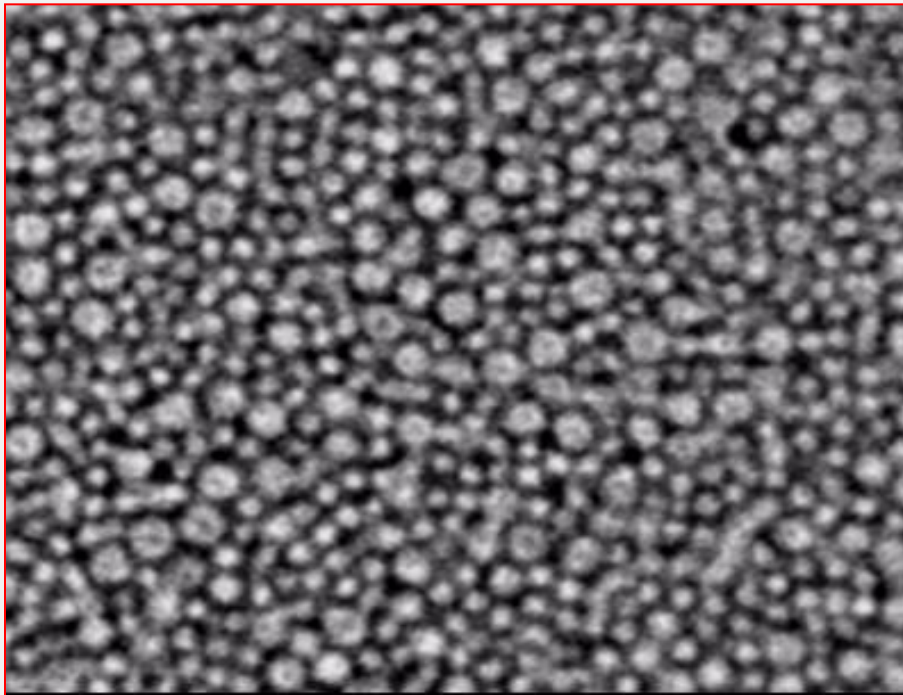


Yielding of a Soft Glass :Signatures of a Non-Equilibrium Phase Transition

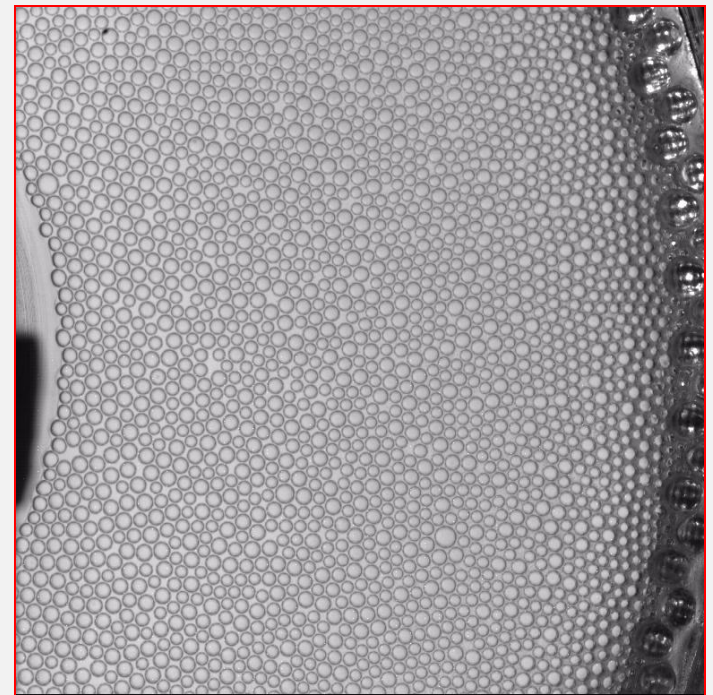
Ajay Sood

Department of Physics

Indian Institute of Science, Bangalore ,India



Binary Colloidal Glass



2D Amorphous Bubble Raft

Collaborators

Hima Manasa (JNC, now at IBS Seoul)

Shreyas Gokhale (IISc, Now at MIT)

Srimayee Mukherjee(JNC)

Neelima Kandula (JNC)

Prof Rajesh Ganapathy(JNC)

More recent experiments on 2D film : Interfacial rheology

Pradip Bera

Dr Ajoy Kandar

Dr R Krishnaswamy

Basic facts of life.....

Solids (Atomic crystals, Metallic glasses, Dense suspensions , gels , foams) exhibit yielding and plastic flow at sufficiently large external stresses.

Crystalline Materials : Yielding via motion of well defined topological defects called dislocations.

Local yield events (dislocations) move collectively and self organize into avalanches that follow power law scaling . Many examples.....

Crystal Plasticity

Atomic

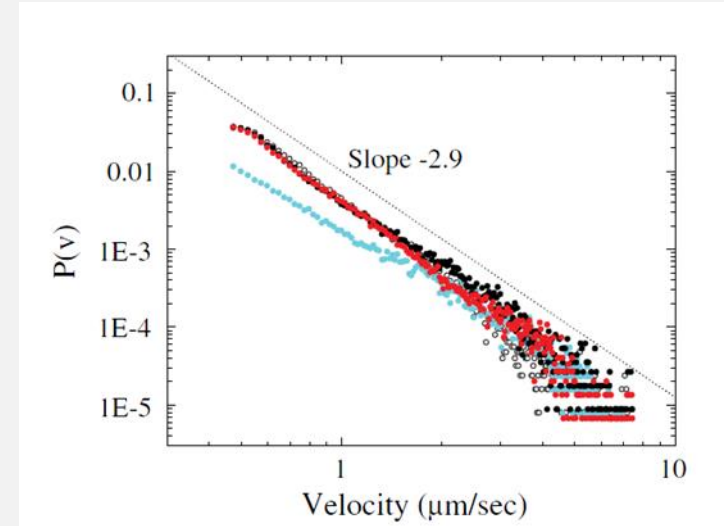
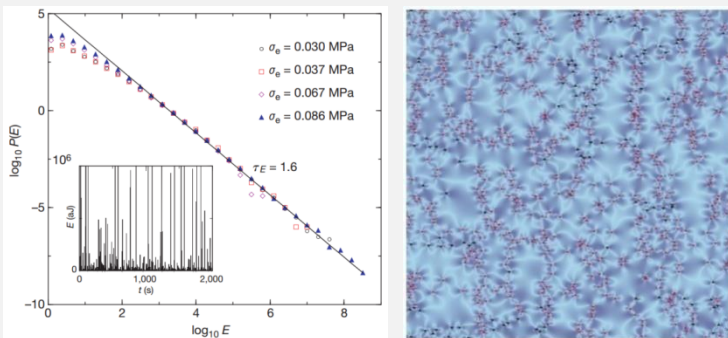
Soft

Intermittent dislocation flow in viscoplastic deformation

M.-Carmen Miguel*†, Alessandro Vespignani*, Stefano Zapperi‡, Jérôme Weiss§ & Jean-Robert Grasso||

NATURE | VOL 410 | 5 APRIL 2001 |

Acoustic emission experiments on bulk single crystals of ice

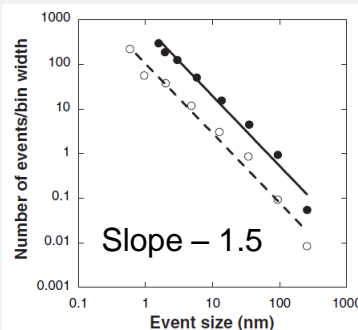


Pertsinidis and Ling, New J Phys 7,33 (2005)

Dislocation velocities exhibit power laws

Scale-Free Intermittent Flow in Crystal Plasticity

Dennis M. Dimiduk, *et al.*
Science 312, 1188 (2006);



Nano-indentation experiments on Ni microcrystals

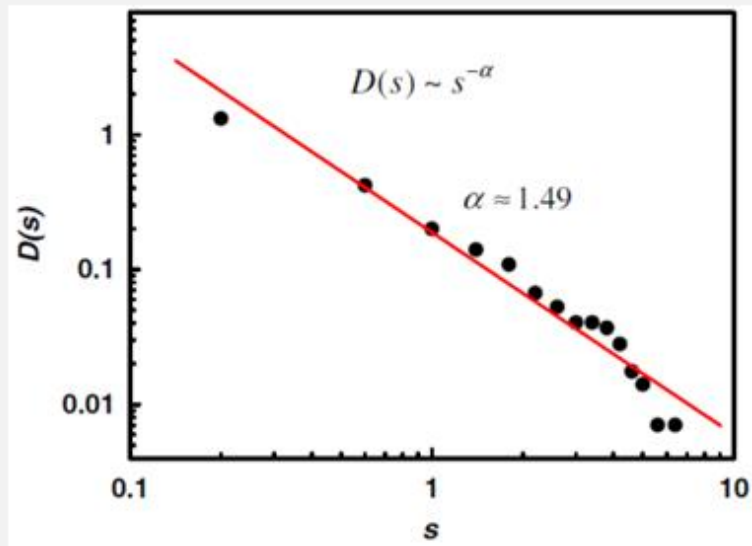
Amorphous Systems

ATOMIC SYSTEMS

PRL 105, 035501 (2010) PHYSICAL REVIEW LETTERS WEEK ENDING
16 JULY 2010

Plasticity of Ductile Metallic Glasses: A Self-Organized Critical State

B. A. Sun, H. B. Yu, W. Jiao, H. Y. Bai, D. Q. Zhao, and W. H. Wang*



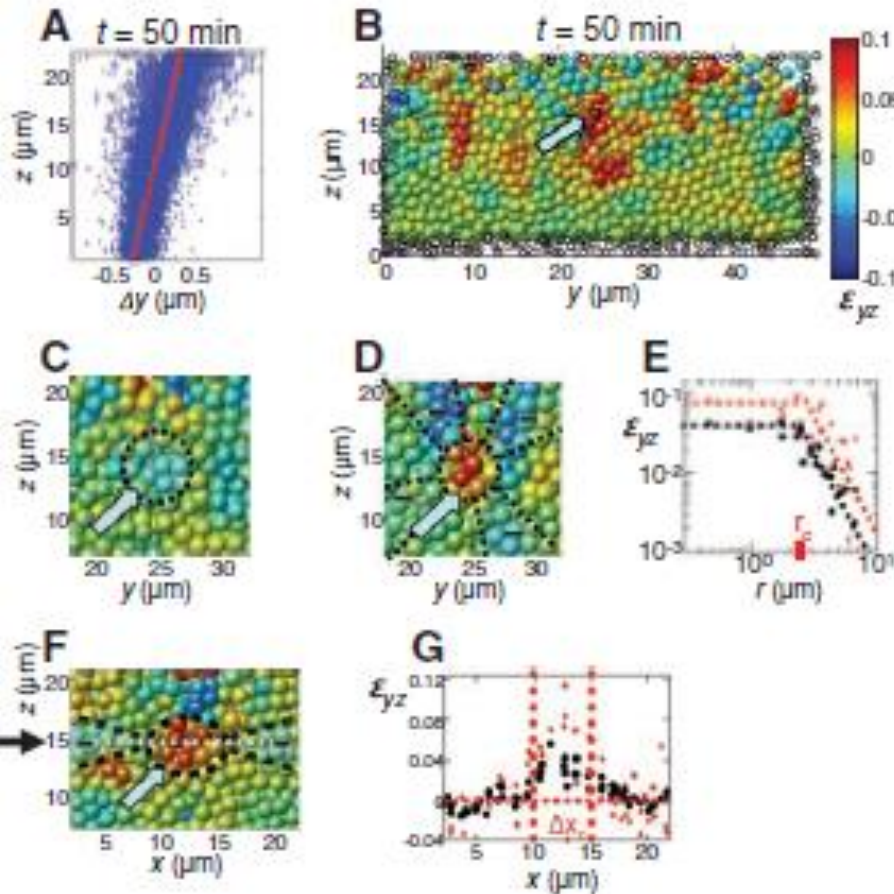
Log-log plot of the density distribution $D(s)$ vs normalized stress drop magnitude s for Cu47:5Zr47:5Al5,

Experiments on colloidal glasses: Steady Shear

Structural Rearrangements That Govern Flow in Colloidal Glasses

Peter Schall,^{1,2*} David A. Weitz,^{2,3} Frans Spaepen²

Science 318,1895(2007)



Fourfold pattern in the Spatial autocorrelation of the local strain field indicate that the dominant deformation mechanisms in glass are STZs(See Falk and Langer , PRE (1998))that behave as Eshelby inclusions: local regions of high plastic Strain that couple elastically to the surrounding material.

Long-Range Strain Correlations in Sheared Colloidal Glasses

Vijayakumar Chikkadi,¹ Gerard Wegdam,¹ Daniel Bonn,¹ Bernard Nienhuis,² and Peter Schall¹

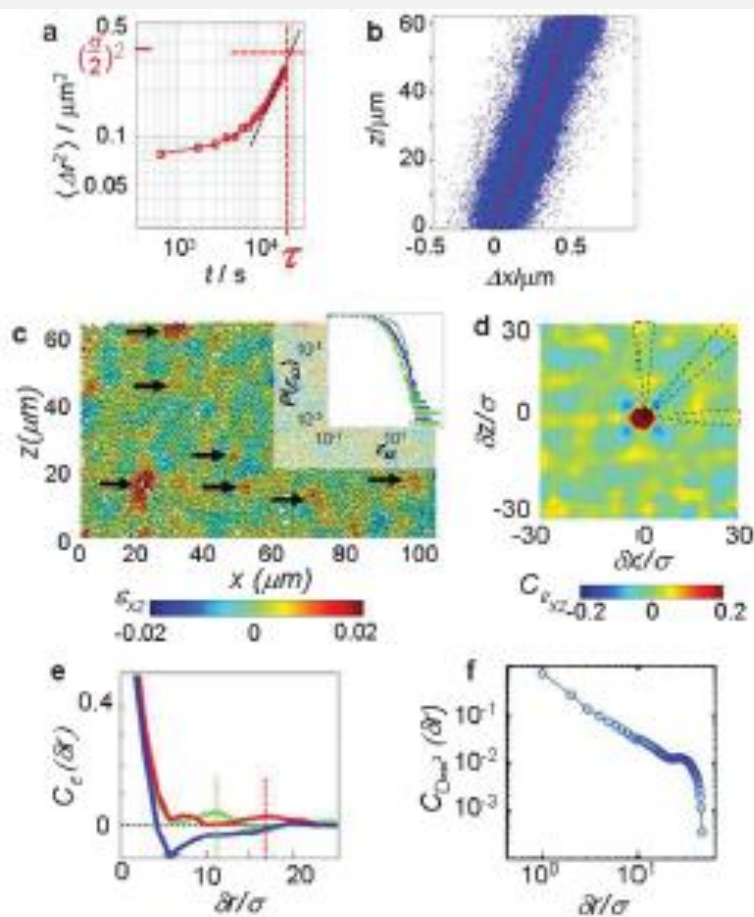


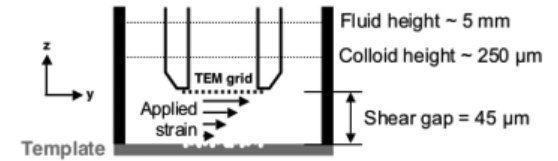
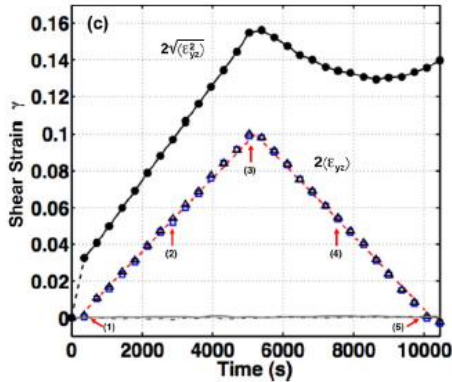
FIG. 1 (color). Homogeneous deformation at a shear rate of $\dot{\gamma} = 1.5 \times 10^{-5} \text{ s}^{-1}$. (a) Mean square displacement of the quiescent glass without applied shear. The structural relaxation time τ of the glass is estimated by extrapolating the mean square displacement in the diffusive regime with a line of slope 1 (dashed black line). (b) Displacements of individual particles (+) and average displacement (dashed line) along the shear direction during $\delta t = 10$ min of shear. (c) $7 \mu\text{m}$ thick reconstruction of the distribution of shear strain after $\delta t = 3$ min of shear. Particle color indicates the value of ϵ_{xz} . The inset in (c) shows the relative frequency of shear strain magnitudes ϵ_{xz} for time intervals $\delta t = 3$ (green stars) and 25 min (blue squares). (d) Angle-resolved spatial correlation $C_{\epsilon_{xz}}$ of the fluctuations of shear strain, in the x - z plane. (e) $C_{\epsilon_{xz}}$ as a function of distance averaged over angular wedges of 10° around the horizontal (red line), the vertical (green line), and the two diagonal directions (blue line). (f) Angle-averaged correlation function $C_{D_{\min}^2}$ as a function of distance in a double-logarithmic representation.

