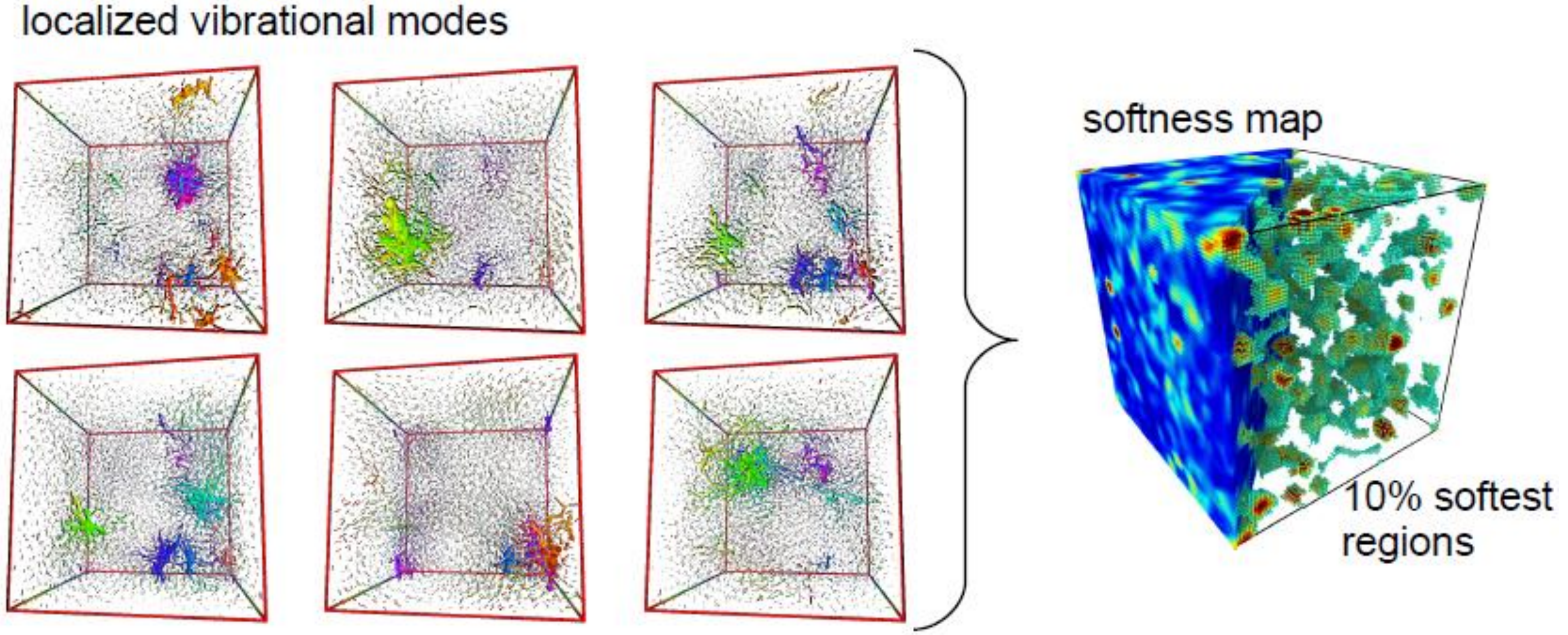
Finding structural defects with soft vibrational modes: From (athermal) crystals to (thermal) glasses



