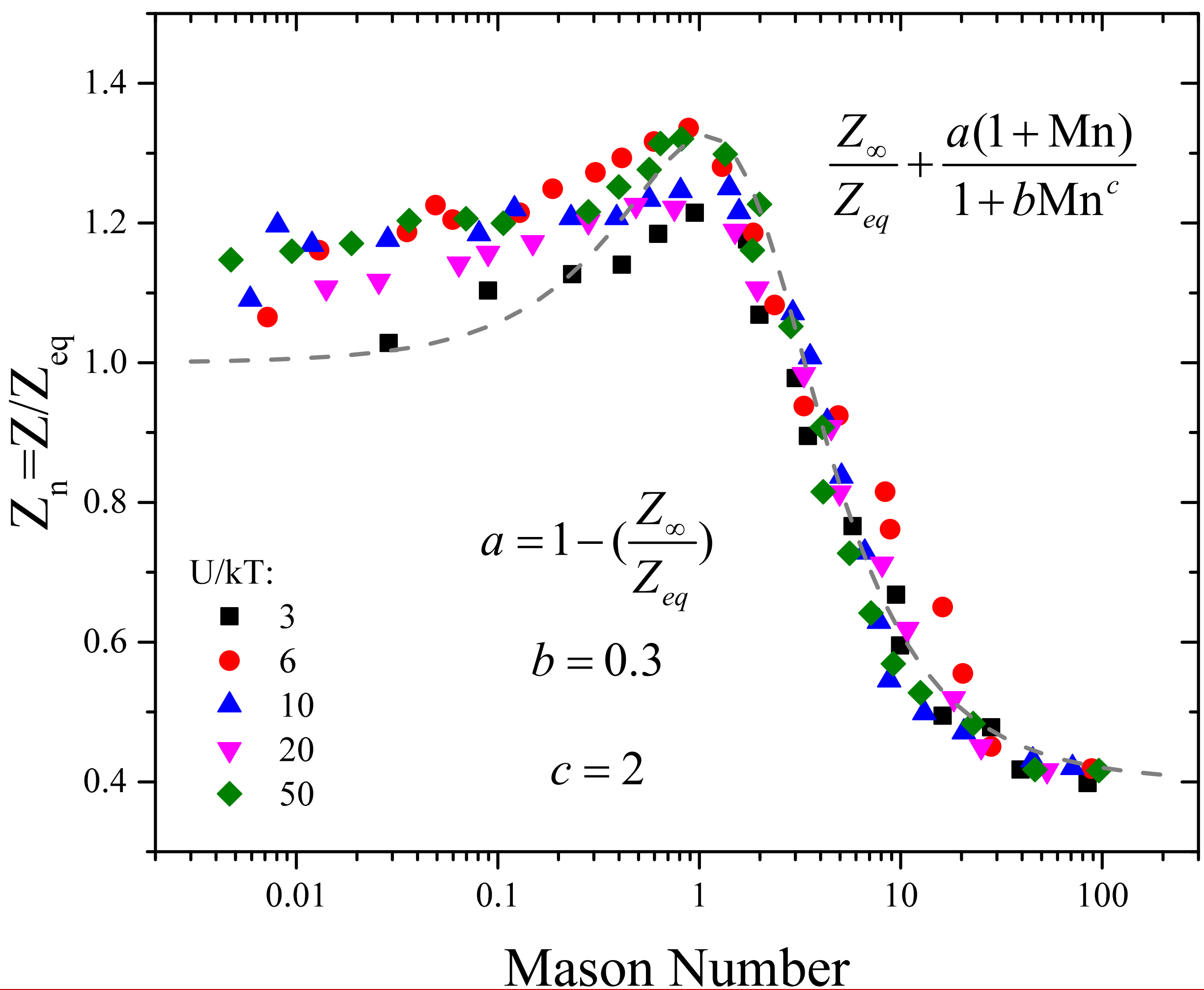


Northeastern University

Jamali, McKinley and Armstrong, *Physical Review Letters*, 2017

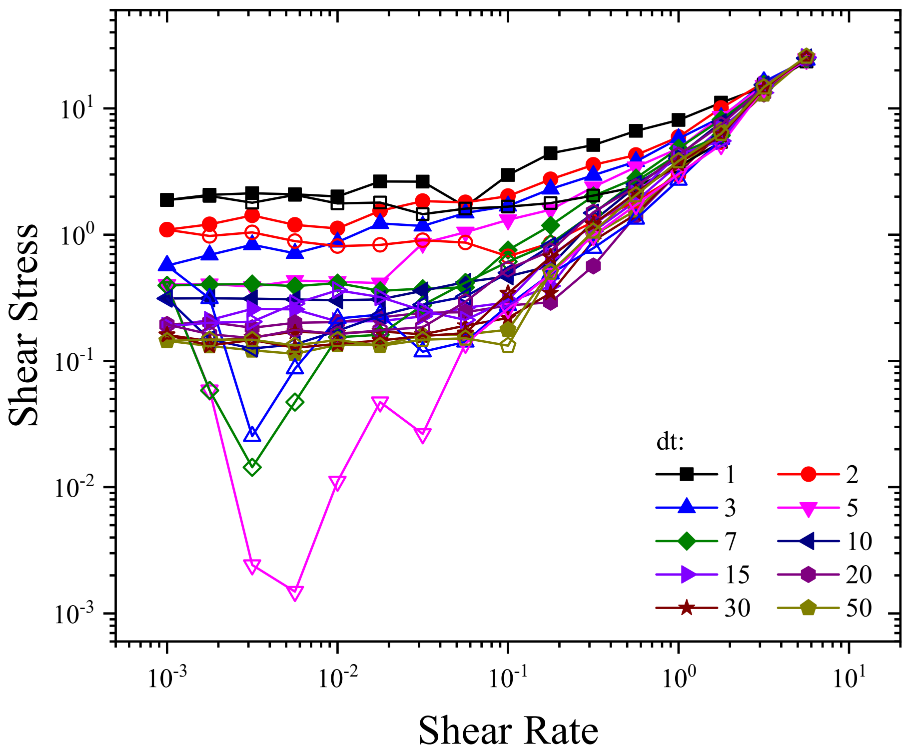
Jamali, McKinley and Armstrong, *Soft Matter*, submitted