Local shear transformations in deformed and quiescent hard-sphere colloidal glasses

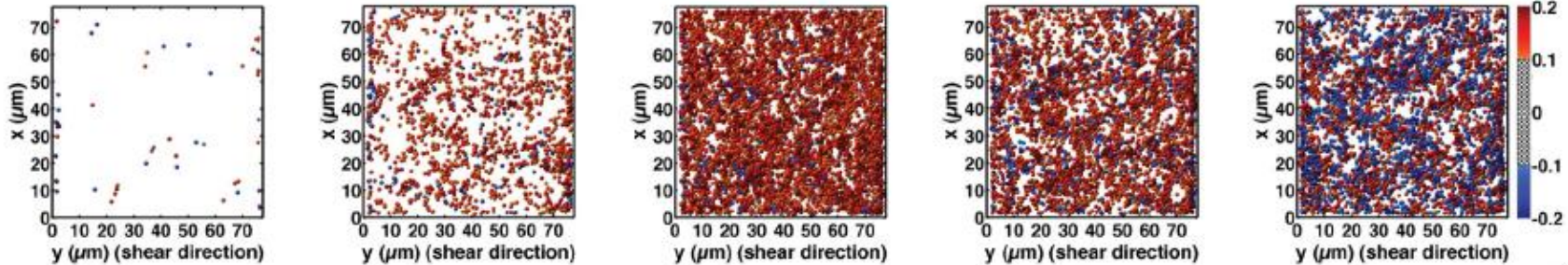
K E Jansen, D A Weitz and F Spaepen

Vol fraction ~62 %
 Constant Strain rate of (1-5) 10^{-5} sec⁻¹.
 Max strain of 10%.

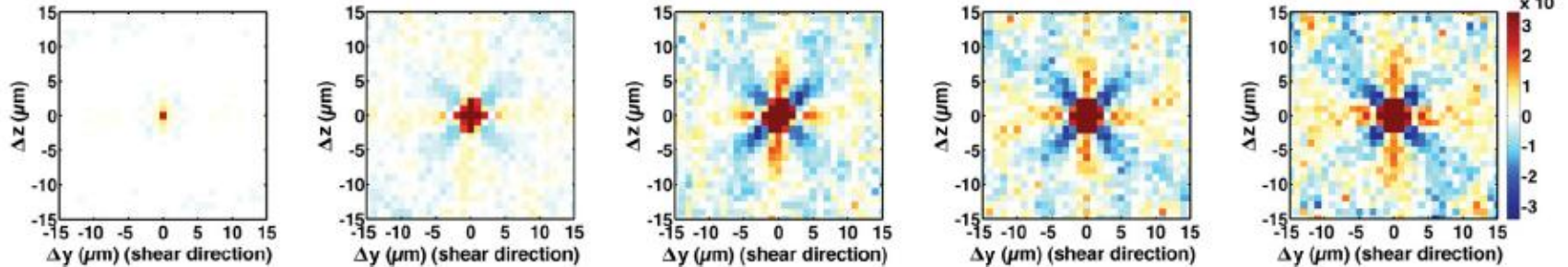
Yield strain ~2 %



(b) Highest Strain 'articles'



(c) Spatial correlation of Local Strain

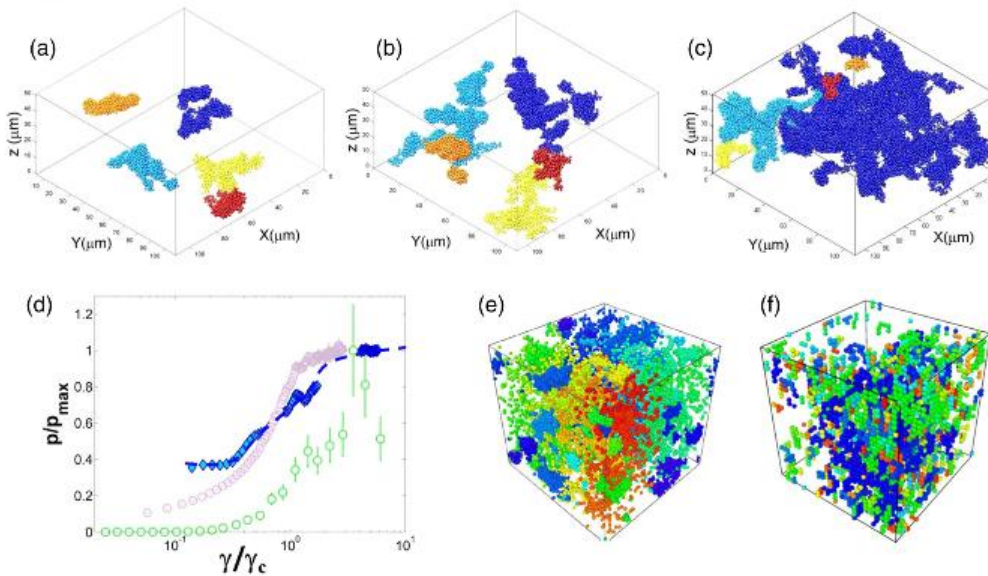


Direct Observation of Percolation in the Yielding Transition of Colloidal Glasses

Antina Ghosh,^{1,2} Zoe Budrikis,³ Vijayakumar Chikkadi,^{1,2} Alessandro L. Sellerio,⁴ Stefano Zapperi,^{4,3,5,6} and Peter Schall¹

PRL 118, 148001 (2017)

PHYSICAL REVIEW LETTERS

week ending
7 APRIL 2017

Hard Sphere Glass
Vol Frac~0.60
Dia=1.3 microns

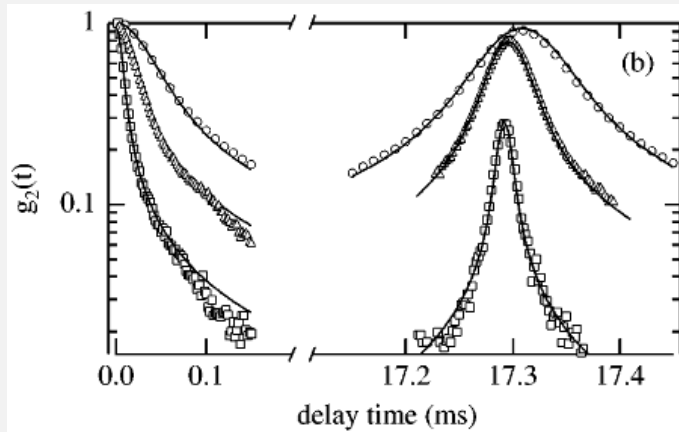
equilibrated state (rejuvenation and subsequent relaxation for three hours) and applied uniform, slow shear at constant rate $\dot{\gamma} = 10^{-4} \text{ s}^{-1}$, of the order of the inverse structural

FIG. 1. Observation of the microscopic yielding transition. (a)–(c) Evolution of highly nonaffine clusters in experiments at strains 2.1, 4.9, and 10.1%. (d) Evolution of fraction p normalized by the maximum fraction p_{\max} of active sites with strain in experiments (diamonds), atomistic simulations (pink circles), and mesoscale simulations (green circles). (e) Clusters of highly nonaffine particles in atomistic simulation at 2% strain. (f) Clusters of active sites in mesoscale simulations at 40% strain.

Growing clusters of nonaffine deformation percolate at Yielding .The spanning Cluster is fractal with $D_f \sim 2$ and the correlation length diverges upon approaching the Critical yield strain.

Microscopic Irreversibility Under Oscillatory Shear

Diffusing Wave Spectroscopy Under Oscillatory Shear DWS Echo Technique



VOLUME 78, NUMBER 24

PHYSICAL REVIEW LETTERS

16 JUNE 1997

Yielding and Rearrangements in Disordered Emulsions

P. Hébraud, F. Lequeux, and J.P. Munch

Laboratoire de Dynamique des Fluides Complexes, 3 rue de l'Université, 67070 Strasbourg, France

D. J. Pine

Departments of Chemical Engineering and Materials, University of California, Santa Barbara, California 93106

(Received 25 March 1997)

Below yield strain (measured separately) several percent of particles were rearranging irreversibly with each cycle.

PHYSICAL REVIEW E 66, 051402 (2002)

Rearrangements in hard-sphere glasses under oscillatory shear strain

G. Petekidis,* A. Moussaïd, and P. N. Pusey

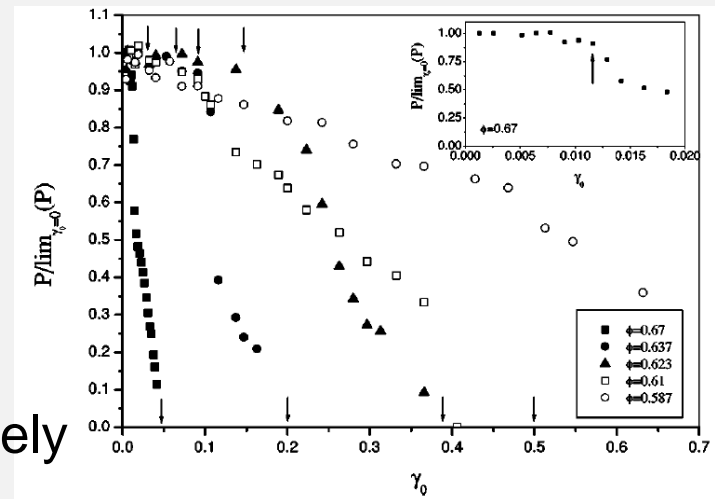
Department of Physics and Astronomy, The University of Edinburgh, Mayfield Road, Edinburgh, EH9 3JZ, United Kingdom

(Received 21 April 2002; published 26 November 2002)

G Petekidis et al PR A 66, 051402(2002)

Yielding marked division between completely Reversible and irreversible behavior.

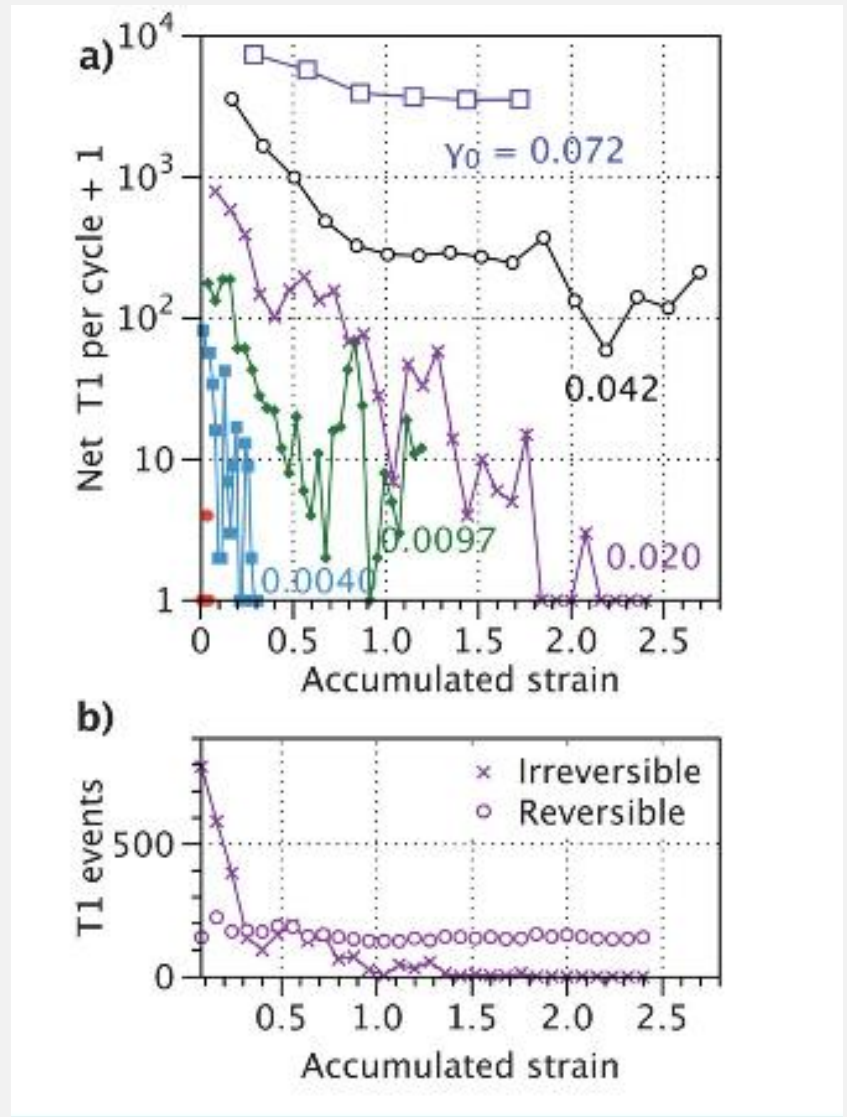
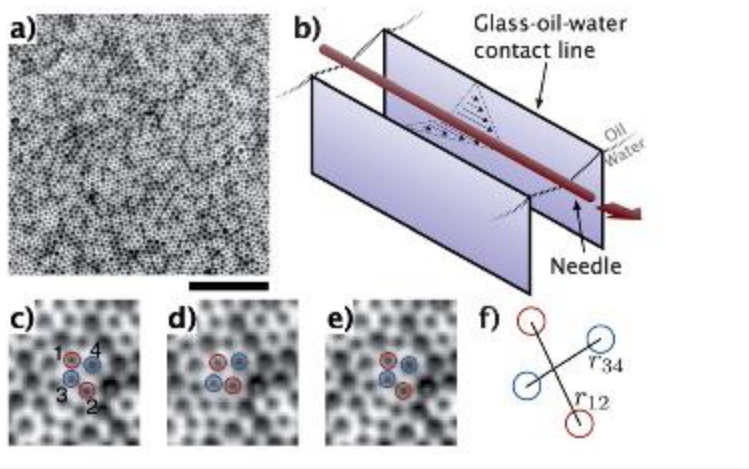
No transient behavior studied.



Yielding and microstructure in a 2D jammed material under shear deformation

†Nathan C. Keim and Paulo E. Arratia, Soft Matter 9, 6222(2013)

*



Local mobility and microstructure in periodically sheared soft particle glasses and their connection to macroscopic rheology