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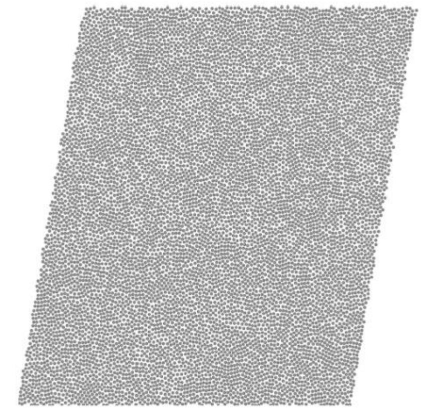
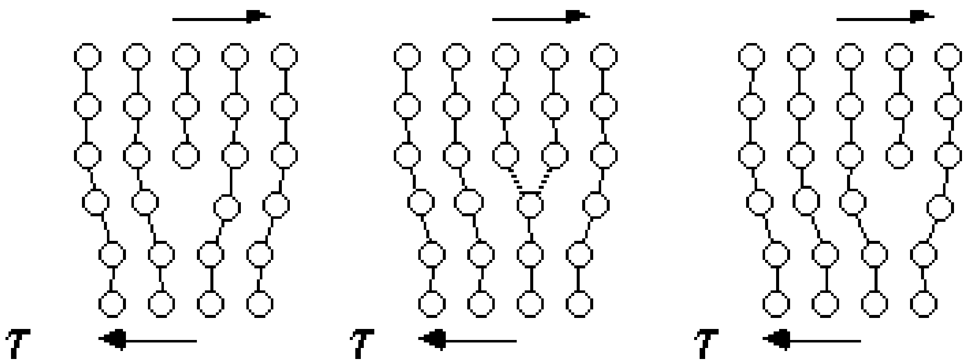
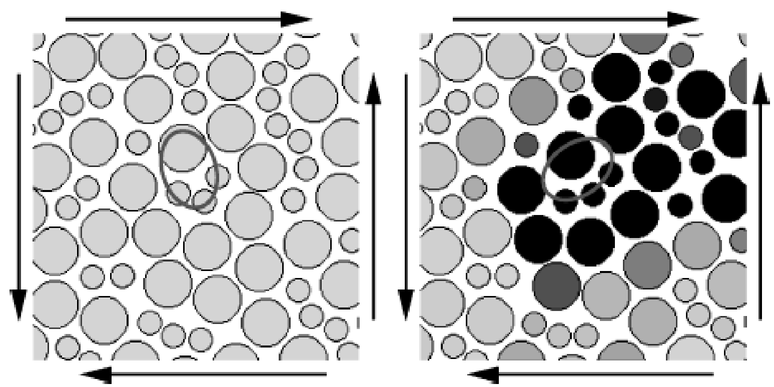
How does an amorphous solid fail?

- Flow of glassy materials is a paradigmatic topic in materials physics: polymers, amorphous metals, foams, emulsions, colloidal mixtures, yield stress fluids



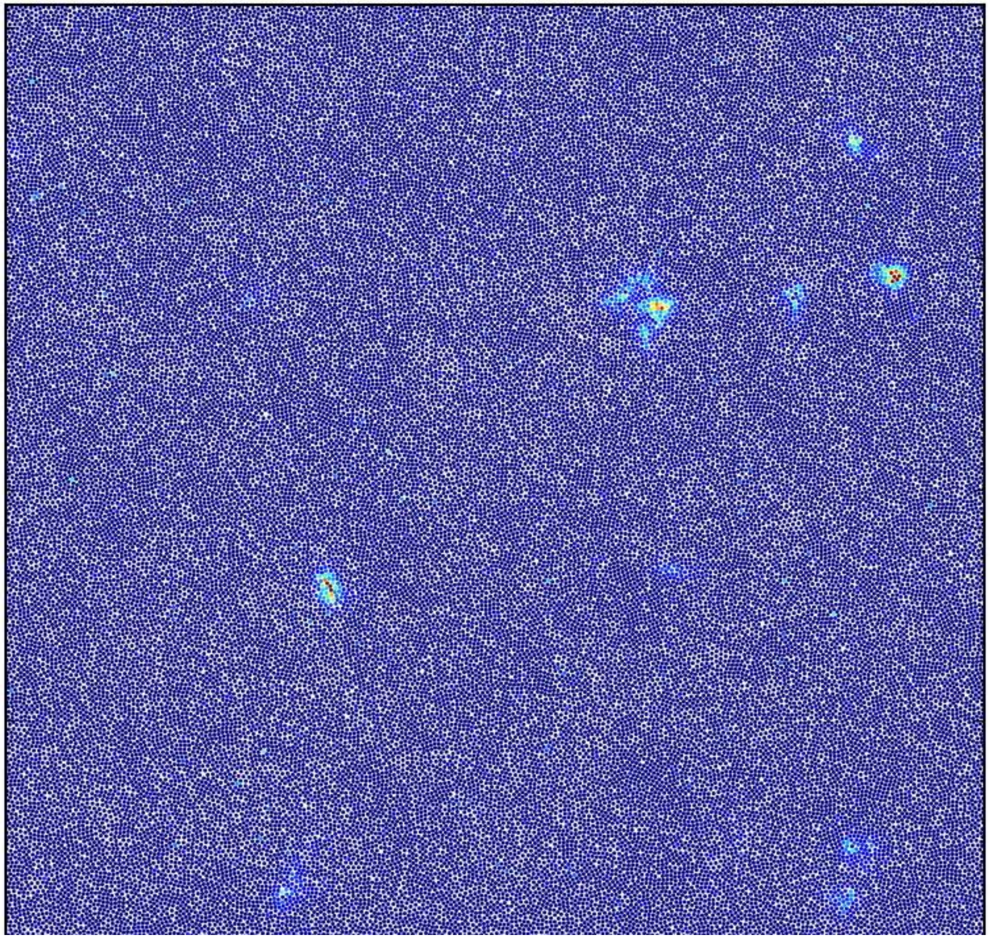




Falk and Langer, Phys Rev E (1998)

- In crystals, defects (dislocations) mediate plasticity
- In amorphous solids, there are local plastic events, but we cannot easily connect them to structural properties

Plastic activity in a flowing glass at very low T



- Can we identify a population of weak sites where rearrangements are statistically more likely to occur than elsewhere?
- How do the plastic events influence each other and how do we build up a statistical description of plastic flow?