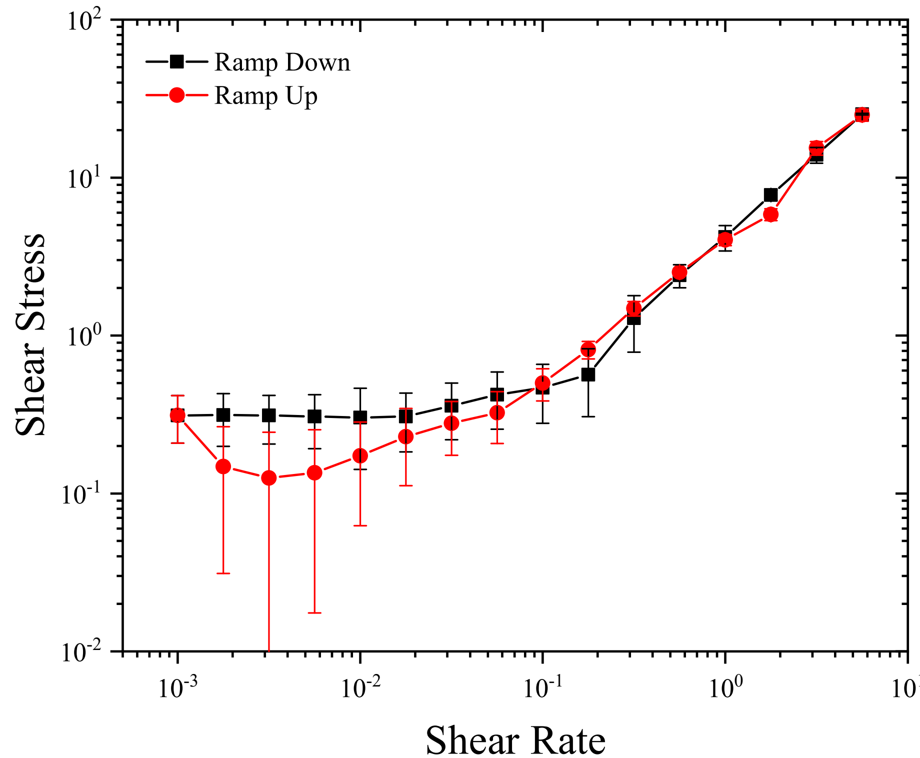
Structure under Flow



Flow Hysteresis

- Protocol:
  - Start from highest shear rate (Mn>>1)
  - Ramp down followed by ramp up, with time dt at each shear rate
  - Explore effect of attraction strength on thixotropic time