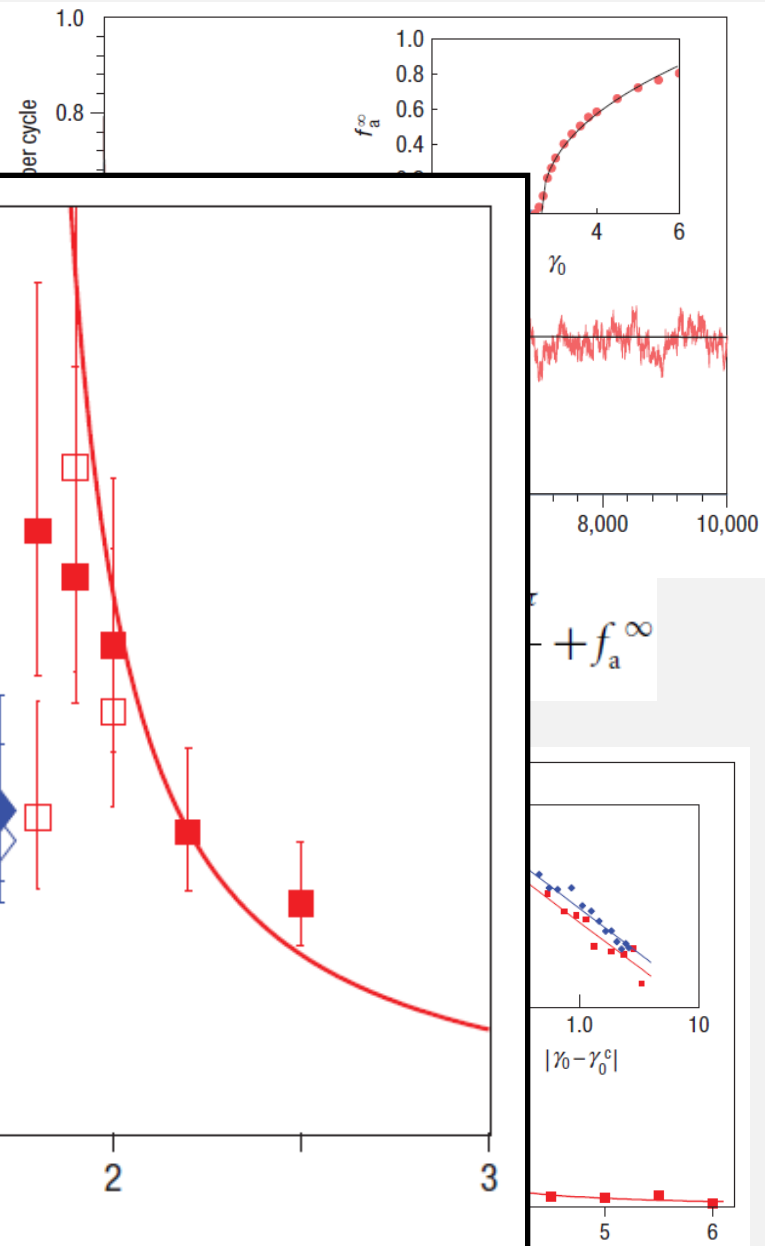
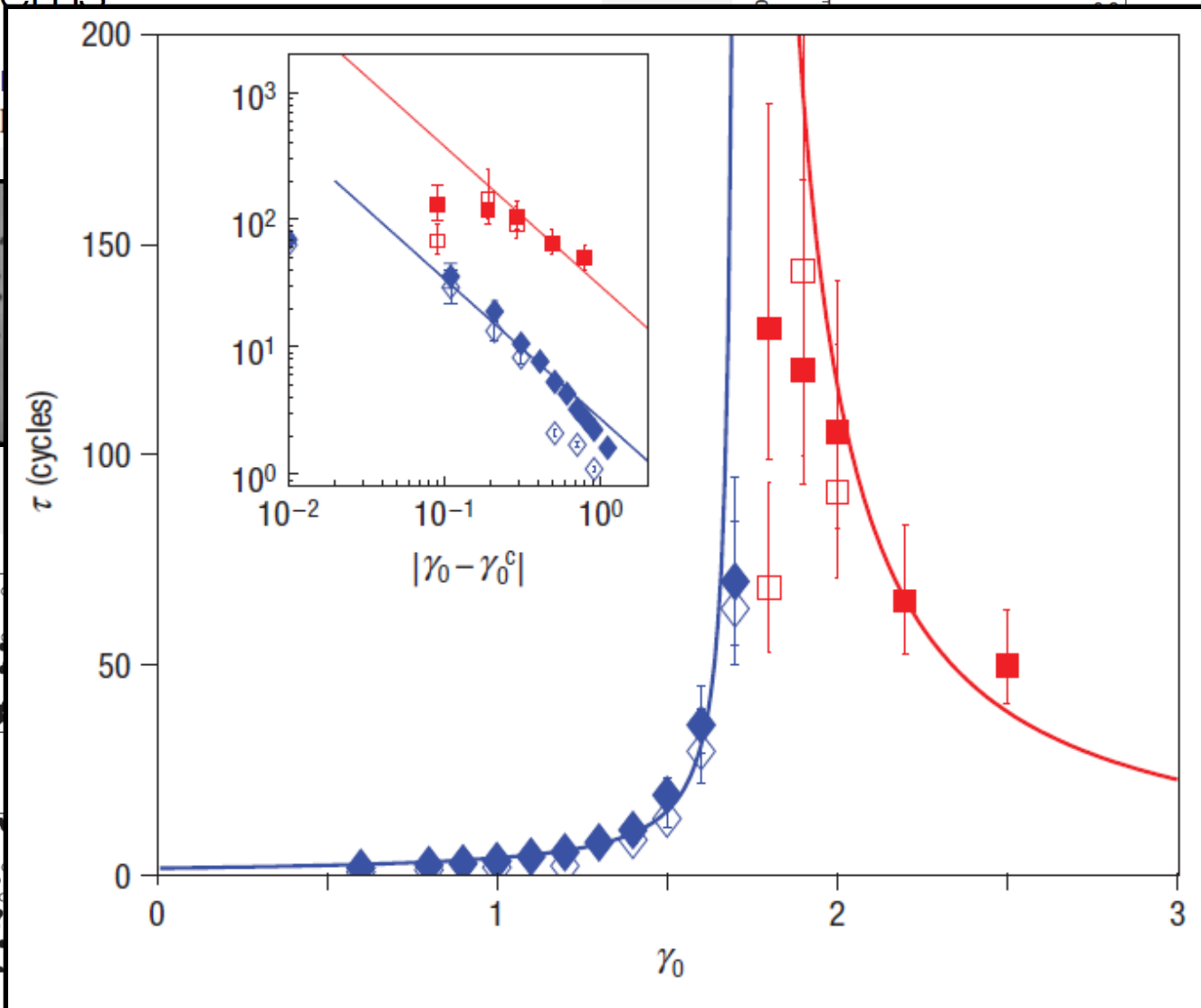
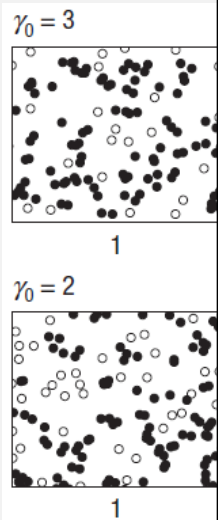
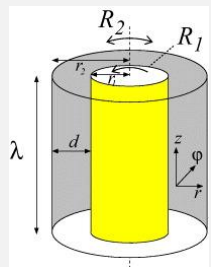
Lavanya Mohan, Charlotte Pellet, Michel Cloitre and Roger Bonnecaze

Journal of Rheology ,57,1023 (2013)

“Our findings concerning the particle scale dynamics also exhibit interesting analogies with the behavior of non-Brownian hard sphere suspensions in a viscous fluid, which are subjected to oscillatory shear motion [Pine et al.(2005)Corte et al.(2008)]. The physical origin of interparticle interactions is a priori significantly different in both systems: In non-Brownian suspensions, particles interact through long-range hydrodynamic interactions; in dense suspensions of soft particles, particles experience short-range EHD forces at particle contacts.”

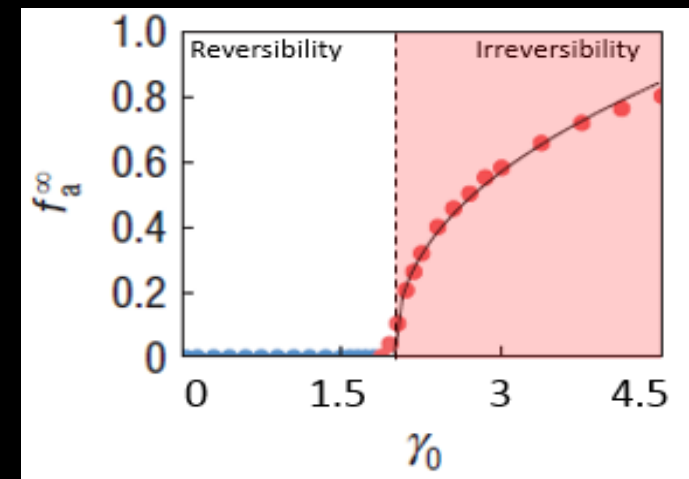
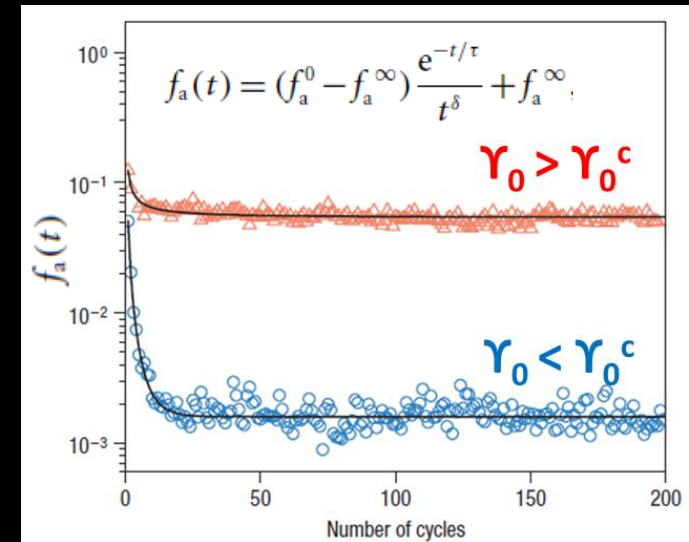
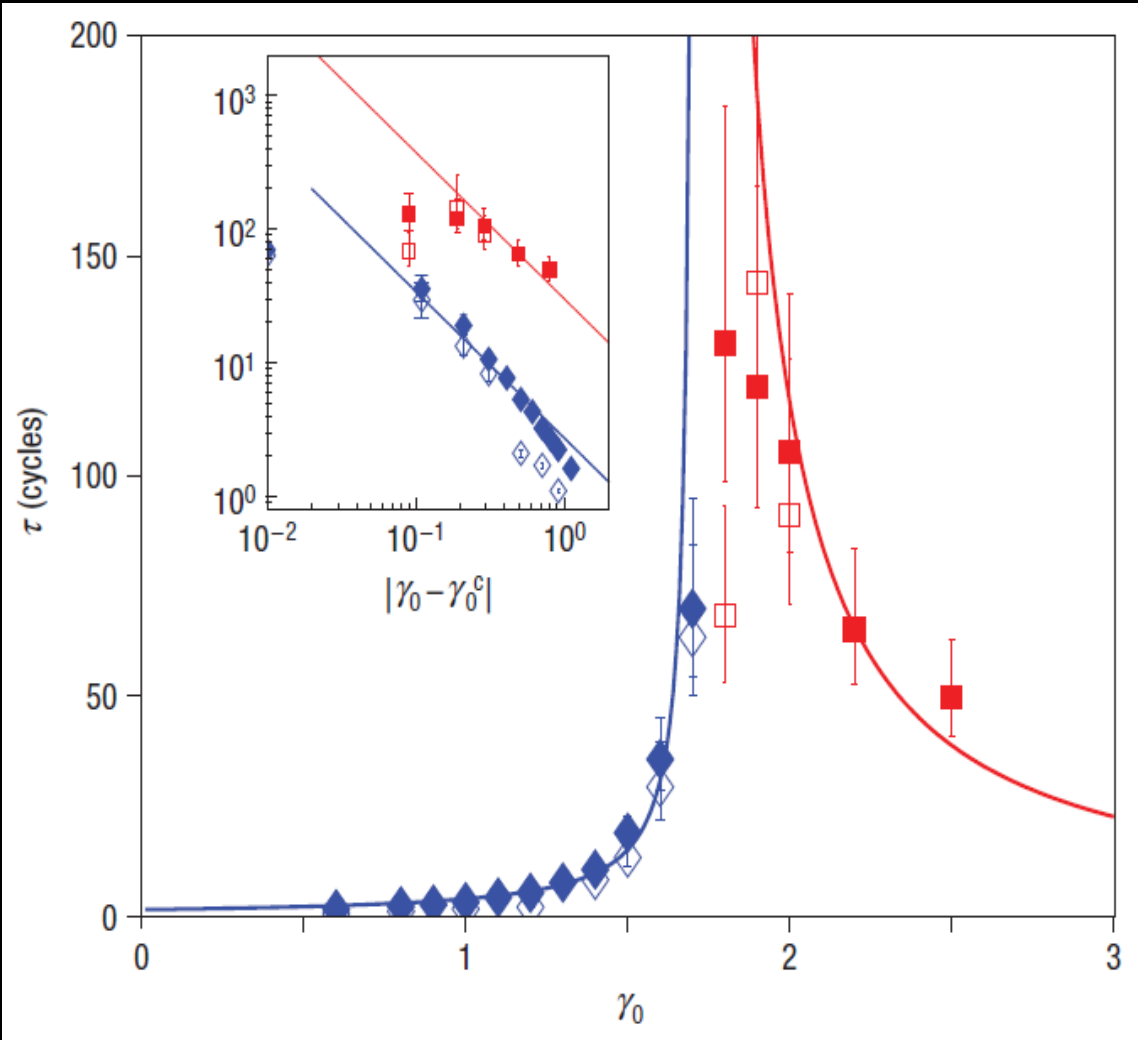
Random organization in periodically driven systems

LAURENT CORTÉ¹, P. M. G.
nature physics | VO



Sharp onset of irreversible dynamics: **Absorbing Phase Transition**

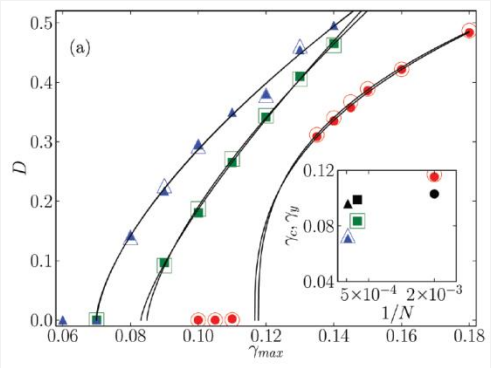
Random Organization & Basis for Mechanical Memory



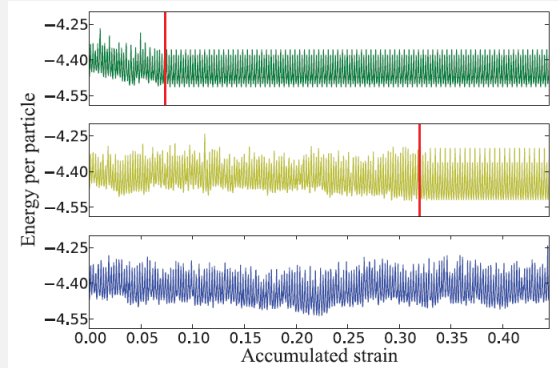
- Irreversibility onset – Non-equilibrium phase transition from an absorbing state to a fluctuating steady state
- System adapts to a particular strain

❖ Can periodically driven amorphous solids show reversible steady states below a threshold stress?

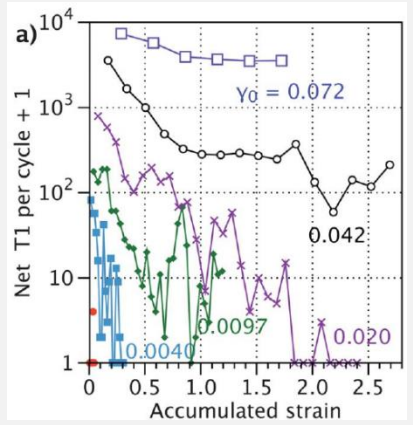
❖ Can ideas from dilute suspensions (Corte' et al.) be extended to denser ones?



Fiocco et al., PRE (R) 88, 020301 (2013)

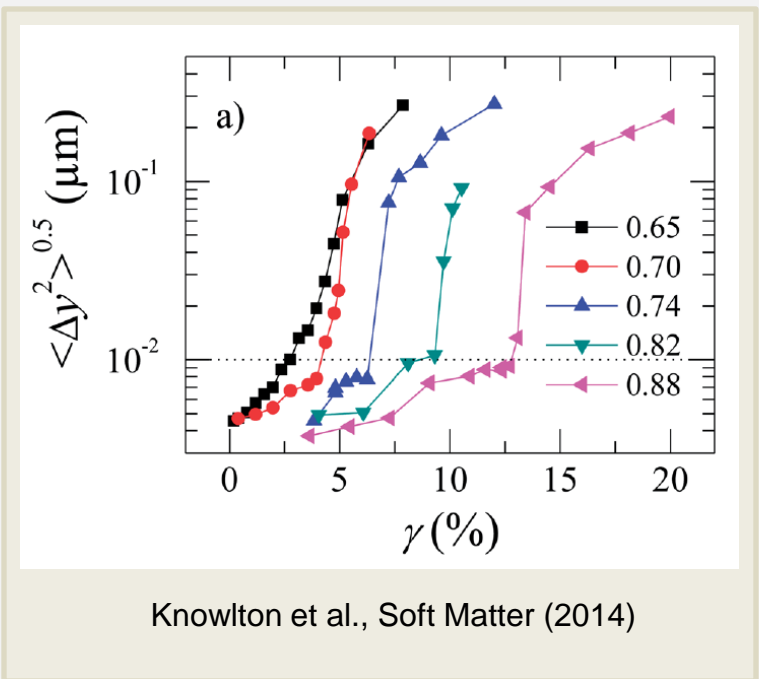


Regev et al., PRE 88 062401 (2013)



Kiem and Arratia
Soft Matter 9, 6222 (2013)

Slotterback et al., PRE 85,
021309 (2012)



Knowlton et al., Soft Matter (2014)

Experimental signatures of a nonequilibrium phase transition governing the yielding of a soft glass

K. Hima Nagamanasa,^{1,*}† Shreyas Gokhale,^{2,*} A. K. Sood,^{2,3} and Rajesh Ganapathy^{3,†}

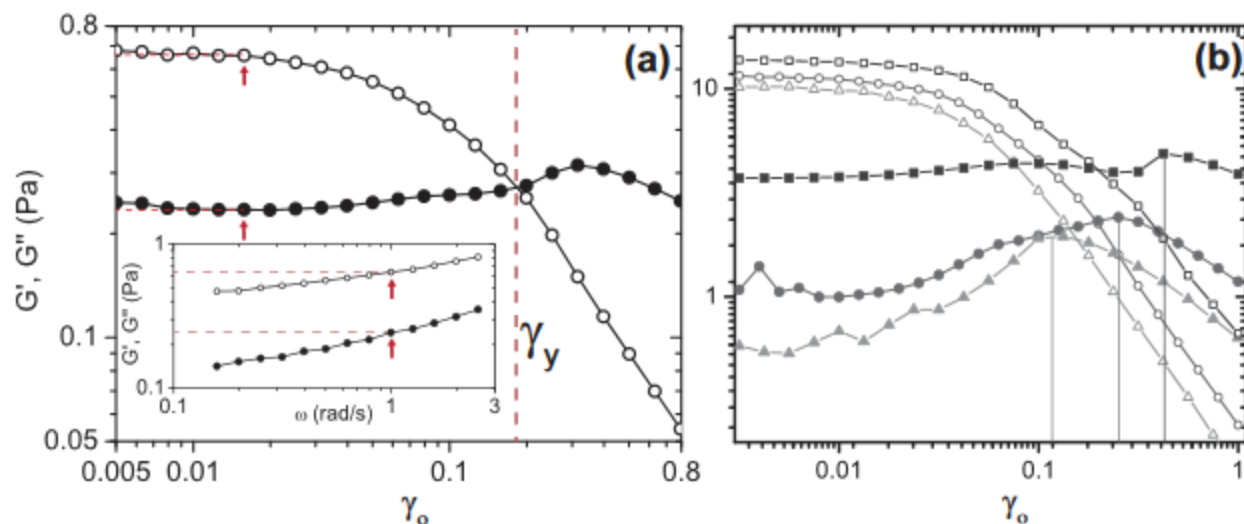
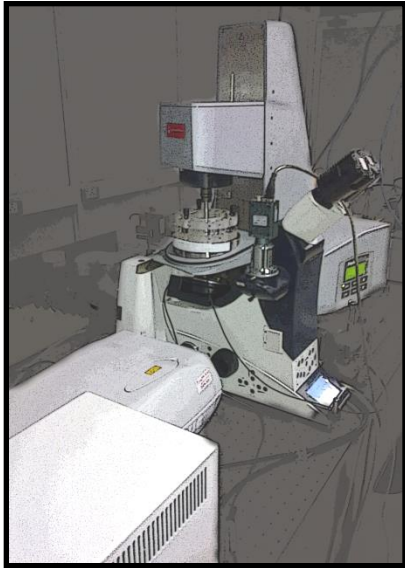