Amorphous packing of large/small disks during simple shear

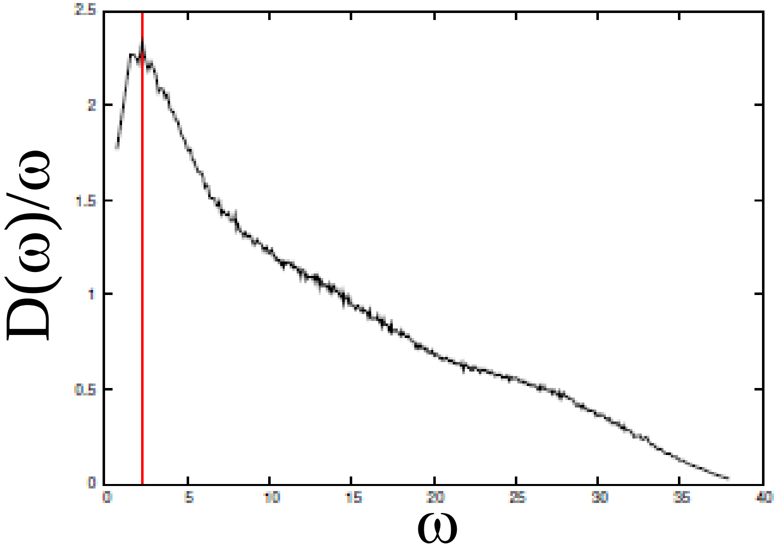
Primary tool: vibrational properties of solids

- In harmonic theory, entirely described by spectrum of dynamical matrix

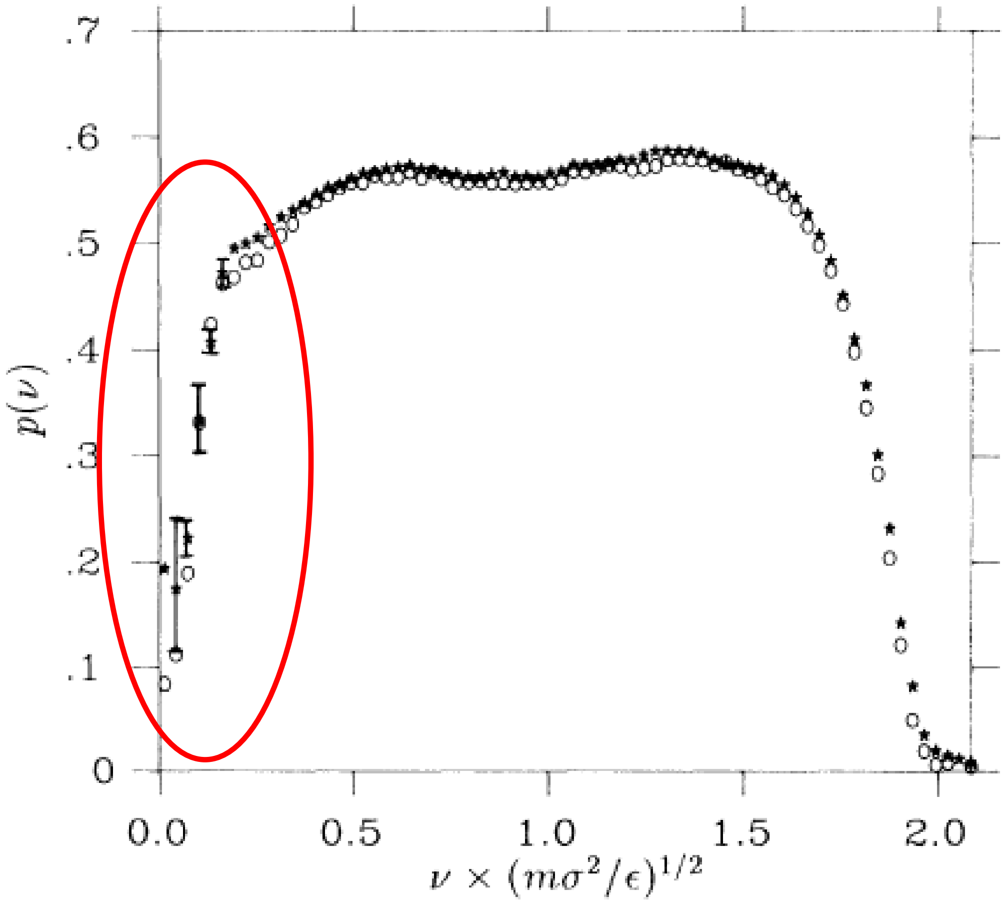
$$\boldsymbol{D}_{i\alpha j\beta} = \frac{1}{\sqrt{m_i m_j}} \frac{\partial^2 U(\boldsymbol{r}_1,\ldots,\boldsymbol{r}_N)}{\partial r_{i\alpha} \partial r_{j\beta}}$$