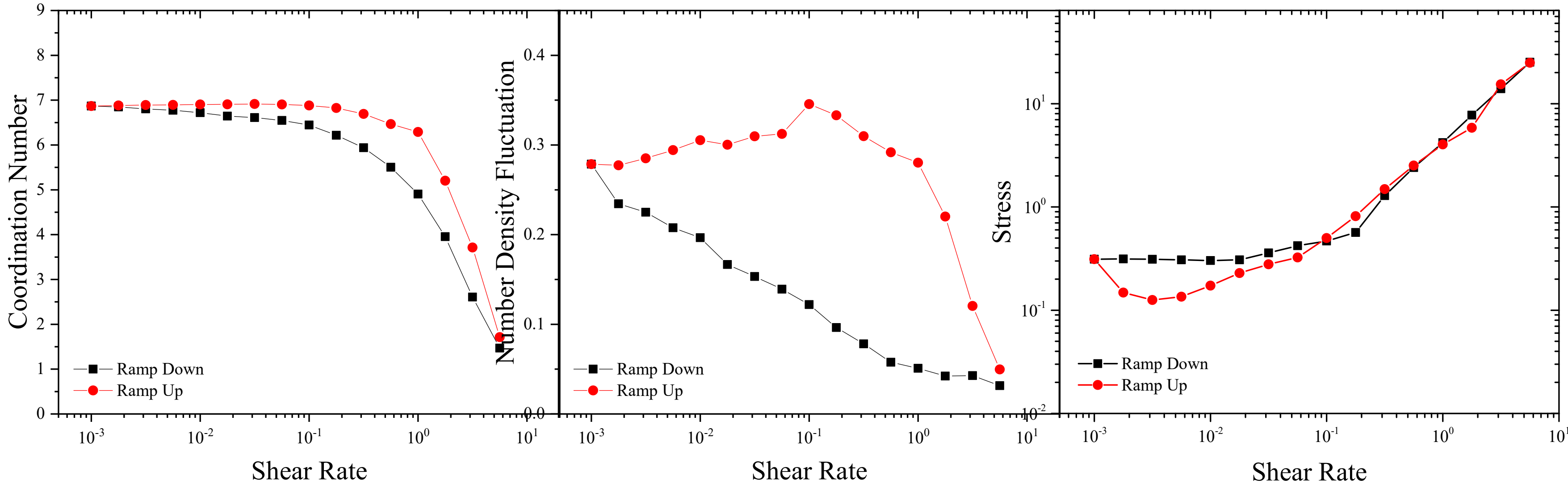




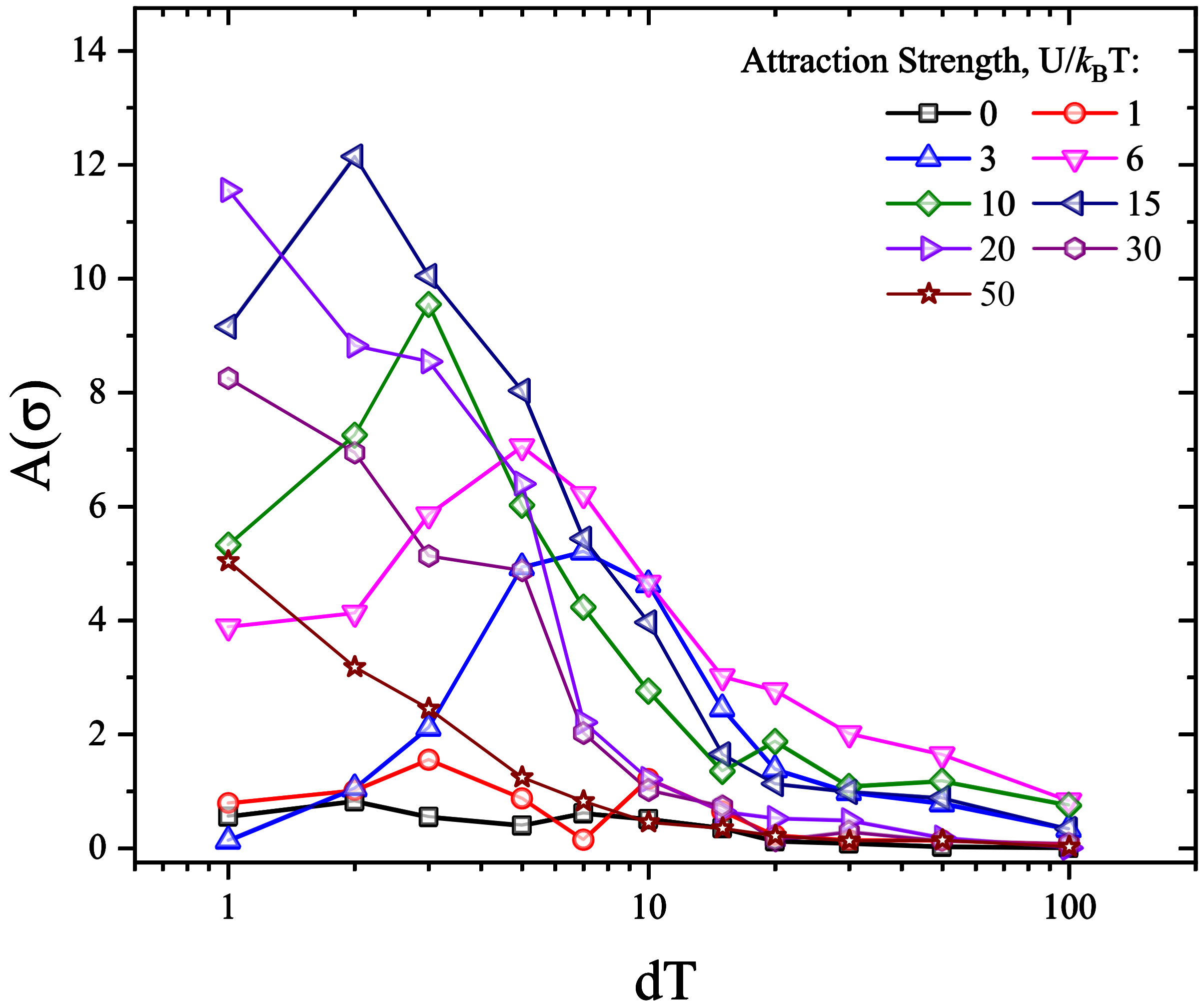
Hysteresis area across scales

- Macro-scale: Stress
- Meso-scale: Number density fluctuation
- Micro-scale: Average coordination number

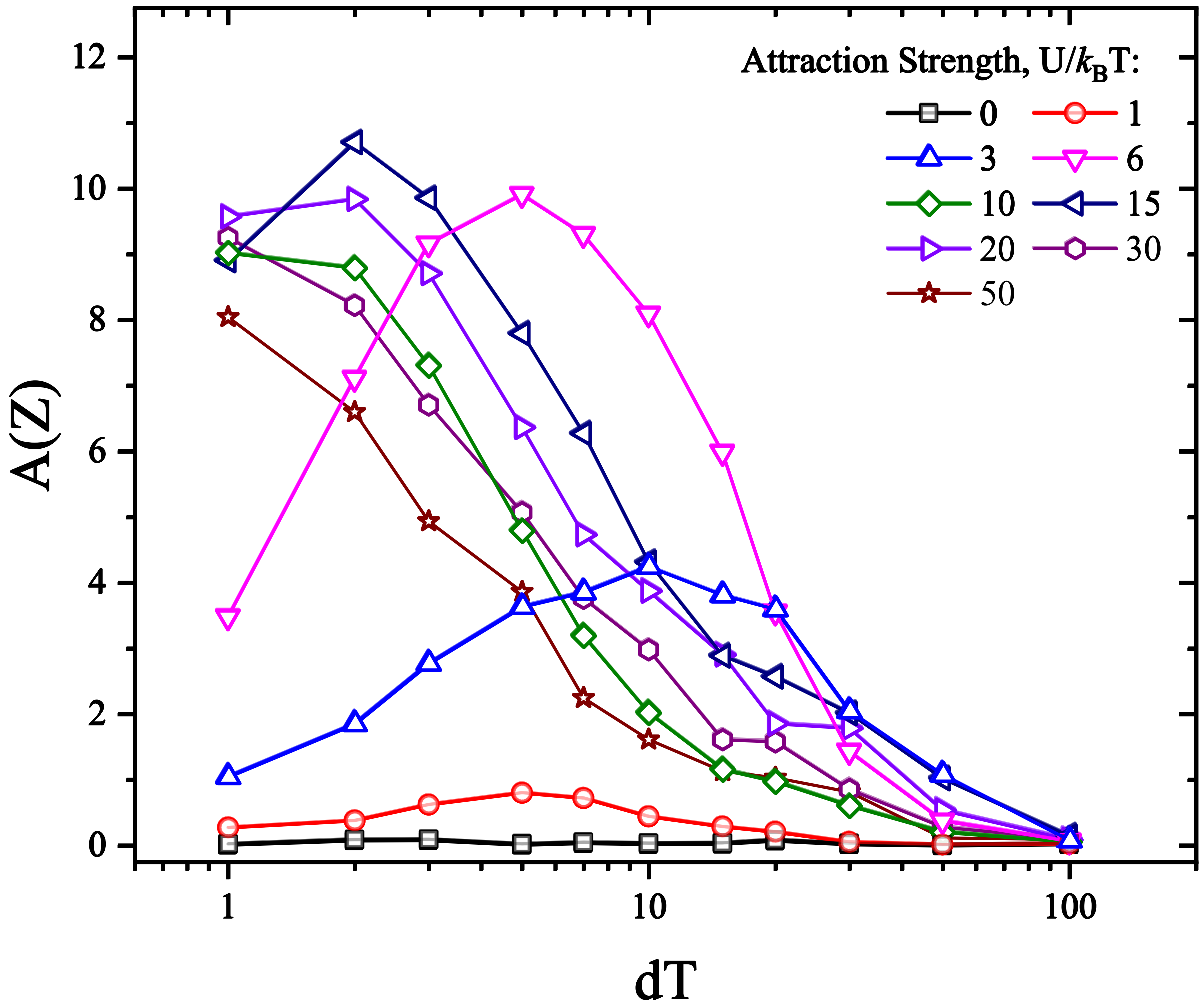
$$A_Z \equiv \int_{\dot{\gamma}_{\min}}^{\dot{\gamma}_{\max}} \left| Z(\dot{\gamma}) \right| d\left(\log \dot{\gamma}\right) \qquad A_{NDF} \equiv \int_{\dot{\gamma}_{\min}}^{\dot{\gamma}_{\max}} \left| \frac{\langle N^2 \rangle - \langle N \rangle^2}{\langle N \rangle} \right| d\left(\log \dot{\gamma}\right) \qquad A_\sigma \equiv \int_{\dot{\gamma}_{\min}}^{\dot{\gamma}_{\max}} \left| \Delta\sigma(\dot{\gamma}) \right| d\left(\log \dot{\gamma}\right)$$