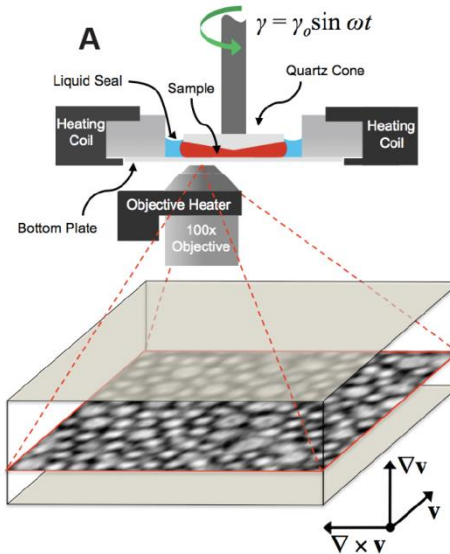
FIG. 2. (Color online) (a) γ_0 -sweep measurements performed at $\omega = 1$ rad/s. Inset shows results from ω -sweep experiments at $\gamma_0 = 0.015$. G' and G'' are denoted by (\circ) and (\bullet) , respectively. The red arrows and dotted lines in the figure and the inset highlight the values of G' and G'' for $\gamma_0 = 0.015$ and $\omega = 1$ rad/s. (b) γ_0 -sweep experiments for $\omega = 0.1$ rad/s (\blacktriangle), $\omega = 1$ rad/s (\bullet) and $\omega = 20$ rad/s (\blacksquare). G' and G'' are denoted by open and filled symbols respectively. The solid lines highlight the γ_0 corresponding to the G'' peak.

Experiments

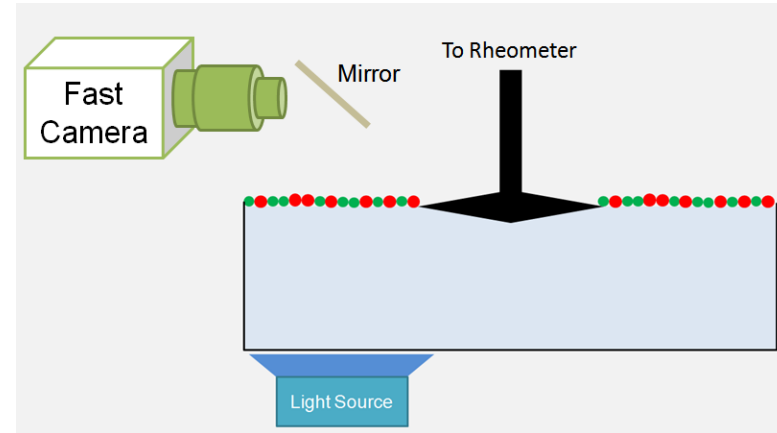
Colloid Experiment



MCR 301 Rheometer mounted atop Visitech Fast Confocal



Bubble Raft Experiment



Bubble size ratio 1:2
Bubble solution + Glycerol + Na Stearate

Measurements

- Apply pre-shear before every run
- Determine yield strain (γ_y) from standard linear rheology
- Subsequently, at fixed ω and for γ_0 across γ_y , explore temporal evolution of viscoelastic moduli and glass microstructure

System

Binary Colloidal Glass made of size tunable Poly N-isopropylacrylamide (PNIPAM) particles



T = 38°C
Size ~ 600 nm



T = 27°C
Size ~ 1 μm

$$n_{\text{small}} : n_{\text{big}} = 3:1,$$

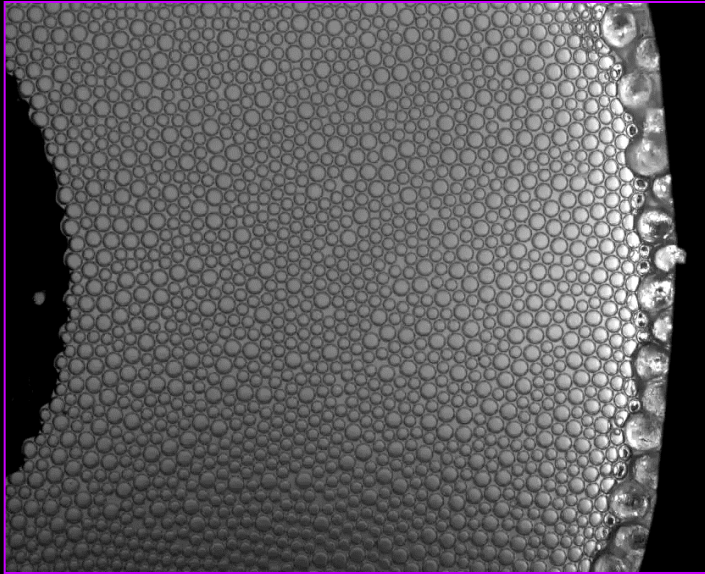
$$R_{\text{small}} : R_{\text{big}} = 1:2$$

$$\phi_{\text{Particle}} > \phi_{\text{glass}} \quad 58\% \text{ at } 27^\circ\text{C}$$

Particles labeled with Rhodamine 6G for confocal imaging, velocity-vorticity plane 7 microns away from bottom plate

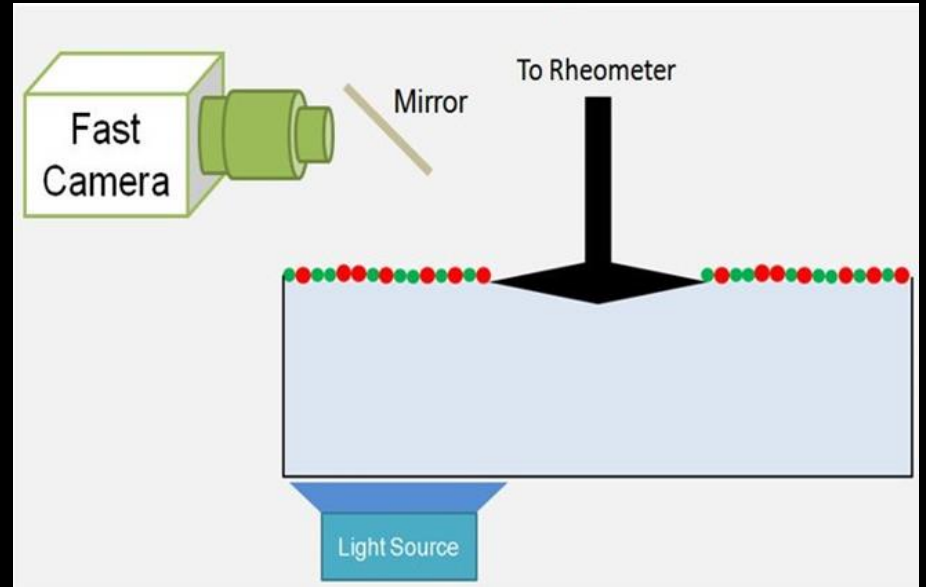
Immobilized layer of particles on top and bottom plates to prevent wall slip

Experimental System – Bubble Rafts



Amorphous Solid: Bi-disperse Bubble Raft

Soft - Frictionless - Athermal Particles

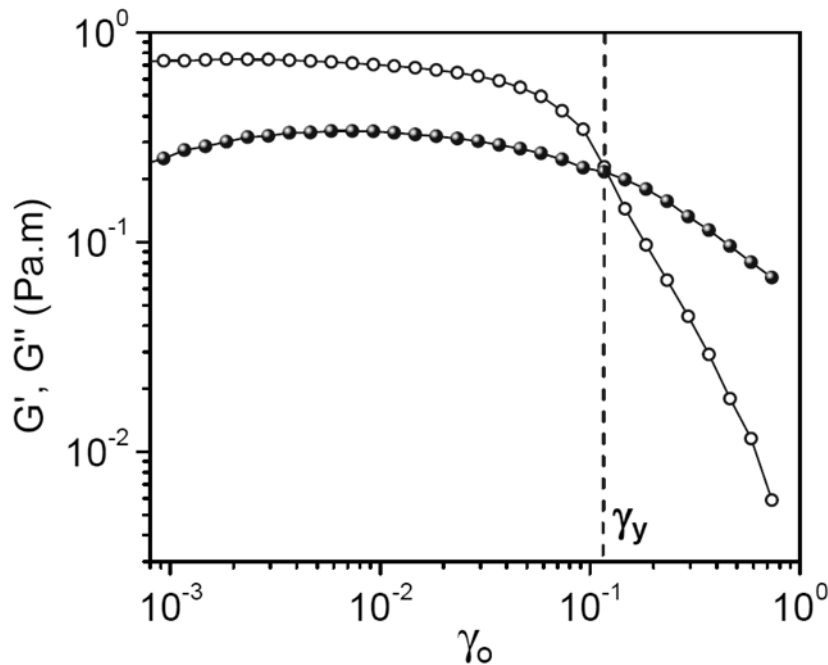


2D Couette Geometry

Area fraction – $\phi > 0.8$

Experimental Details – Bubble Rafts

$\omega = 0.5 \text{ rad/s}$



Basic characterization:

Couette geometry

Area fraction: $\phi > 0.8$

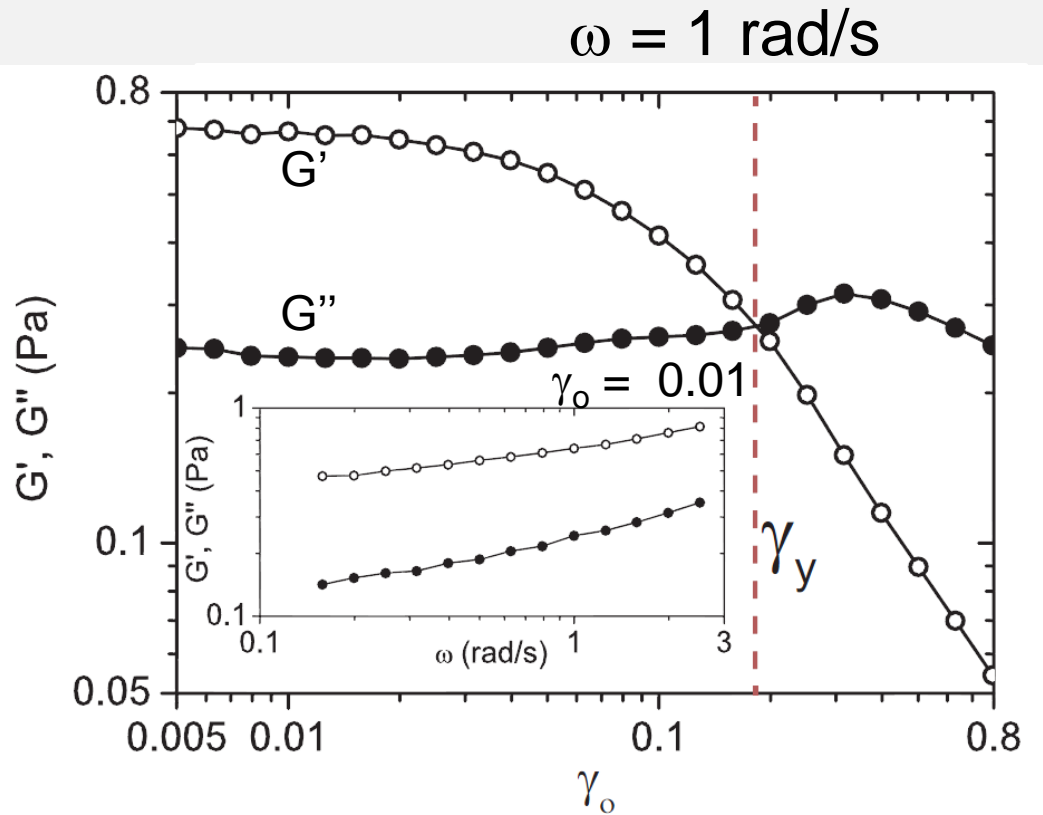
Type of raft: Bidisperse amorphous with some polydispersity

Number ratio of bubbles used: On an average for every 8 big bubbles there are 11 small bubbles in this raft.

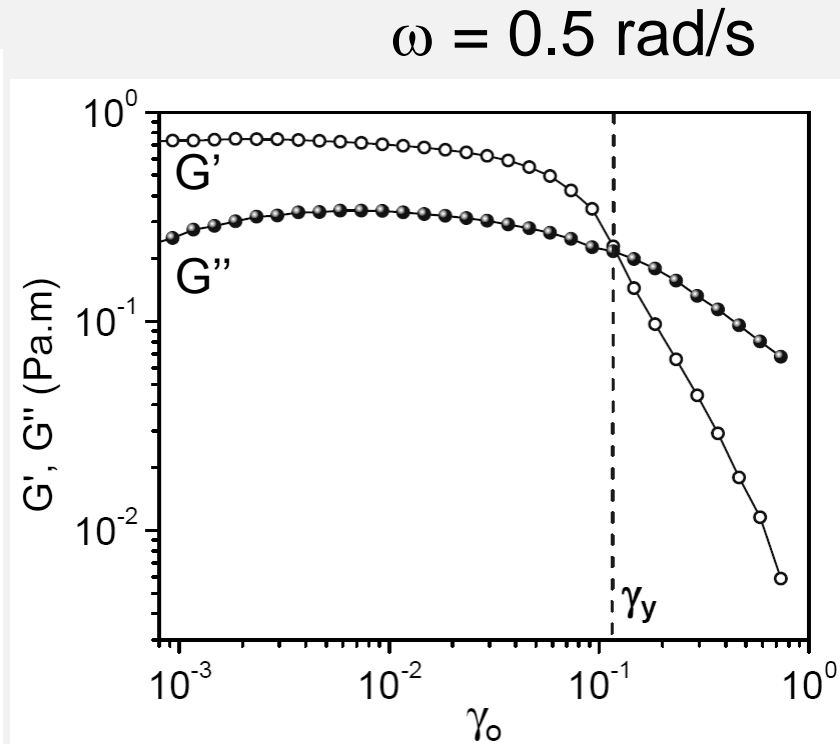
Size of bubbles: Between 1mm and 2.5mm in diameter.

Number of bubbles in the field of view: ~ 1300 .

Determining the Yield Strain



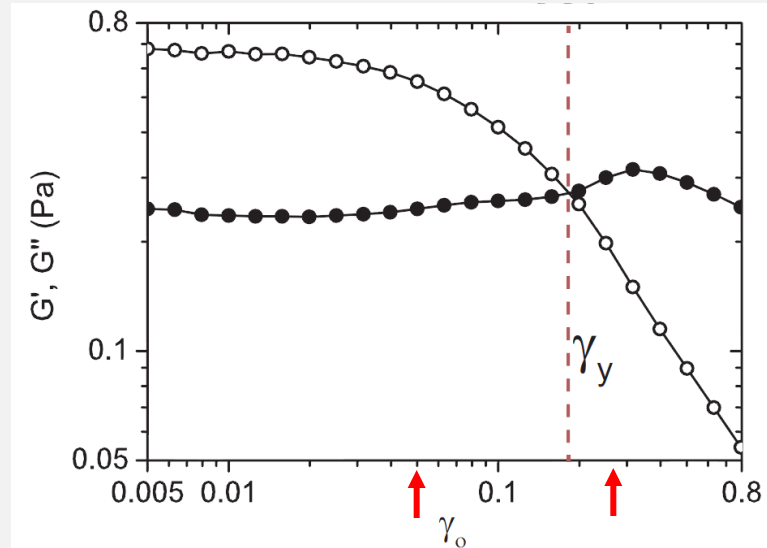
Binary Colloidal Glass



2D Amorphous Bubble raft

Koumakis, Brady and Petekidis, PRL 110, 178301 (2013): Yielding peak in G'' due to two mechanisms: Low Freq – Brownian motion assisted cage escape and Escape through shear induced collisions at High Freqs.