with the total potential energy $U(r_1, \ldots, r_N) = \sum_{i,j}^{N} u(r_{ij})$

- There are d x N eigenfrequencies ω_i and eigenvectors $\mathbf{u}_i$
- In perfect crystals, density of states has the Debye form $D(\omega) \sim \omega^{d-1}$
- Disordered materials have an excess number of modes at low frequency → Boson peak



Quasilocalized low energy modes in glasses



Participation ratio

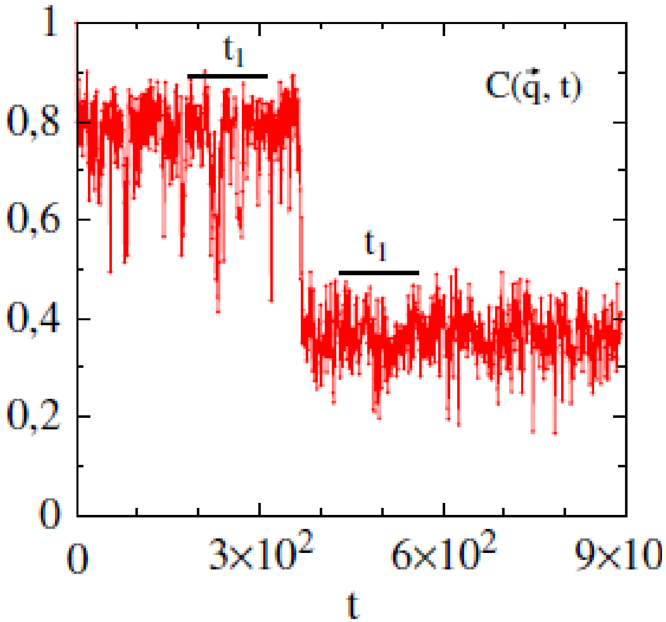
$$p(\omega_j) = \frac{1}{N} \frac{\left(\sum |u_{n,i}|^2\right)^2}{\sum |u_{n,i}|^4}$$

Laird and Schober, PRL (1991)

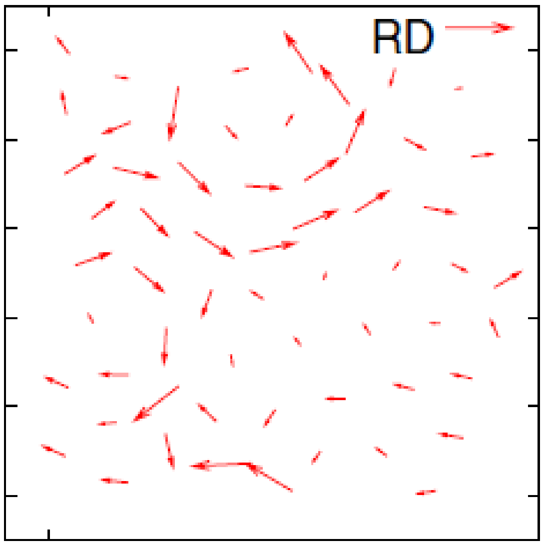
Measures degree of localization of mode:
1 for translational invariance, 3/8 for plane wave

Structural relaxation occurs along soft mode

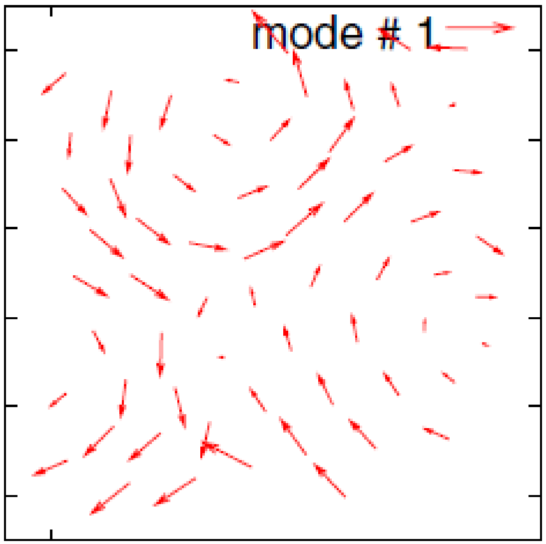
Equilibrated supercooled liquid of 64 hard discs (2D), event driven MD