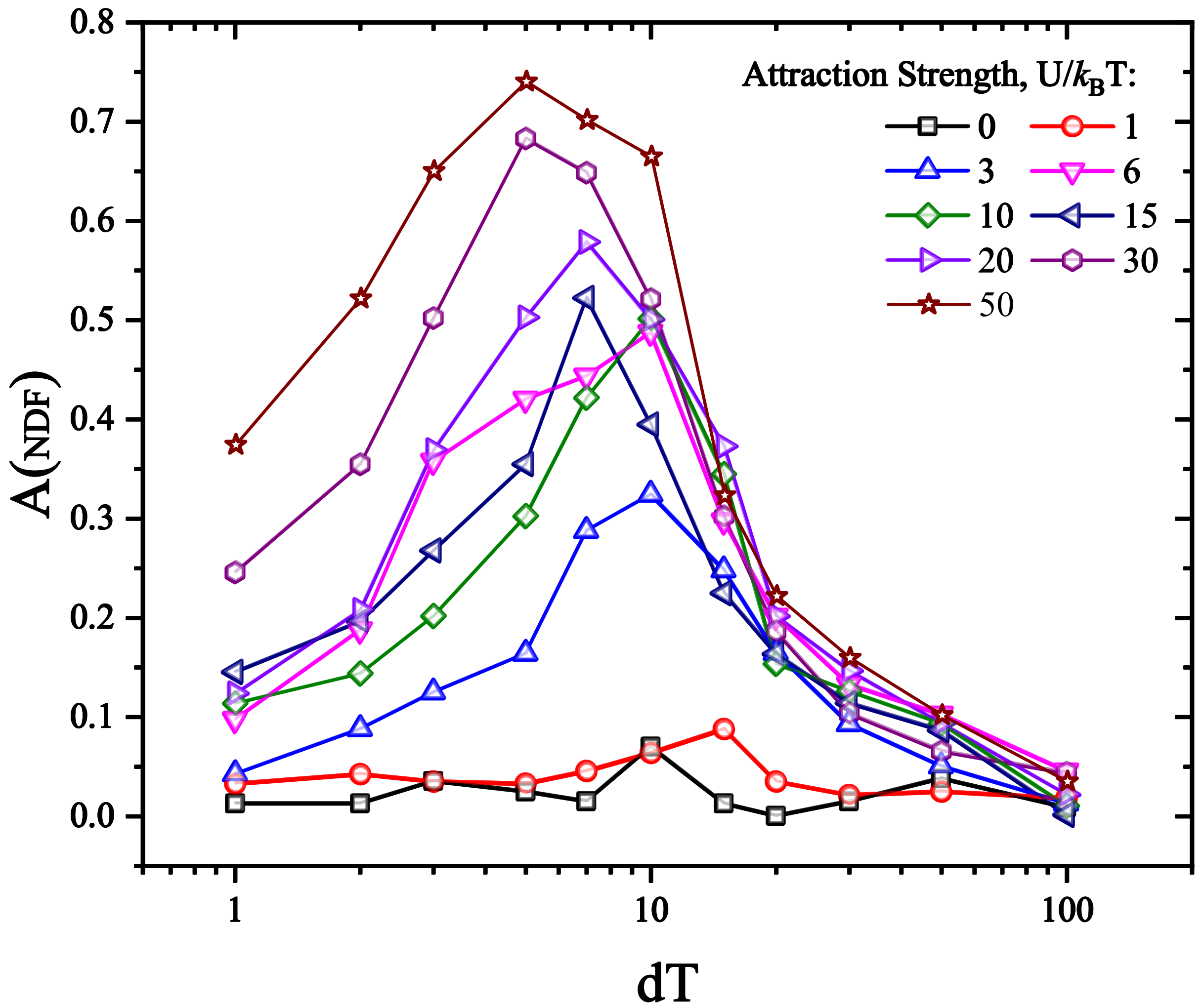
(Macroscopic) Thixotropic time



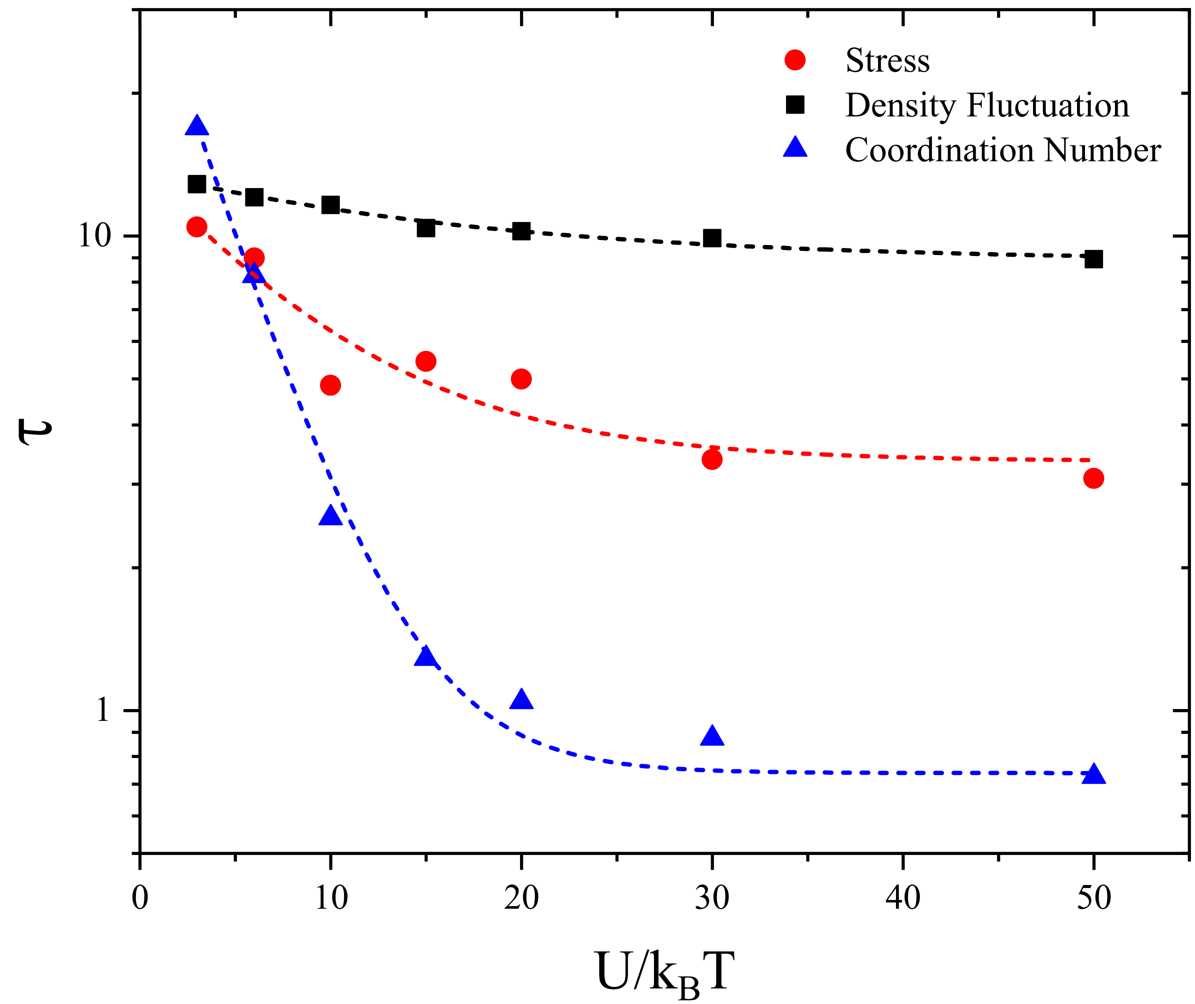
(Microscopic) Thixotropic time



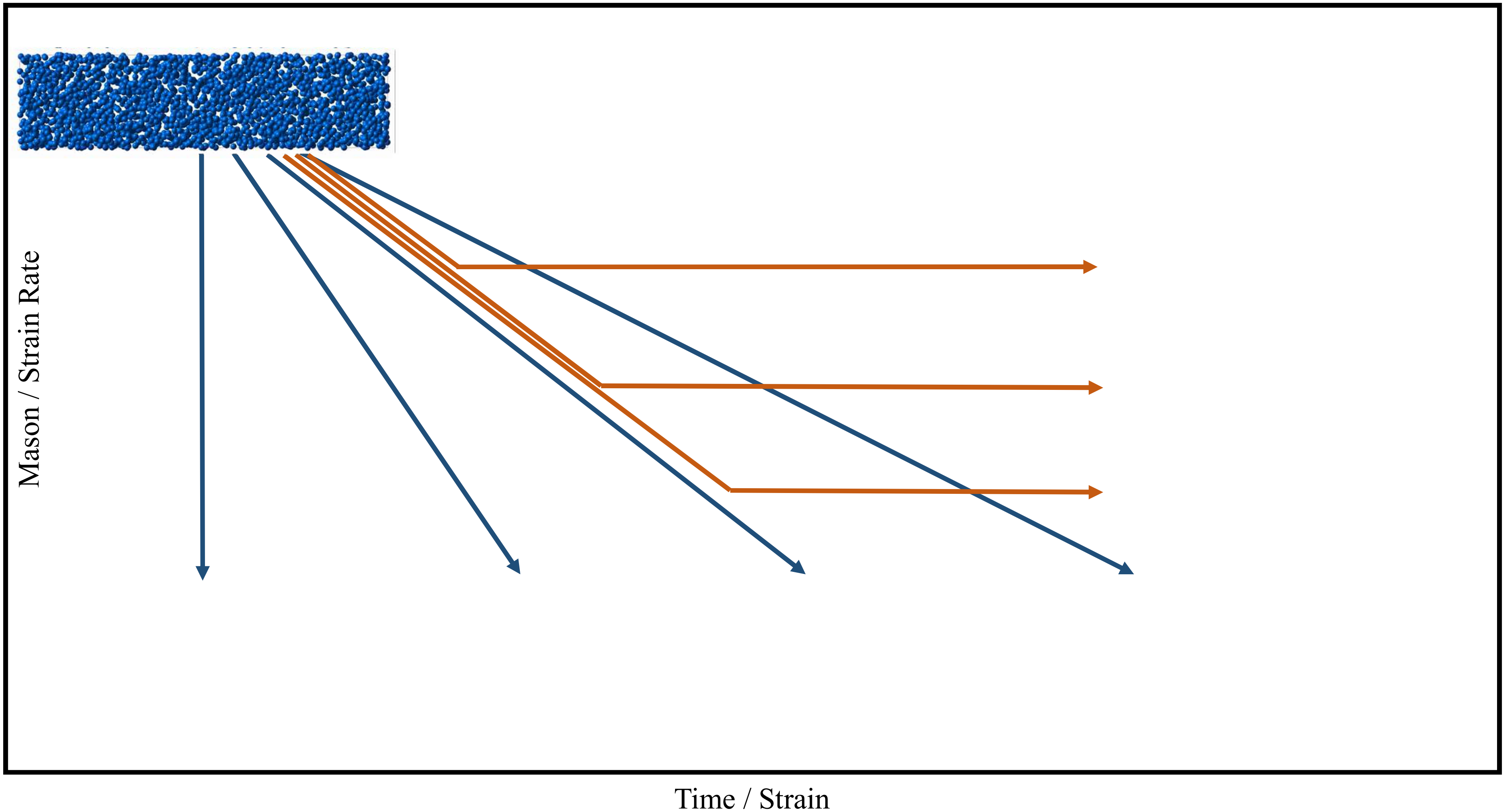
(Mesoscopic) Thixotropic time