Colloids - Single-Particle Dynamics



Stroboscopic Imaging of the Binary Colloidal Glass Under Oscillatory Shear

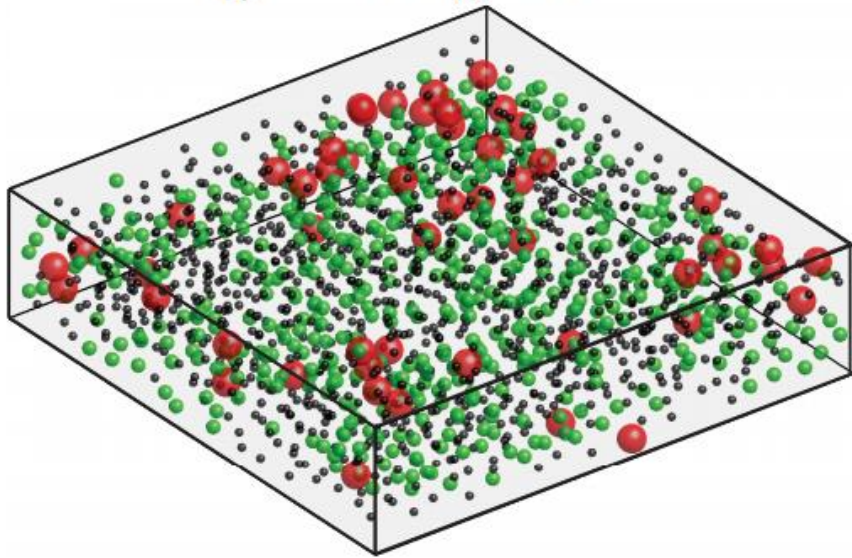
Below γ_y

Above γ_y

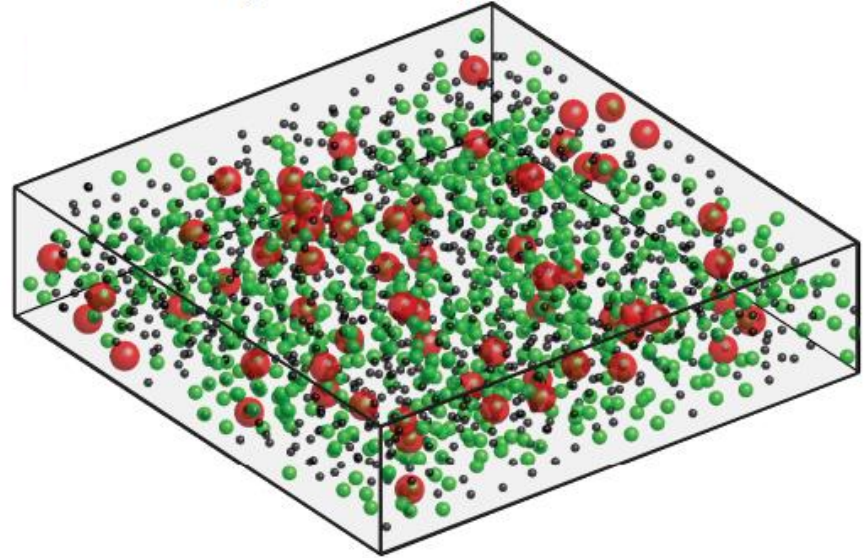
Quantifying Irreversible Yield Events (3D)

Fraction of particles (f_{IR}) that do not return to their positions after one complete oscillation cycle

$$\gamma_o = 0.12, f_{IR} = 0.37$$

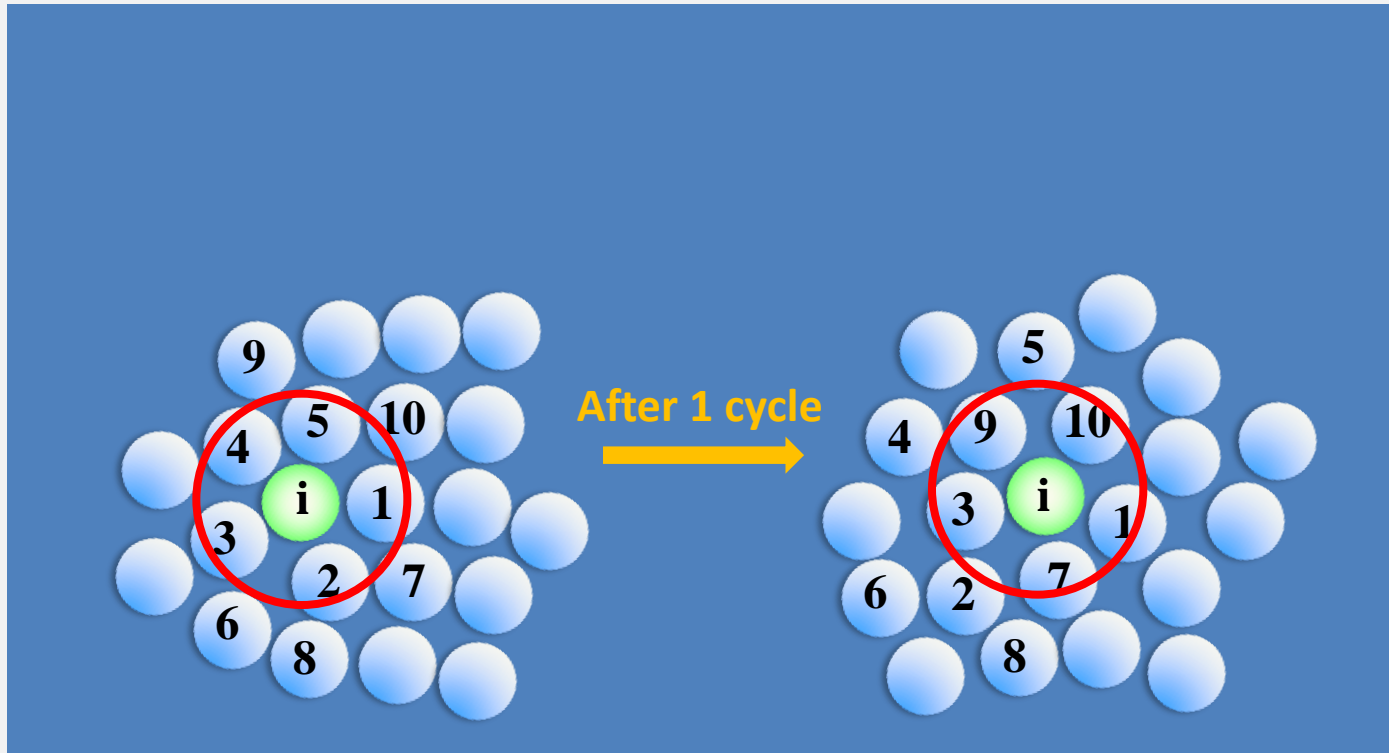


$$\gamma_o = 0.25, f_{IR} = 0.45$$



Red: Big Irreversible Particles
Green: Small Irreversible Particles
Black: Reversible Particles

Quantifying Irreversible Yield Events



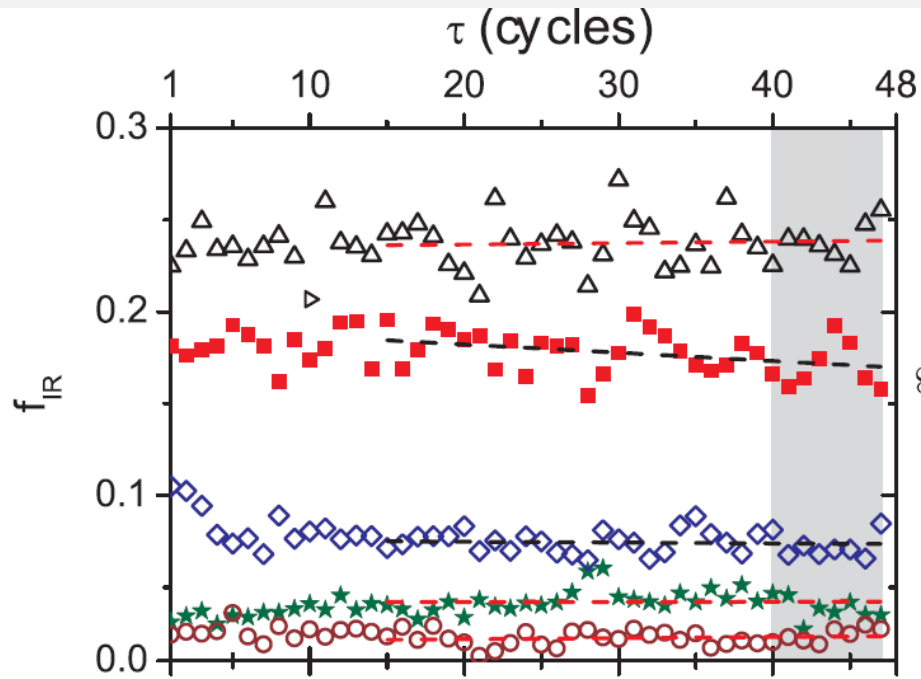
No. of nearest neighbors of i initially, N_{ini}

No of common neighbors of i , after one cycle, N_{fin}

The particle i is labeled irreversible if $N_{fin} / N_{ini} < 0.66$

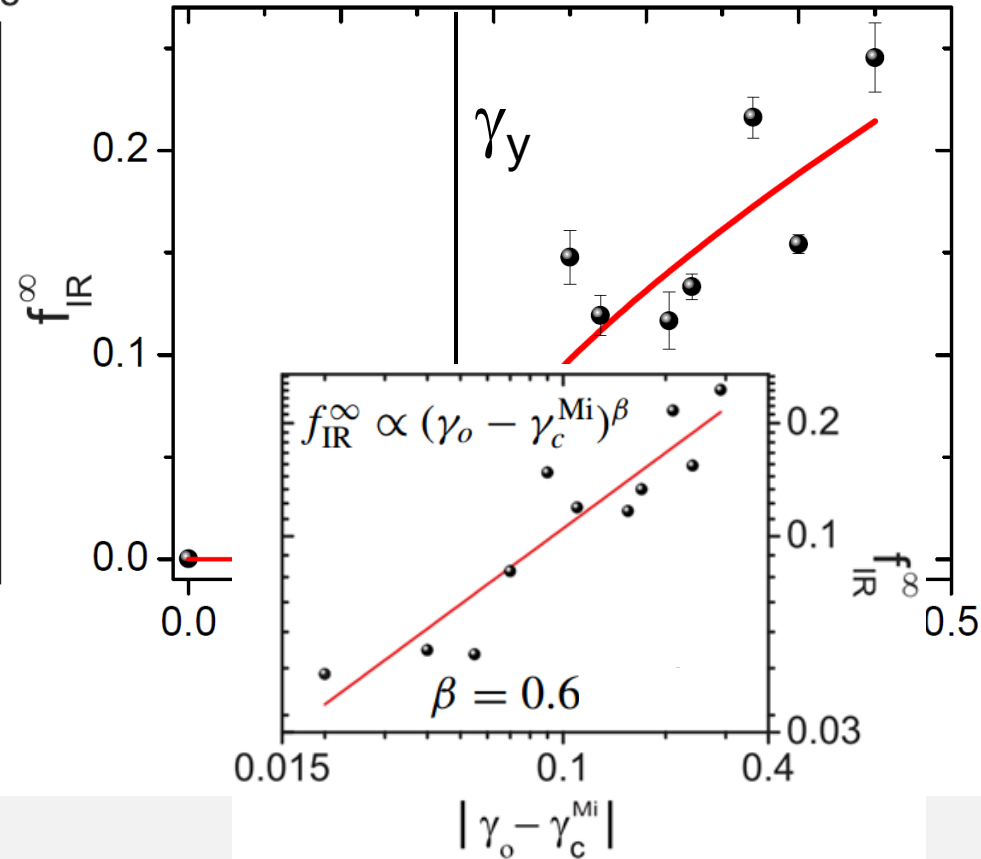
Colloidal Glass - Time Evolution of Irreversibility

Irreversible fraction corrected for thermal contribution



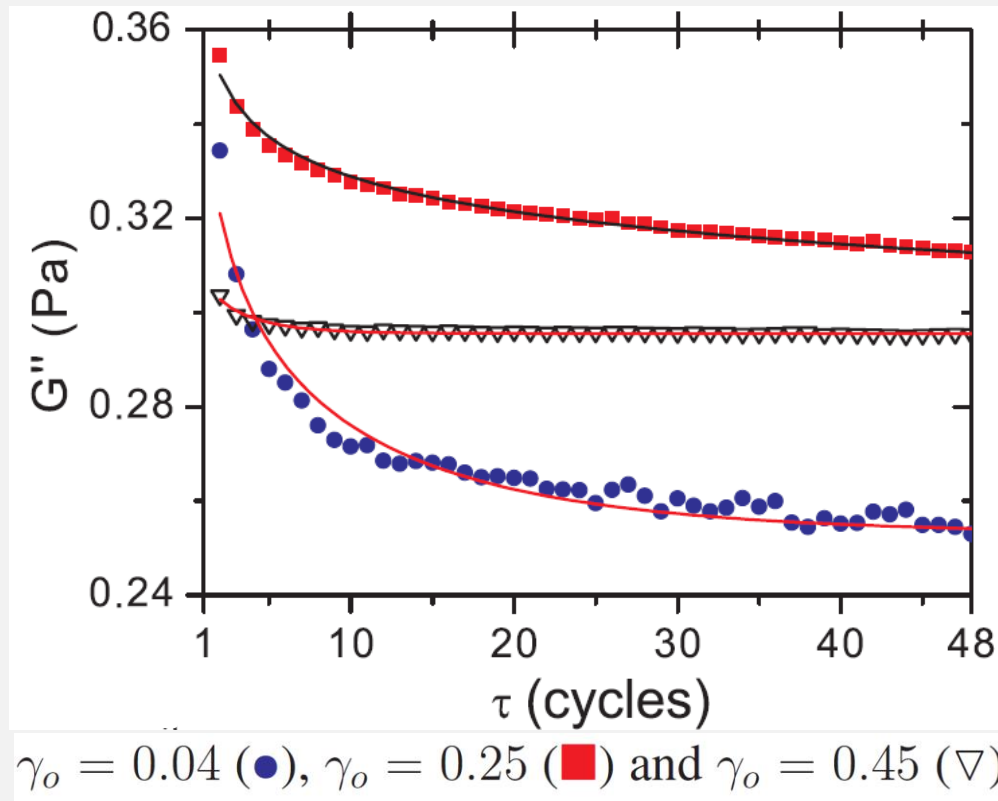
$\gamma_o = 0.05$ (\circ), $\gamma_o = 0.12$ (\star), $\gamma_o = 0.215$ (\diamond),
 $\gamma_o = 0.25$ (\blacksquare) and $\gamma_o = 0.37$ (\triangle)

f_{IR}^∞ – behaves like an order-parameter



Colloidal Glass - Time Evolution of Irreversibility

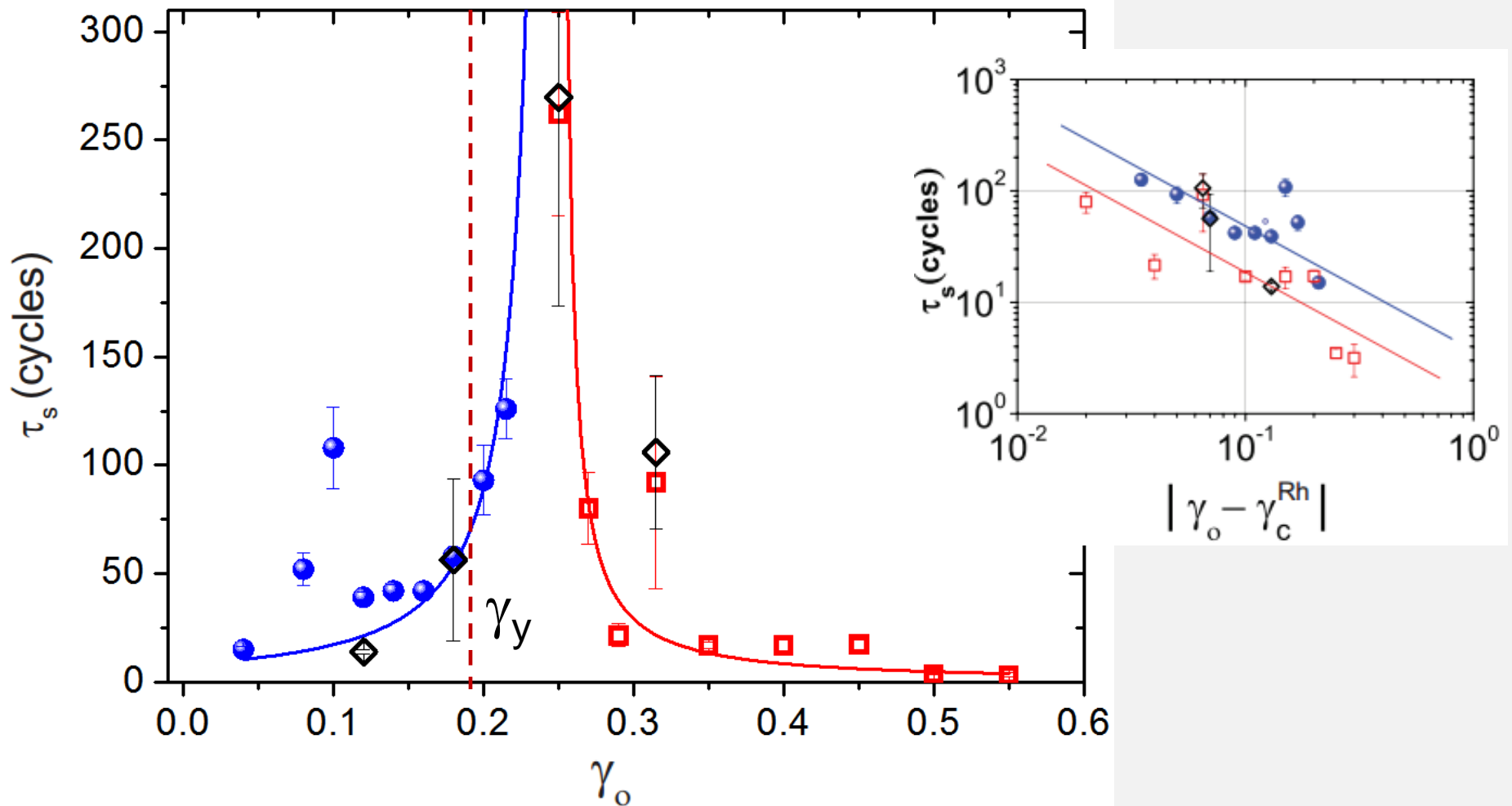
Irreversible events lead to dissipation of energy and should also reflect in the loss modulus G''



$$G''(\tau) = (G''_o - G''_\infty) \frac{e^{-\tau/\tau_s}}{\tau^\delta} + G''_\infty$$