Sudden jump in density correlations



Displacement field

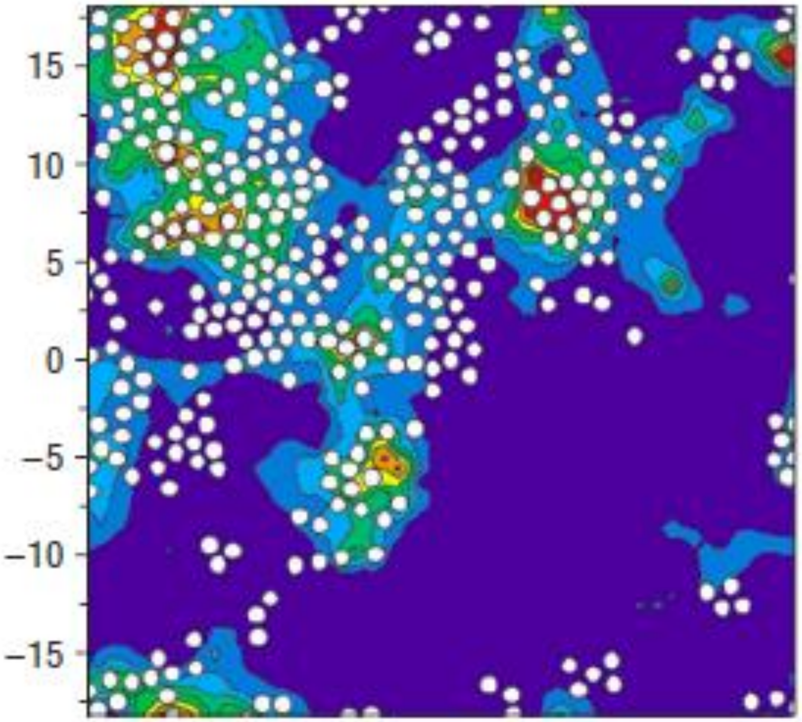


Lowest frequency normal mode

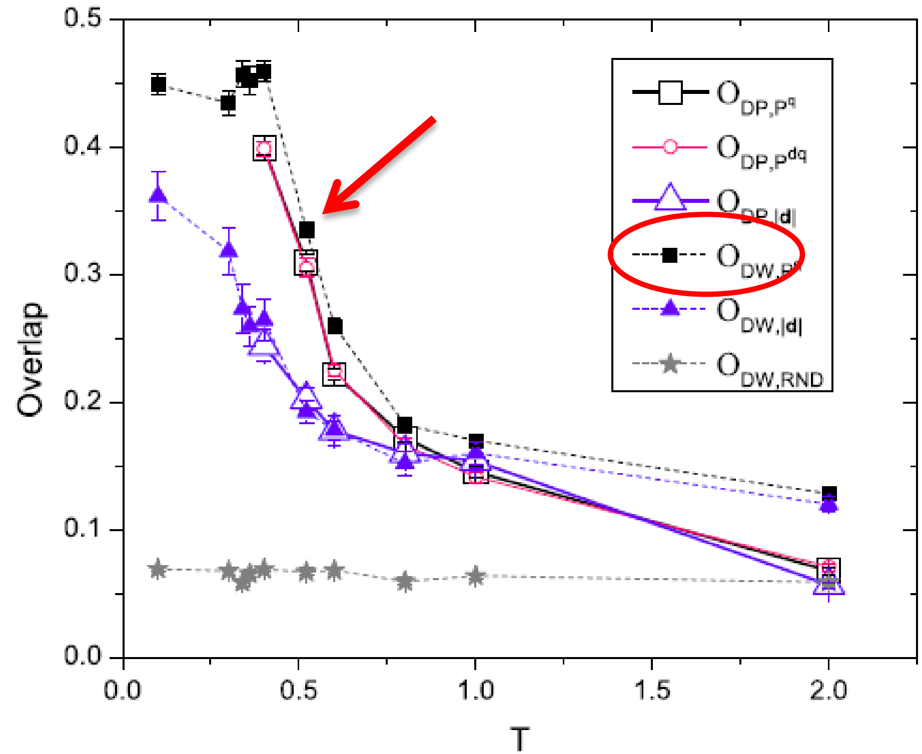
System spanning relaxation event along (a few) low energy modes