Thixotropic timescale vs. Attraction



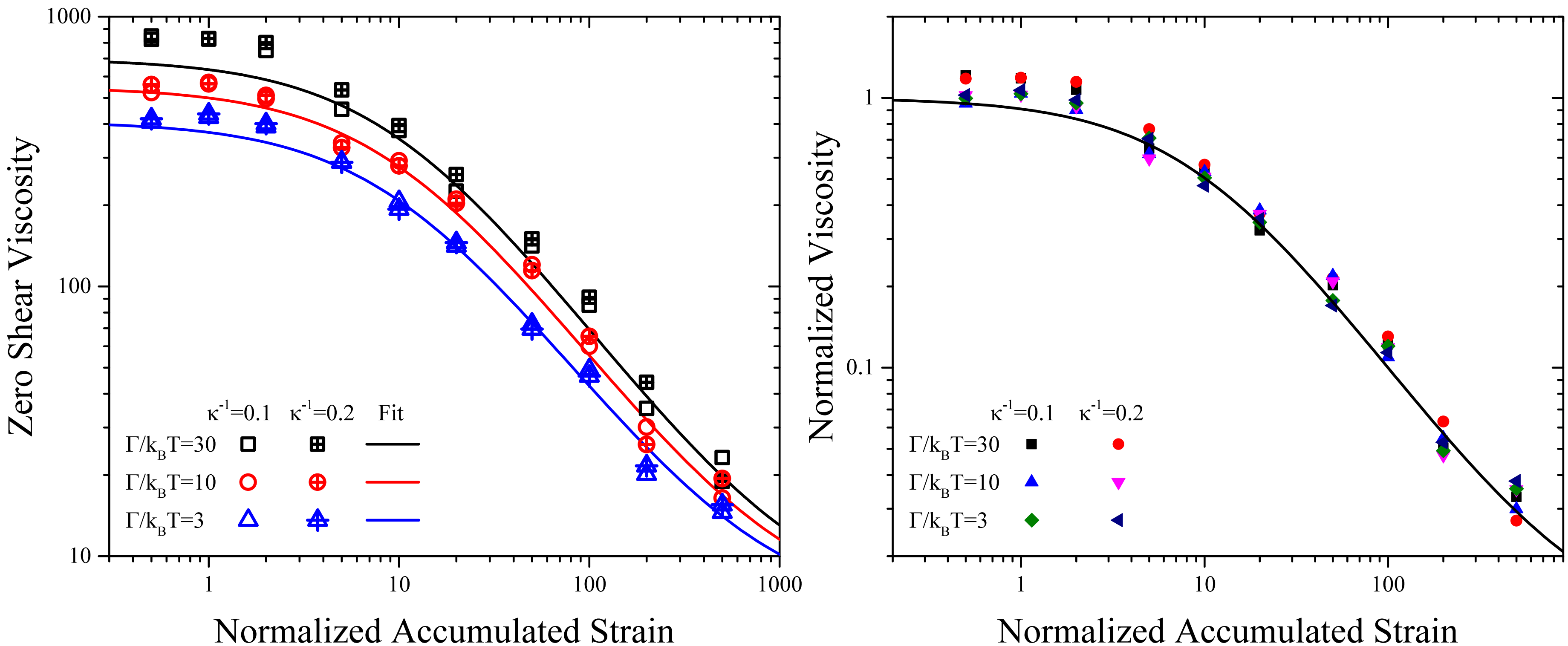
Time-Mason-Transformation



Jamali, McKinley and Armstrong, “Time-Mason-Transformation”, *in prep.*