Signatures of Slowing Down Near Yield

Manasa et al., PRE 89, 062308 (2014)



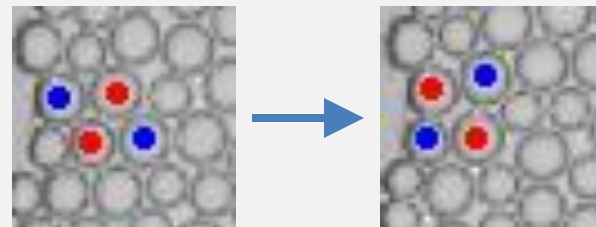
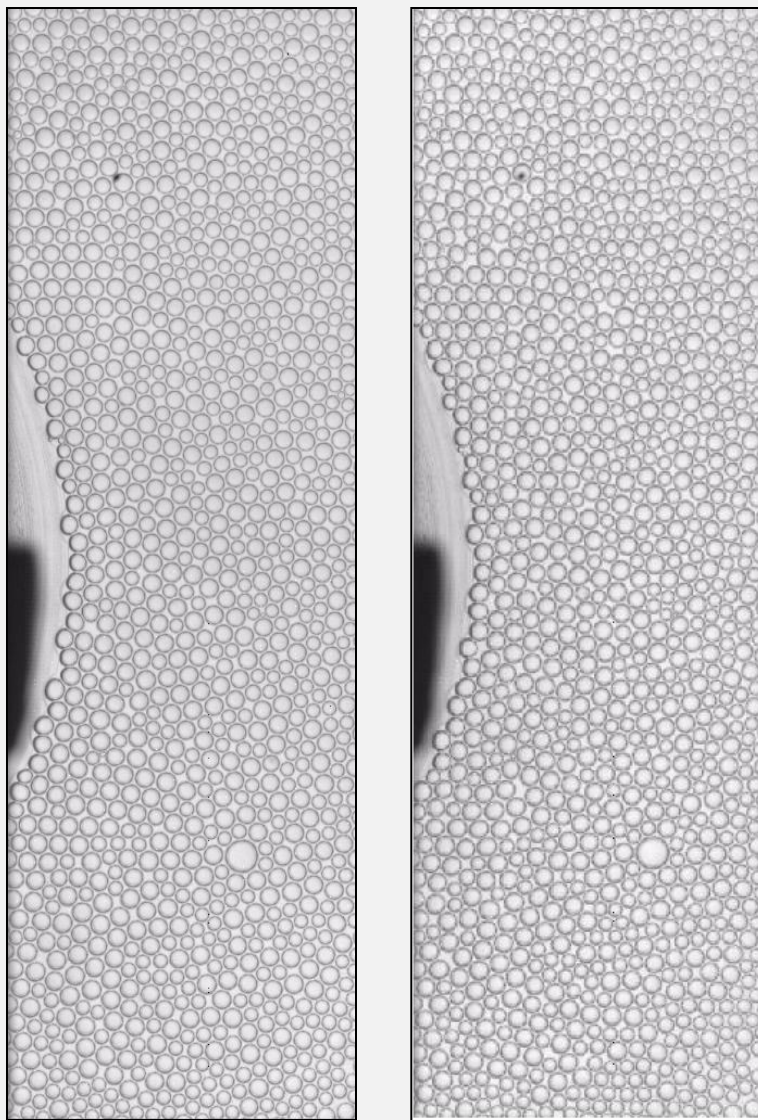
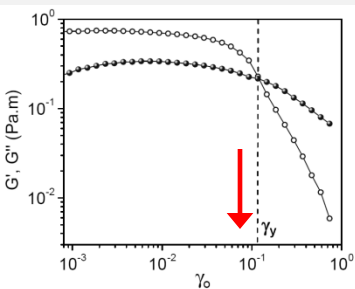
$$\tau_s \propto |\gamma_o - \gamma_c|^{-\alpha}, \text{ with } \alpha = 1.1$$

“Absorbing Phase Transition”, CDP Universality Class

Menon and Ramaswamy PRE 79, 061108 (2009)

Removing "T" from the Picture

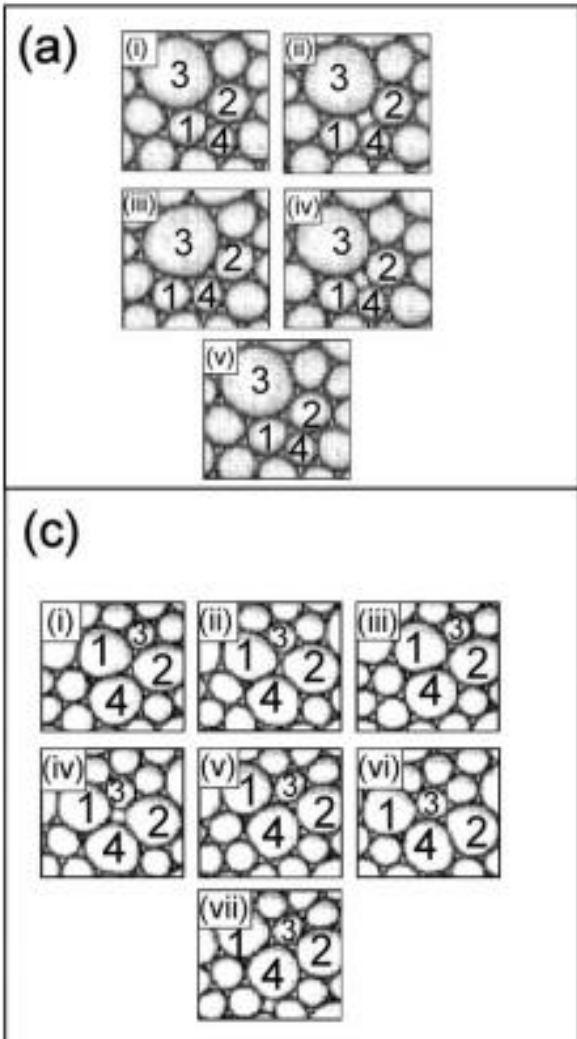
Irreversible T1 Event



A True "Absorbing/Reversible State" below Yield

Reversible plastic events in amorphous materials

Micah Lundberg,¹ Kapilanjnan Krishan,¹ Ning Xu,^{2,3} Corey S. O'Hern,^{4,5} and Michael Dennin¹



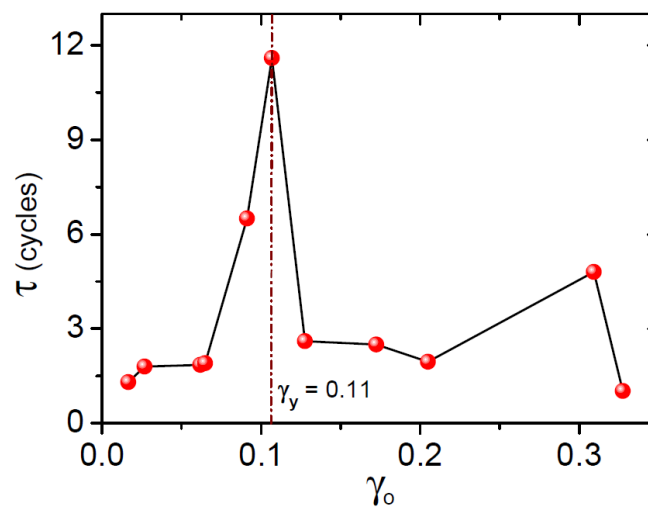
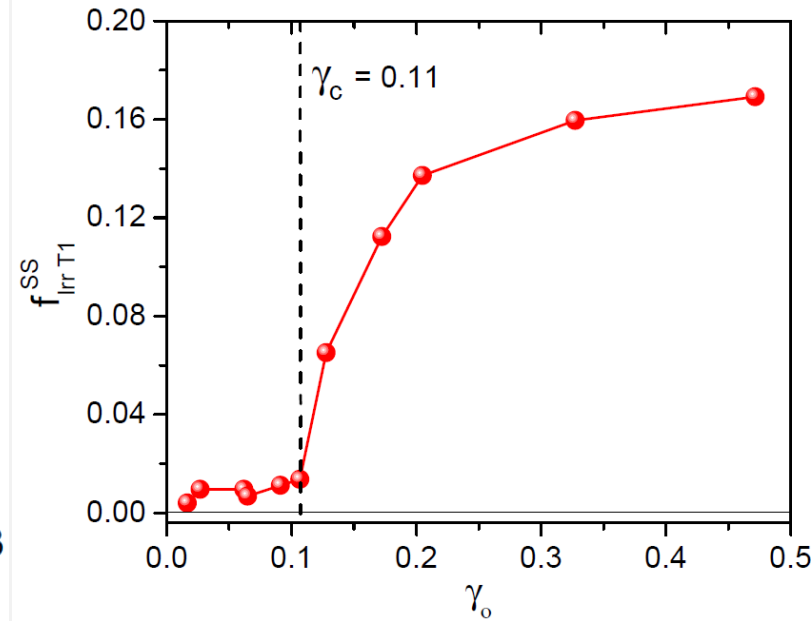
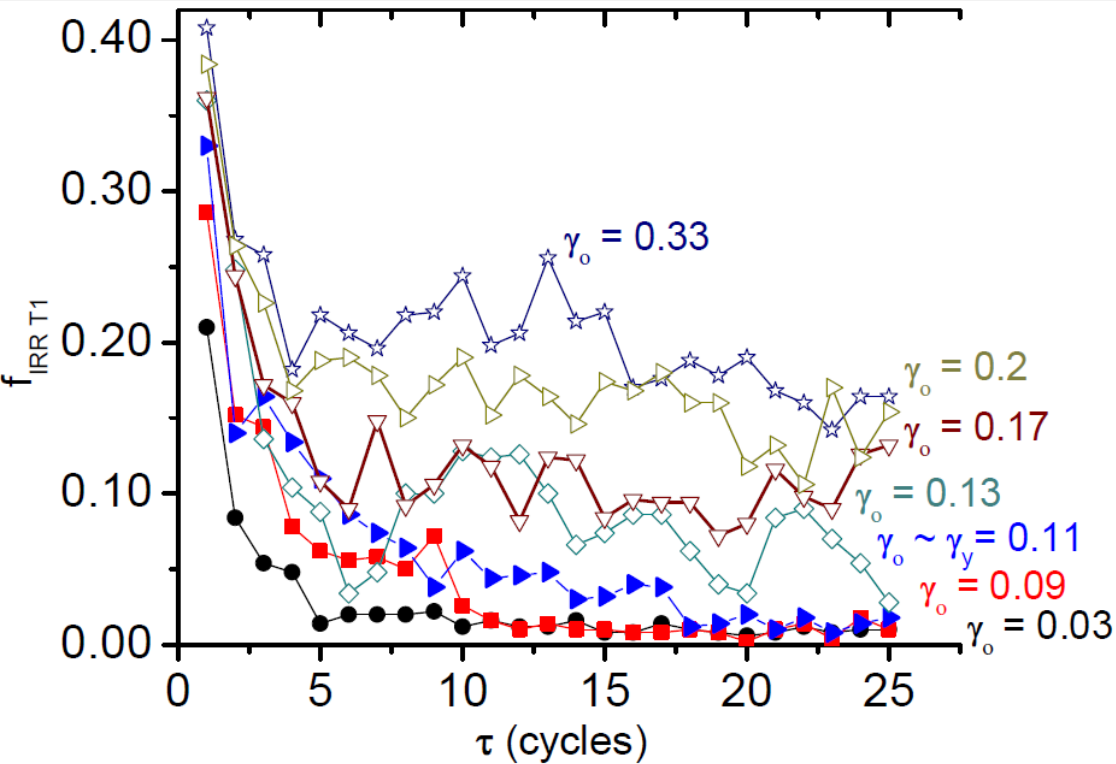
T1 events correspond to a neighbor switching event in which two neighboring bubbles lose contact, and two nextnearestneighbors become neighbors

Initial state i, middle of the T1 event ii, second state iii, middle of the T1 event under reversal of shear iv, and the return to the initial state v

Irreversible T1 event: initial state i-iii, middle of the T1 event iv, and the second state v-vii.

“Transition from an elastic deformation with mostly reversible T1 events to macroscopic plastic flow with mainly irreversible T1 events. This is a direct experimental confirmation of reversible microscopic plastic zones or shear transformation zones STZs. The concept of a STZ as a reversible, two-state transition within a material that can be created or destroyed was first proposed by Falk and Langer.”

Bubble Rafts – Time Evolution of Irreversibility



Is there Spatial Correlations between local irreversible rearrangements near Yielding?

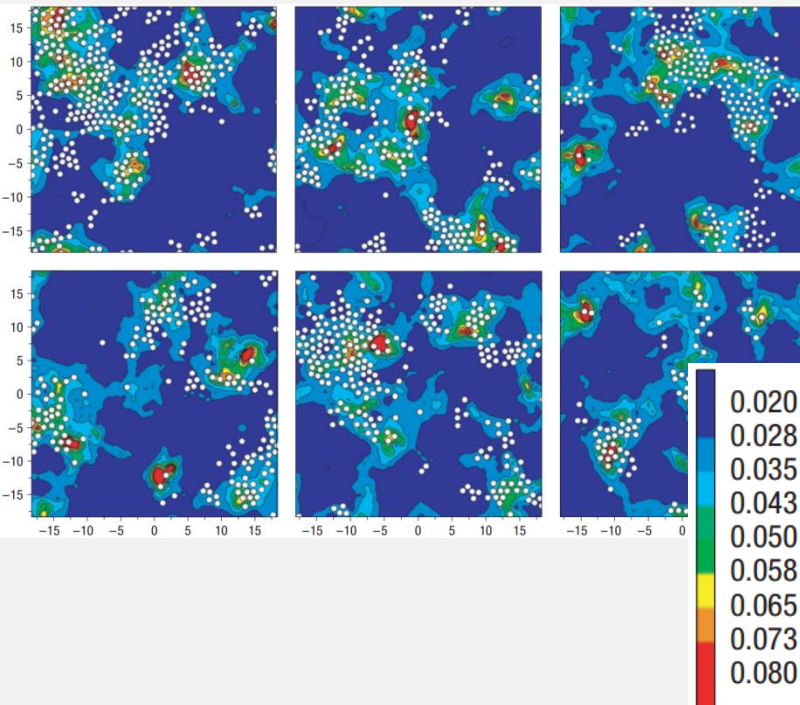
Nature Physics **4**, 711 - 715 (2008)

Published online: 20 July 2008 | doi:10.1038/nphys1025

Irreversible reorganization in a supercooled liquid originates from localized soft modes

Asaph Widmer-Cooper¹, Heidi Perry², Peter Harrowell¹ & David R. Reichman²

Local plastic events in glasses originate from spatially Localized low freq Vib modes.