Brito and Wyart, J. Stat. Mech (2007)

Soft mode map of a supercooled liquid



Widmer-Cooper et al, Nature Physics (2008)



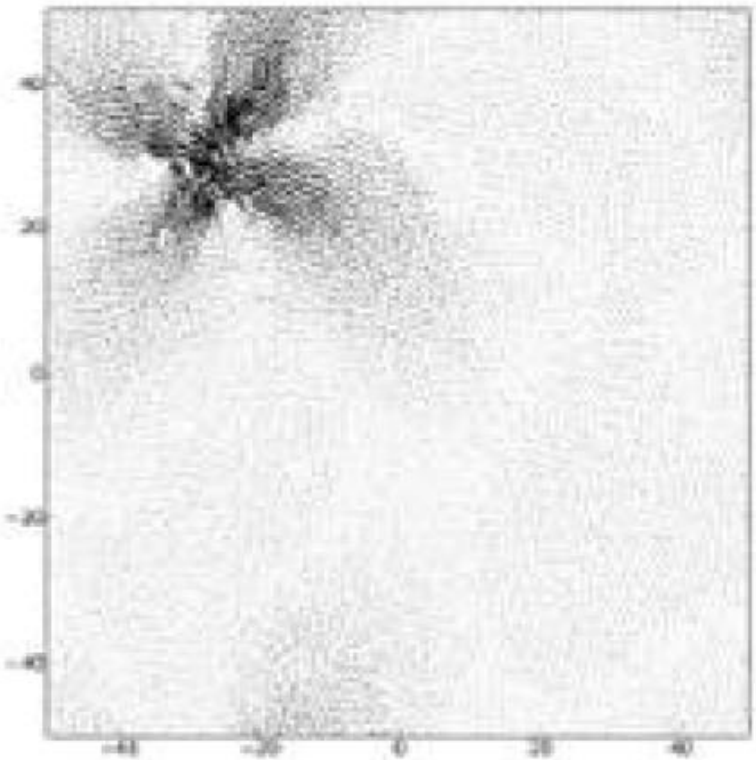
Mosayebi et al, PRL (2014)

Color map: **superposition** of 30 lowest energy normal modes (1024 LJ particles)
Qualitative correlation with irreversible rearrangements (points)

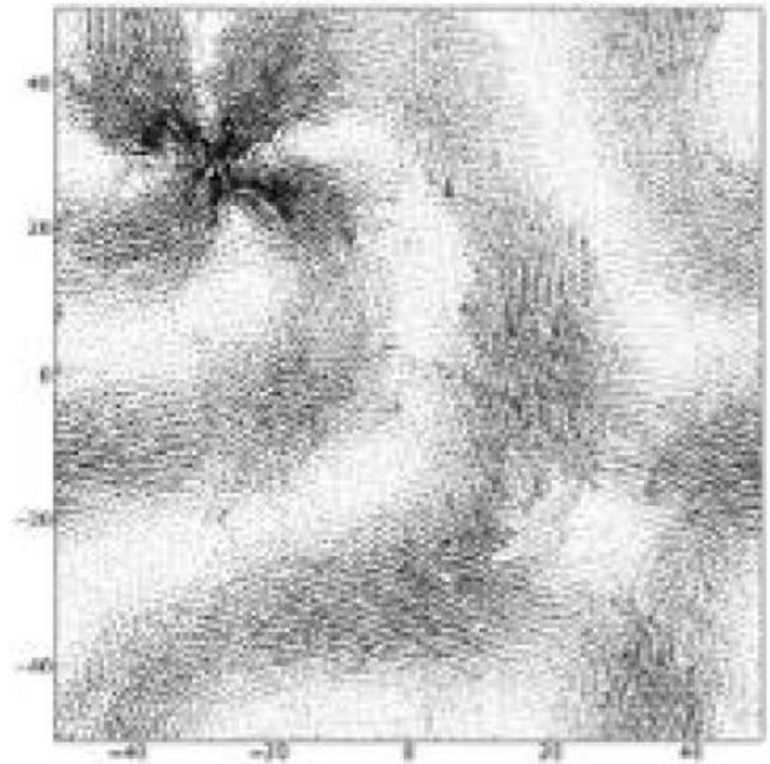
Quantitative overlap and temperature dependence