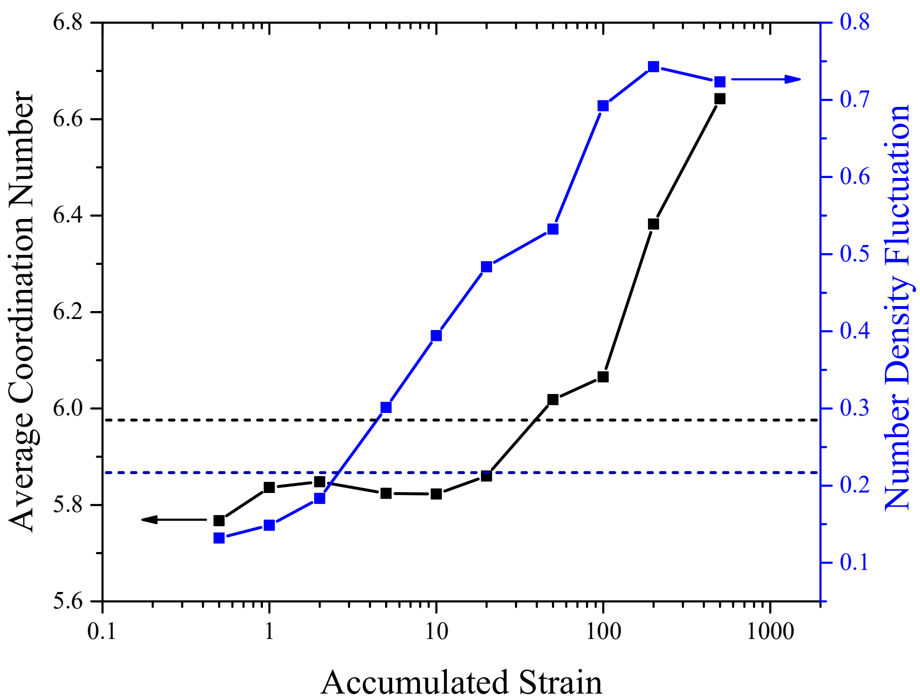
Viscosity-History relationship

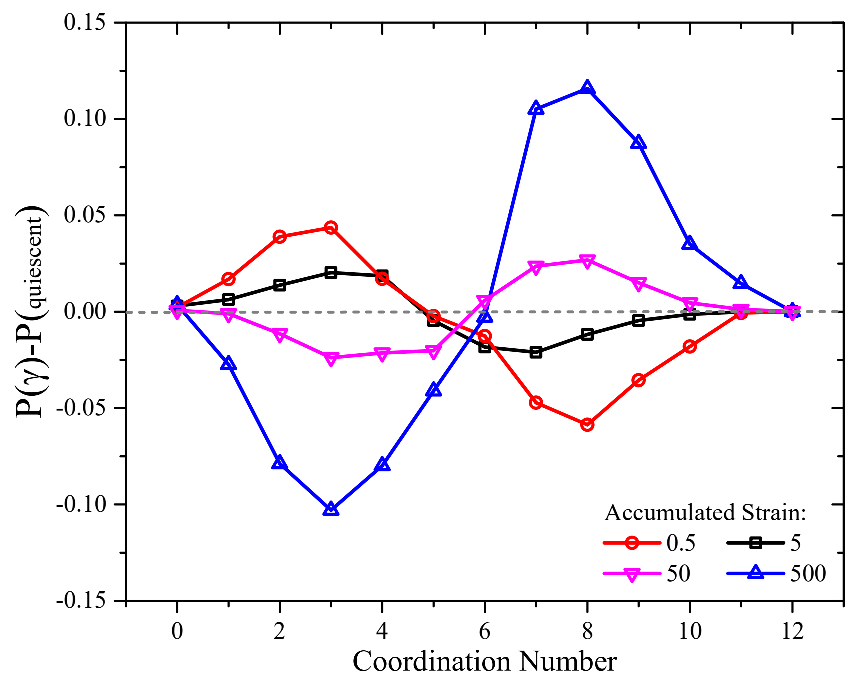
$$\frac{\eta_0 - \eta_\infty}{\eta - \eta_\infty} = 1 + \left(\frac{\kappa^{-1}}{a}\right)\gamma$$