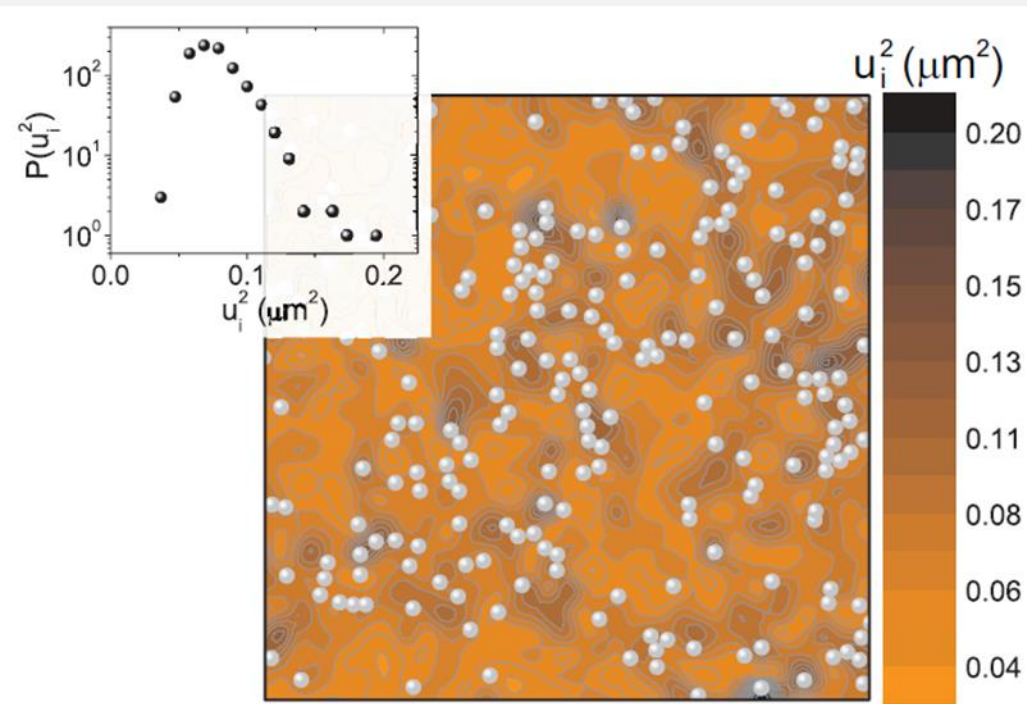
We identify them by local measure of elasticity : DW

Debye-Waller factor: mean squared deviation of a particle from its mean position

$$DW_i = \left\langle \left(\langle r_i \rangle - r_i(t) \right)^2 \right\rangle_{\text{cycle}}$$

$r_i(t)$ instantaneous displacement

$\langle r_i \rangle$ average position of the particle



Averaged over suitable time window.
For each strain, for different time Windows Δt (less than the cage Breaking time), we clustered top 10 % of the high u_i particles based on nn distances.

Average cluster size as a fn of time Exhibits a max at t_{max} which is chosen as time interval to compute Dw_i .

U_i averaged over an oscillation cycle
For $\gamma_0=0.25$ and $\tau=2$.

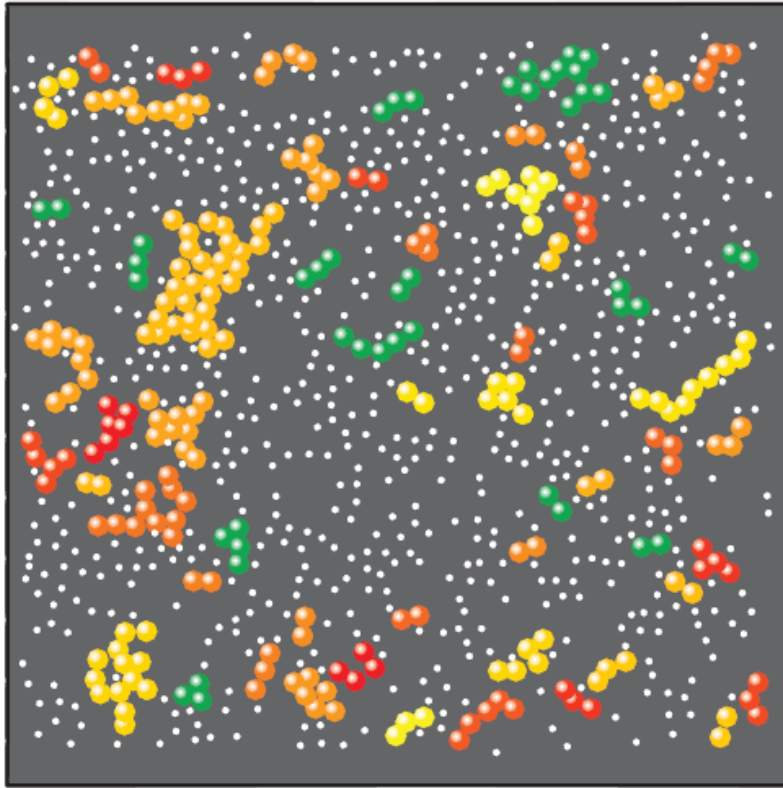
Gray circles: irreversible arrangements

High Debye-Waller (DW) Particles are Spatially Clustered

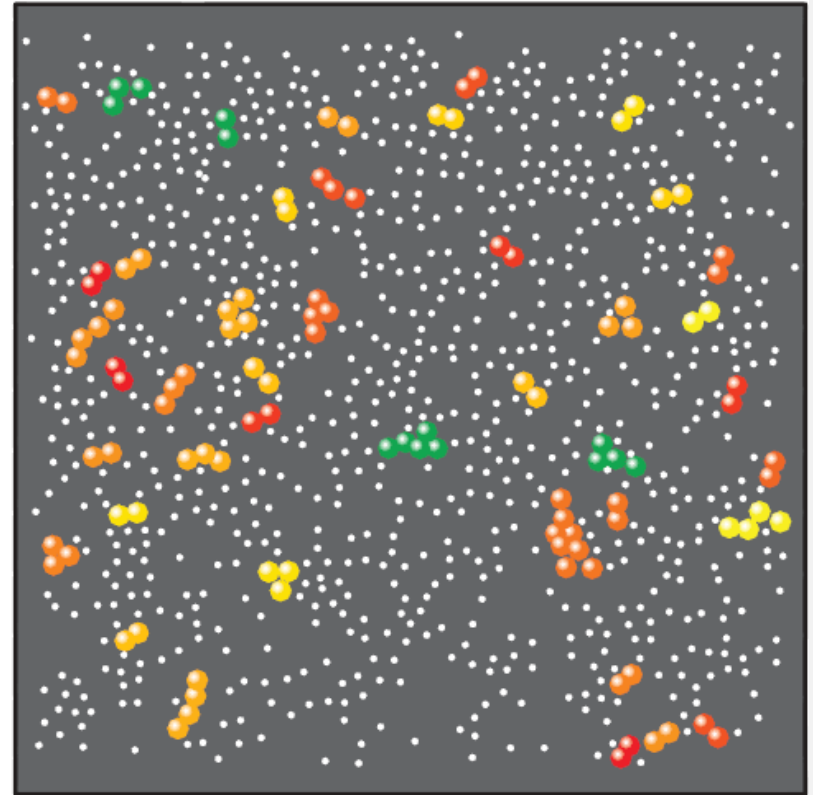
$$\gamma_0 = 0.27$$

Fewer larger clusters

$\tau = 3$ (cycle)

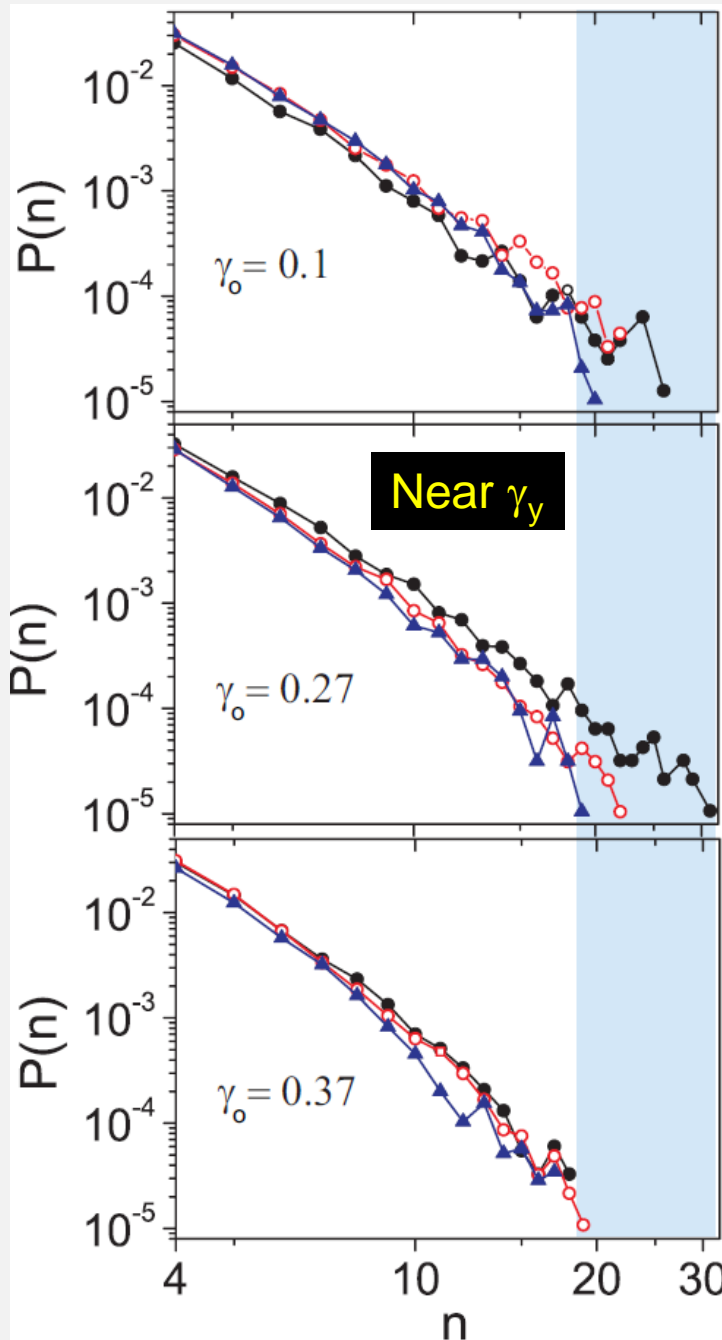
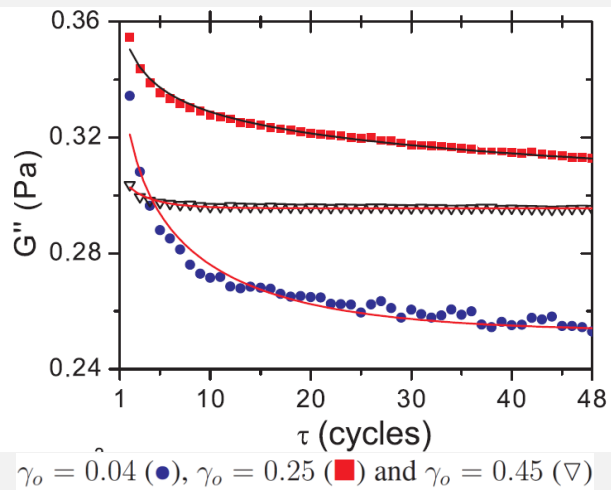


$\tau = 47$ (cycle)



Top 10 % of the high U_i particles are shown as big solid spheres and remaining as small circles. Colors are a visual aid to demarcate clusters.

Time Evolution of DW Particle Clusters



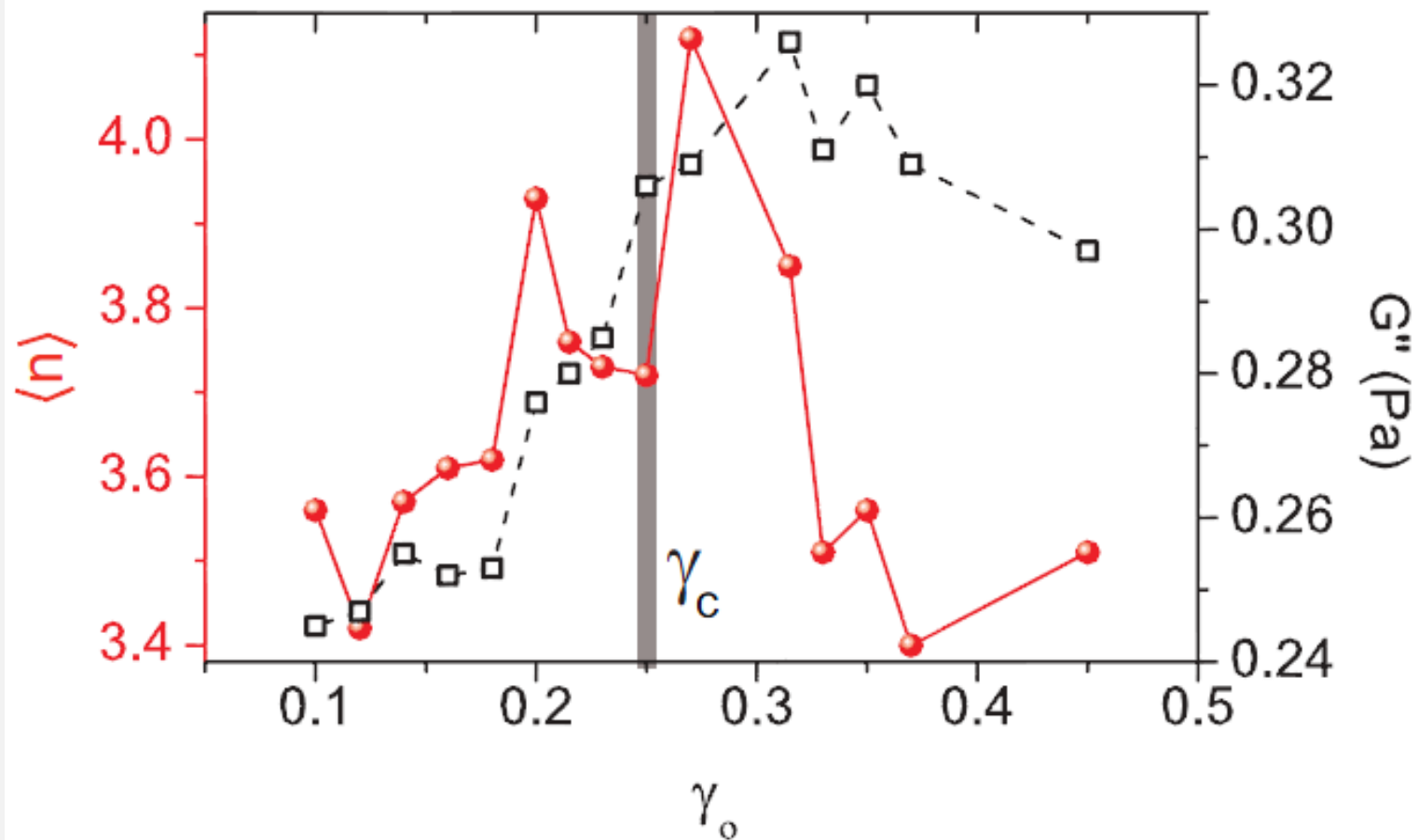
$\tau = 2$ to 10 (●)
 $\tau = 20$ to 30 (○)
 $\tau = 37$ to 47 (▲)

Clusters are larger here .

Here $P(n)$ is stationary

A Growing Length Scale

Cooperative dynamics near yielding



$$\text{average cluster size } \langle n \rangle = \frac{\sum n^2 P(n)}{\sum n P(n)}$$

Note we are imagining in velocity-vorticity plane and NOT in velocity-Gradient plane.

Close to critical strain, correlations between local yield events trigger a cascade of Irreversible rearrangements, manifested as growing Corr length and hence slowing down

Conclusions so far.....

- ❖ Consistent with some recent simulations and experiment – existence of a reversible/absorbing state for strains lower than threshold (γ_y).
- ❖ Slowing Down Accompanied by a Growing Length Scale .
 - Signatures of a Non-Equilibrium Phase Transition Near Yielding ?

Variable-amplitude oscillatory shear response of amorphous materials

Nathan Perchikov and Eran Bouchbinder

1. Two weakly coupled subsystems: slow conf deg of freedom (inherent structures) :
Charaterized by T_{eff}

Fast kinetics-vib deg of freedom

2. Plastic deformation mediated by STZs .
STZ can make transitions between its internal states.

3. Yielding transition is associated with exchange of stability (bifurcation) in the dynamical eq for order parameter m (coarse grained internal variable).

Slowing down mechanism is associated with the exchange of stability in dynamics of m . STZ-STZ interaction is not included.

Above Yield

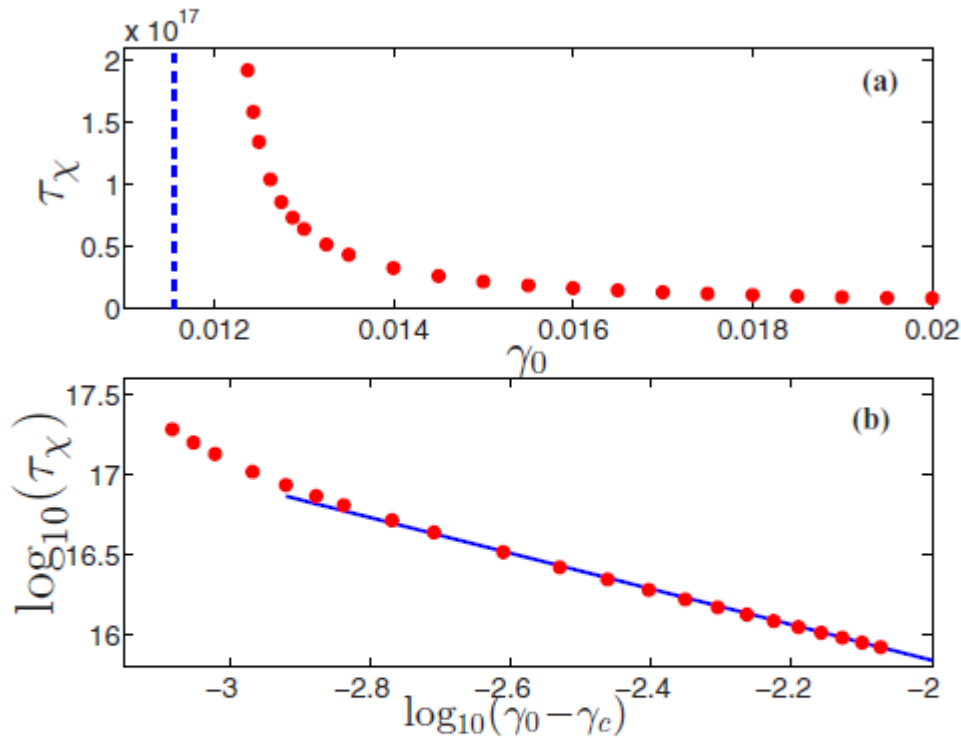


FIG. 4. (Color online) (a) The relaxation time to steady state τ_χ as a function of γ_0 . The dashed vertical line corresponds to $\gamma_0 = \gamma_c$. (b) The same data as in (a), but on a log-log scale. The solid line has a slope of -1.1 , which is consistent with the theoretical prediction in Eq. (19), $\tau_\chi \sim (\gamma_0 - \gamma_c)^{-1}$.