Plasticity at zero temperature

10,000 Lennard Jones particles, amorphous packing, athermal quasistatic shear



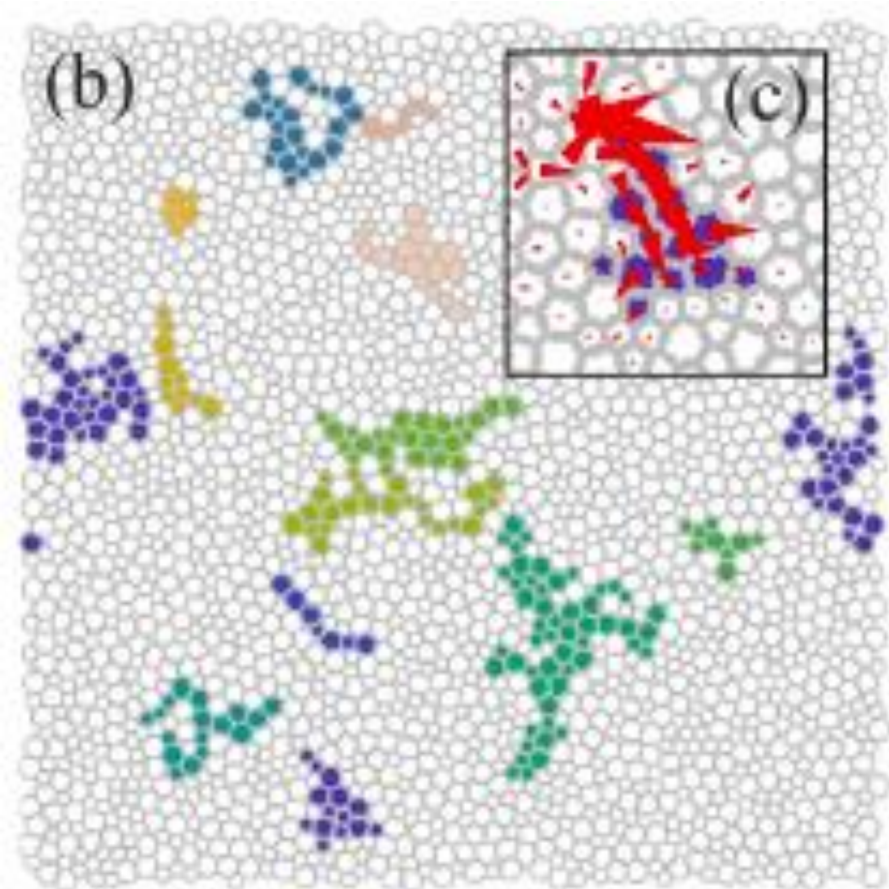
Nonaffine displacement field



Soft mode right before instability

Tanguy et al, EPL (2010)