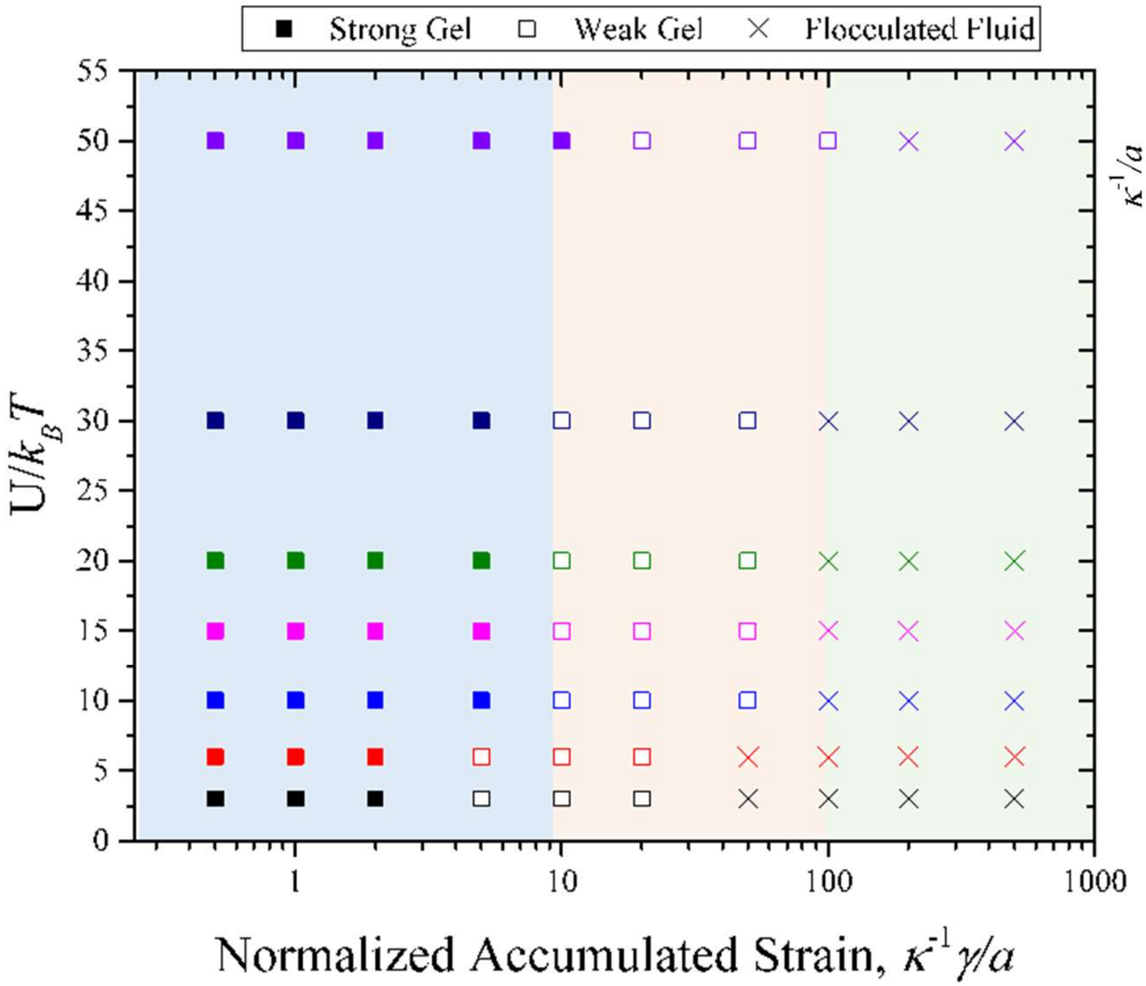
(Micro/Meso) Structure-History

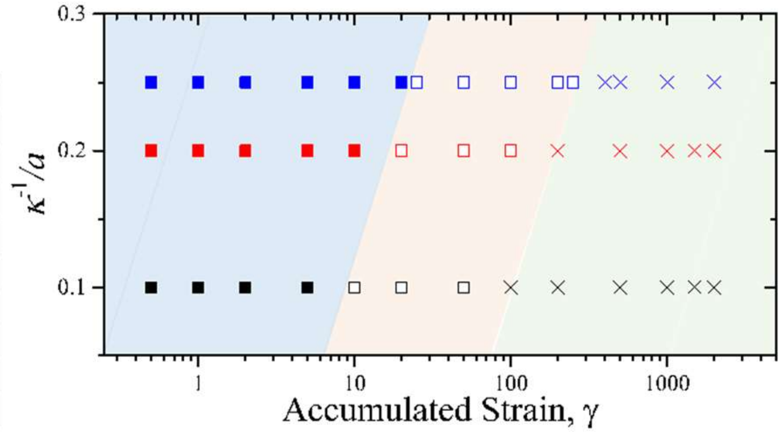
- 3 regimes:
  - Strong gels
  - Weak gels
  - Flocculated fluids

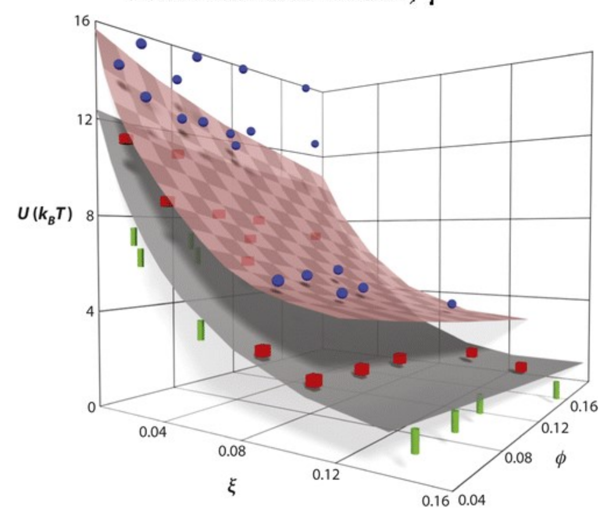




Effect of Particle Characteristics

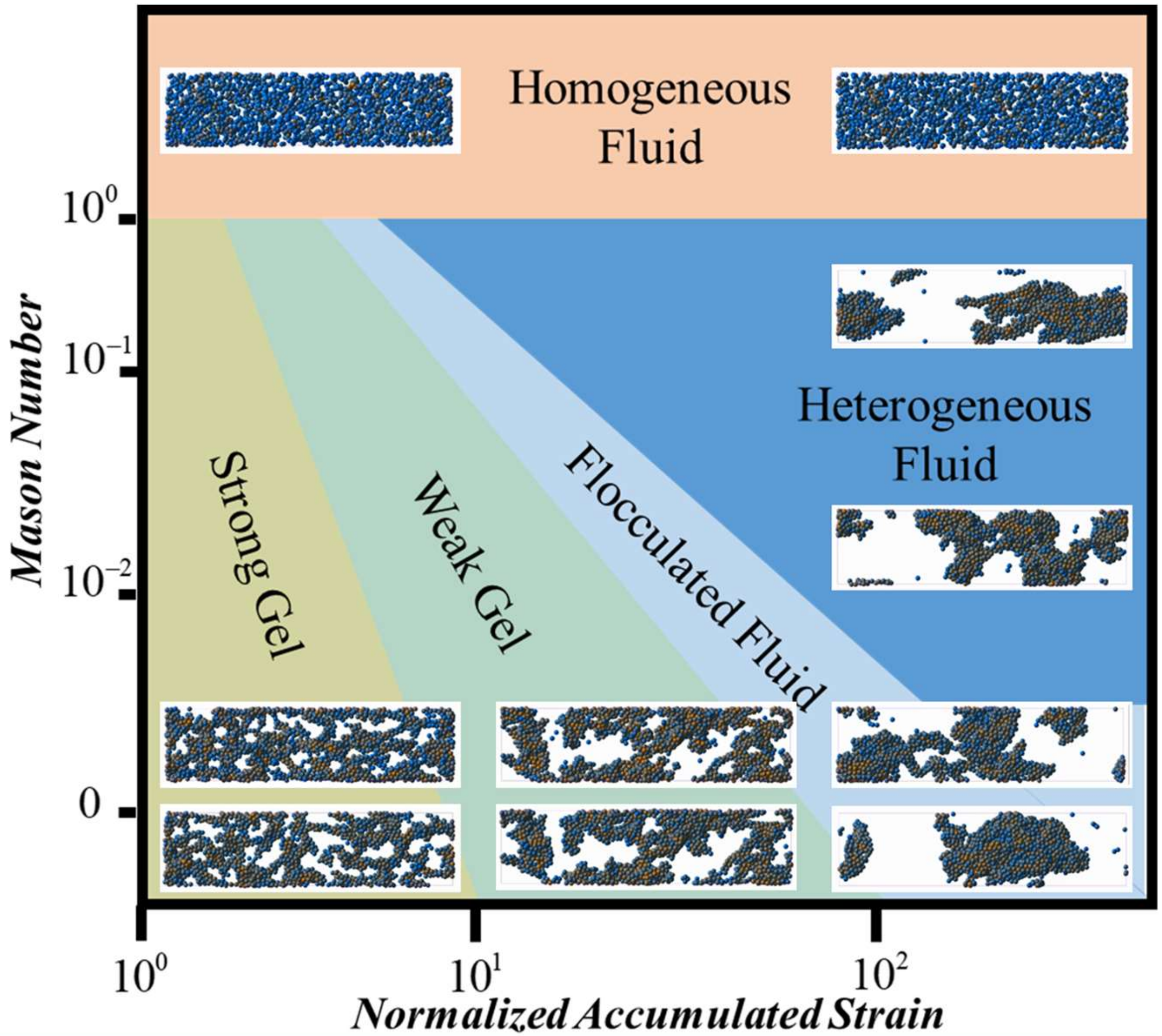






Lu (陸述義) PJ and Weitz DA. 2013.
Annu. Rev. Condens. Matter Phys. 4:217–233

Time-Mason-Transformation



Vorticity-Aligned Structures