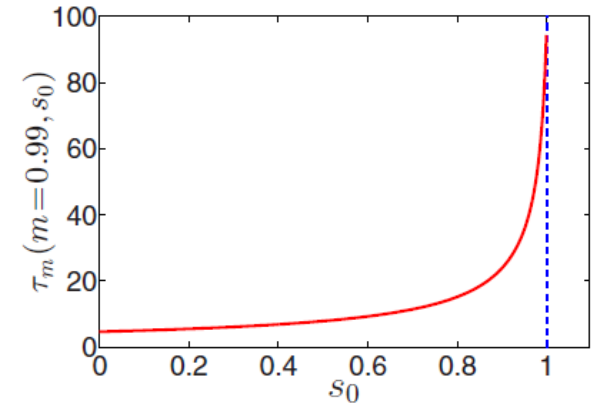


FIG. 5. (Color online) The normalized time $\tau_m(m = 0.99, s_0)$ [where the thermal activation rate factor is suppressed, cf. Eq. (18)] it takes an initially isotropic system, $m(t = 0)$, to reach $m = 0.99$ as a function of s_0 , the magnitude of an applied *unidirectional* (i.e., *not oscillatory*) shear stress in the subyield regime, $s_0 < 1$.

evidence for a similar slowing-down behavior, dominated by a $(\gamma_c - \gamma_0)^{-1}$ variation of the relaxation time, also in the oscillatory case [though it was difficult to detect it numerically]

“ Finally, we note that in [13] it was suggested that the growing relaxation time is associated with a growing correlation length scale due to interaction between different rearrangements, a feature that is absent from the mean-field STZ model.

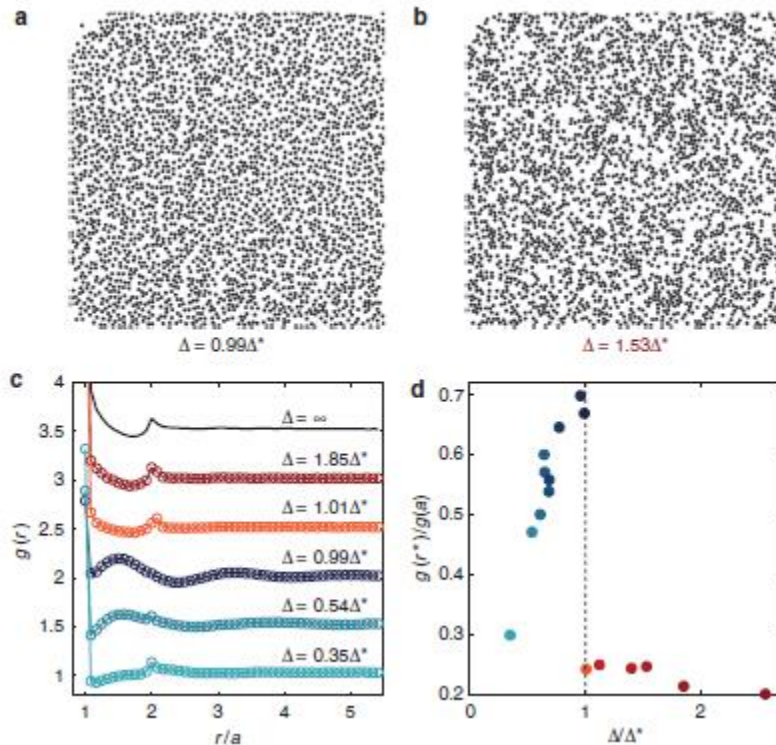
It is also worth noting that while the STZ slowing-down mechanism is related to the dynamics of the orientational order parameter m , measurements of a growing relaxation time scale were based either on the evolution of the energy [9, 10] or on the fraction of particles that do not return to their initial positions at the end of a strain cycle [13]. We have not discussed possible relations between them ”

Order of the Transition??

First order or Second order?

Geometrically protected reversibility in hydrodynamic Loschmidt-echo experiments

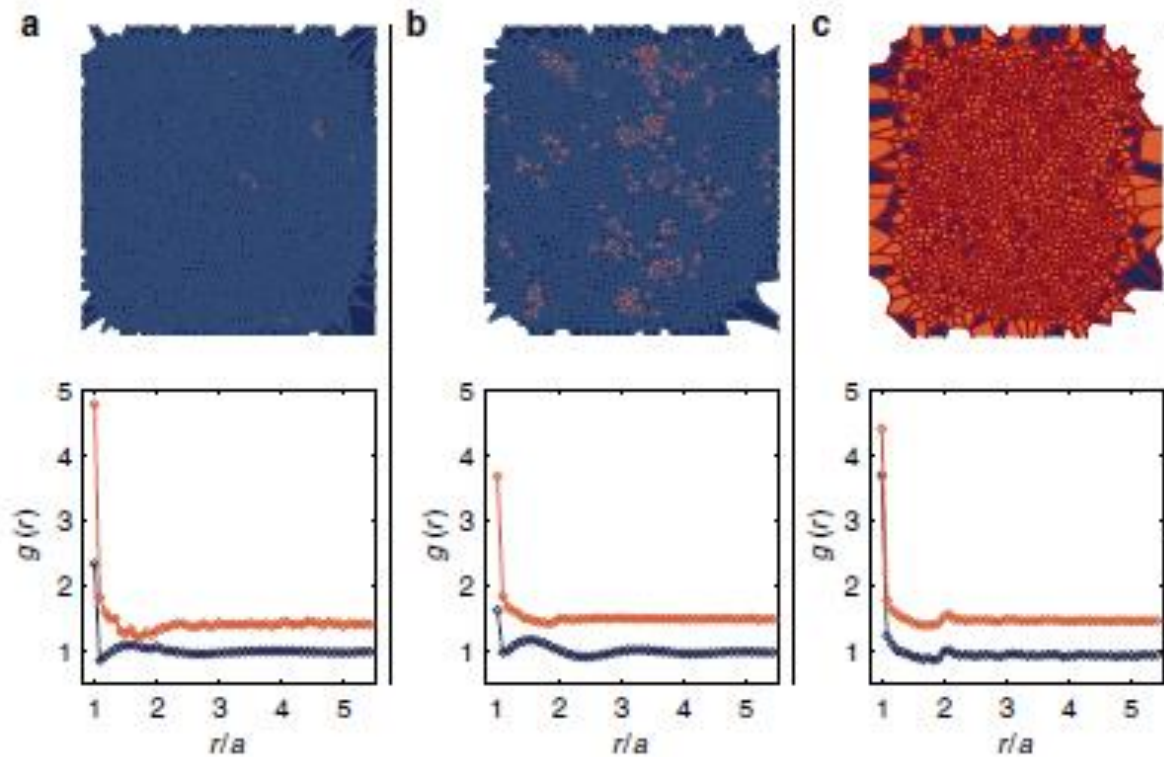
Raphaël Jeanneret¹ & Denis Bartolo^{1,2}



Microfluidic emulsions periodically driven
By a syringe pump.

Oil droplets in aqueous medium,
Dia 25.5 microns,
Area fraction=0.36

Above a maximal shaking amplitude,
structural order and reversibility are lost
through a FIRST ORDER NE Phase
Transition.



0.54

0.99

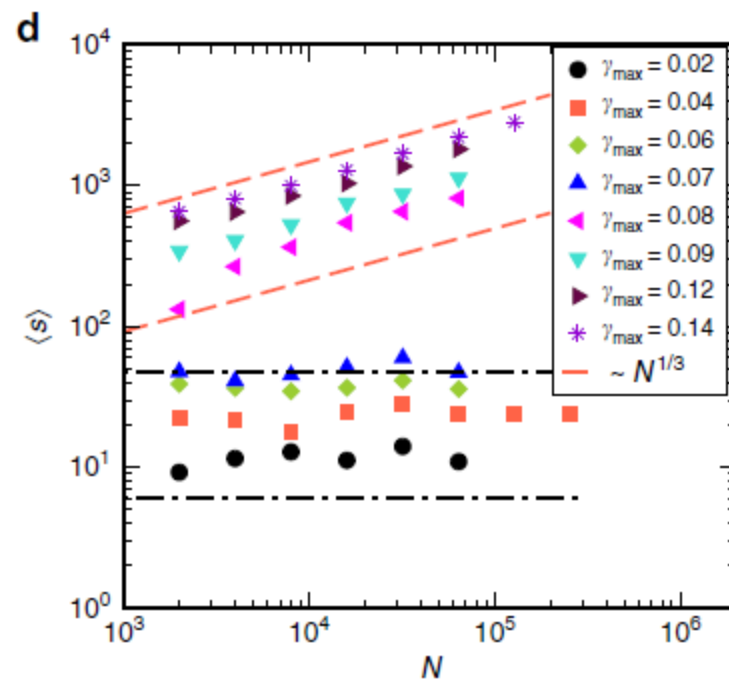
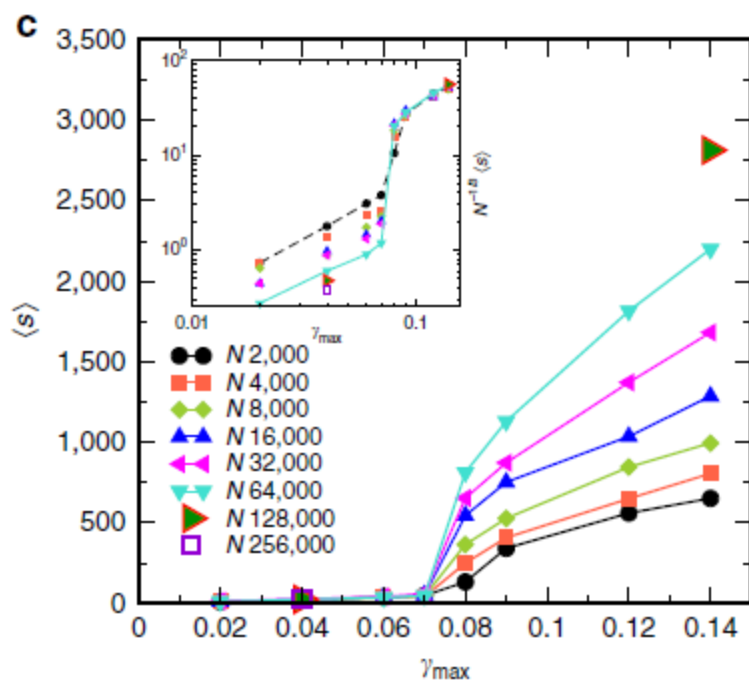
1.53

The irreversible population displays a structure akin to hard attractive disk fluid for all driving amplitudes.

In contrast, in the reversible regions, the emulsion droplets self organise to form a soft repulsive disc structure

The yielding transition in amorphous solids under oscillatory shear deformation

Premkumar Leishangthem¹, Anshul D.S. Parmar^{1,2} & Srikanth Sastry¹



Shear bands as manifestation of a criticality in yielding amorphous solids

Giorgio Parisi¹, Itamar Procaccia^{2,1}, Corrado Rinaldi³, and Murari Singh³

PNAS May 2017, p 5577

Here, we offer such a theory: The mechanical yield is a thermodynamic phase transition, where yield occurs as a spinodal phenomenon. At the spinodal point, there exists a divergent correlation length that is associated with the system-spanning instabilities (also known as shear bands), which are typical to the mechanical yield. The theory, the order parameter used, and the correlation functions that exhibit the divergent correlation length are universal in nature and can be applied to any amorphous

The relevant order parameter for the problem at hand is the overlap function Q_{ab} , which measures the distance between two configurations a and b of the same system. Denoting the position of the i th particle as \mathbf{r}_i^a in configuration a and \mathbf{r}_i^b in configuration b , we define

$$Q_{ab} \equiv \frac{1}{N} \sum_{i=1}^N \theta(\ell - |\mathbf{r}_i^a - \mathbf{r}_i^b|), \quad [1]$$

where $\theta(x)$ is the Heaviside step function, and ℓ is a constant length, which is taken below to be $1/3$ in Lennard–Jones units (numerical details are given below). Thus, $Q_{ab} = 1$ for two identical configurations, and $Q_{ab} = 0$ when the distance between the positions of all of the particles i in the two configurations exceeds ℓ . Based on the introductory discussion, we now derive an expres-

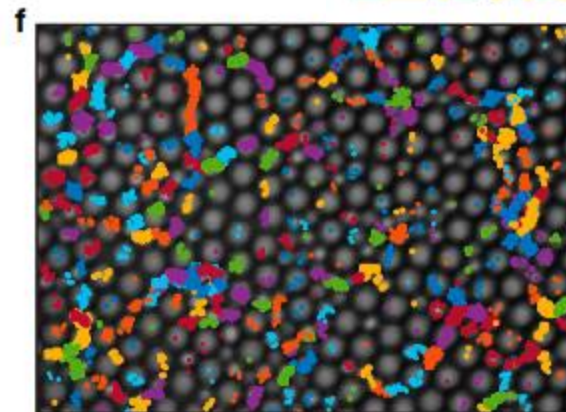
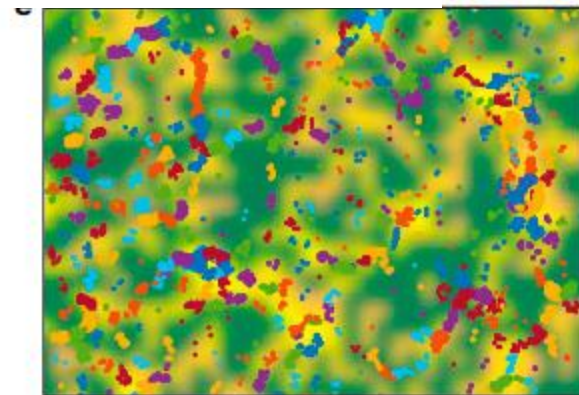
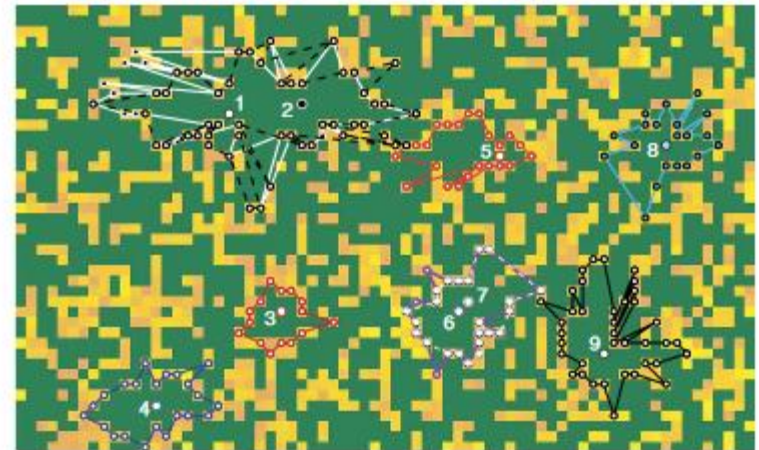
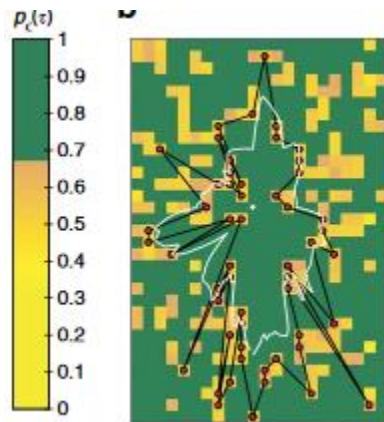
Yield as first order transition ;
Order parameter jumps .

Speculation: Where do the local plastic events occur??

At the amorphous-amorphous interface !

Measurements of growing surface tension of amorphous-amorphous interfaces on approaching the colloidal glass transition

Nat Comm (2018)

Divya Ganapathi¹, K. Hima Nagamasana^{2,5}, A.K. Sood^{1,3} & Rajesh Ganapathy^{3,4}

squares). **e** The background image represents $p_c(t = 7t^*)$ for $\phi = 0.79$. **f** Snapshot of liquid configuration corresponding to **e**. In **e** and **f**, the trajectories of the top 1% most-mobile particles are shown by the colored symbols

Thank you