Soft mode map of a glass → Soft Spots

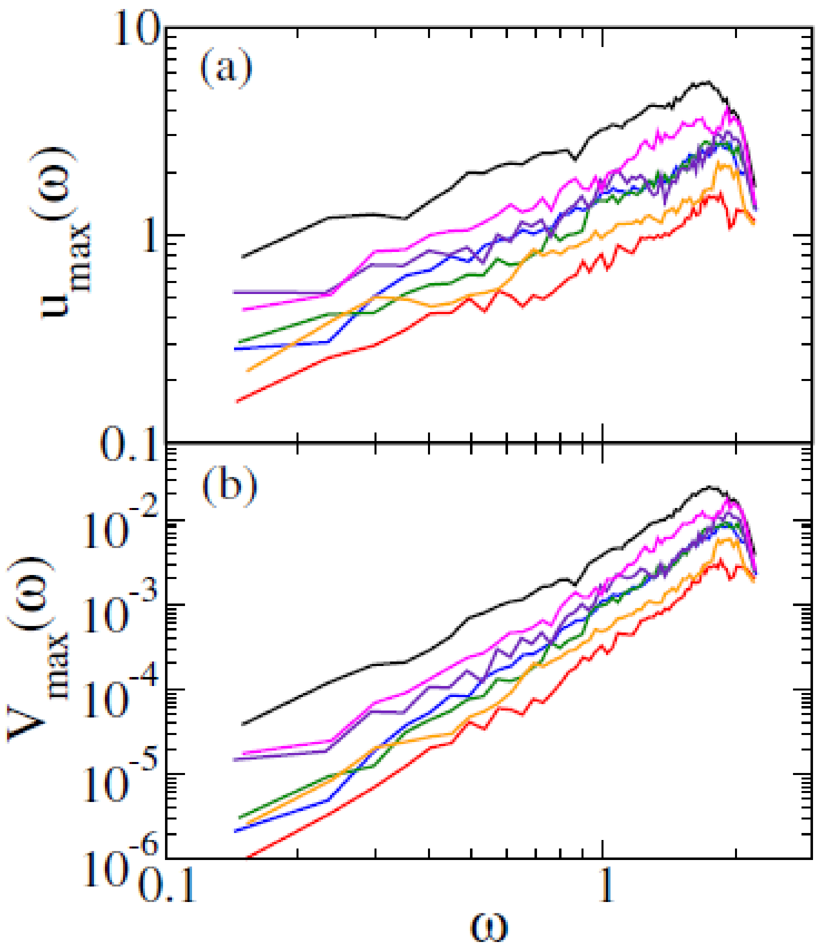


- 2,500 purely repulsive particles amorphous packing
- A(thermal) Q(uasistatic) S(hear) protocol
- Consider lowest N_m=30 modes
- Find N_p=20 particles with largest polarization vector in each mode and create (binary) map of these particles

→ The next rearrangement occurs at one of these spots: best overlap 64%

Manning and Liu, PRL (2011)

Why should this work?



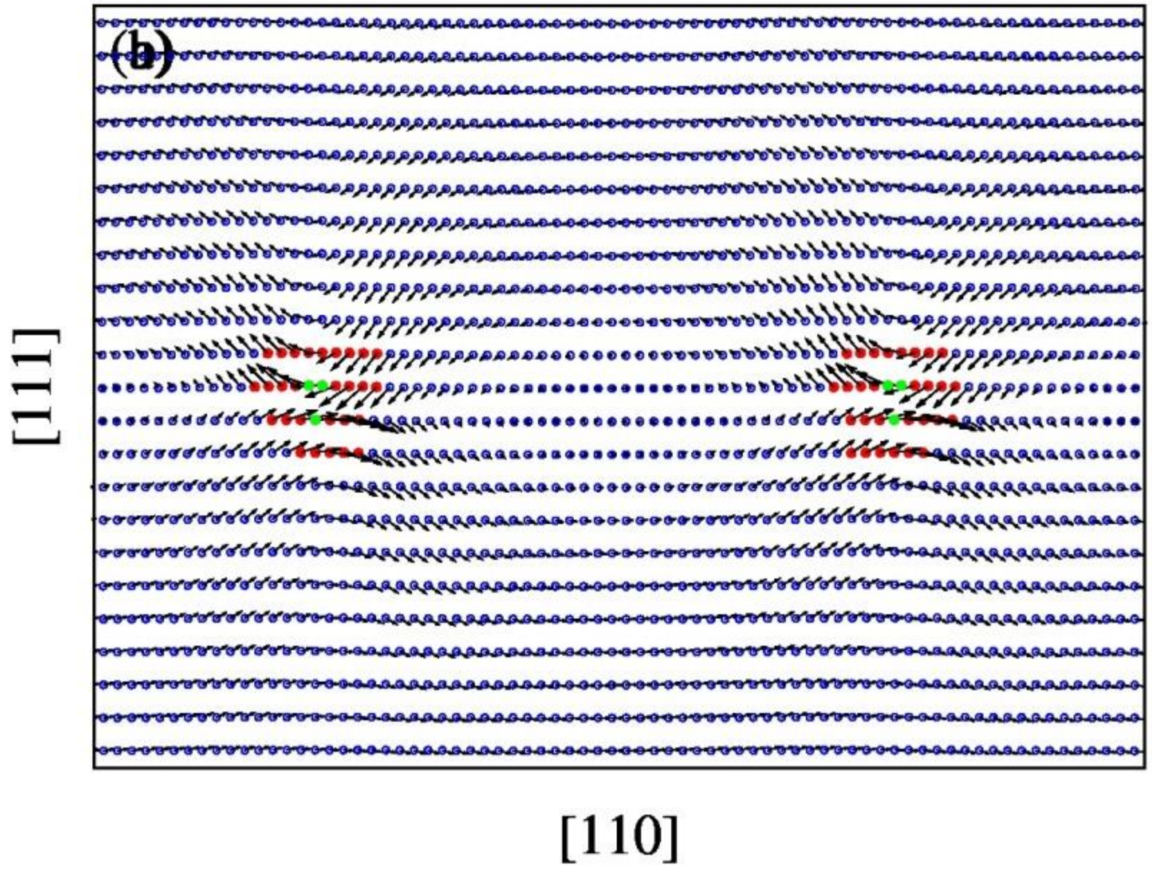
Xu et al, EPL (2010)

- Dynamical matrix relates to curvature of potential energy landscape at local minima, but plastic events require barrier crossing?
- But: barrier height $V \sim \omega^2$
- So low frequency modes are least stable, most easily driven by thermal energy or deformation

Question 1:

Can soft modes generically identify defects in solids, be they crystalline, polycrystalline, or amorphous?

Test case: split partial dislocation in fcc crystal



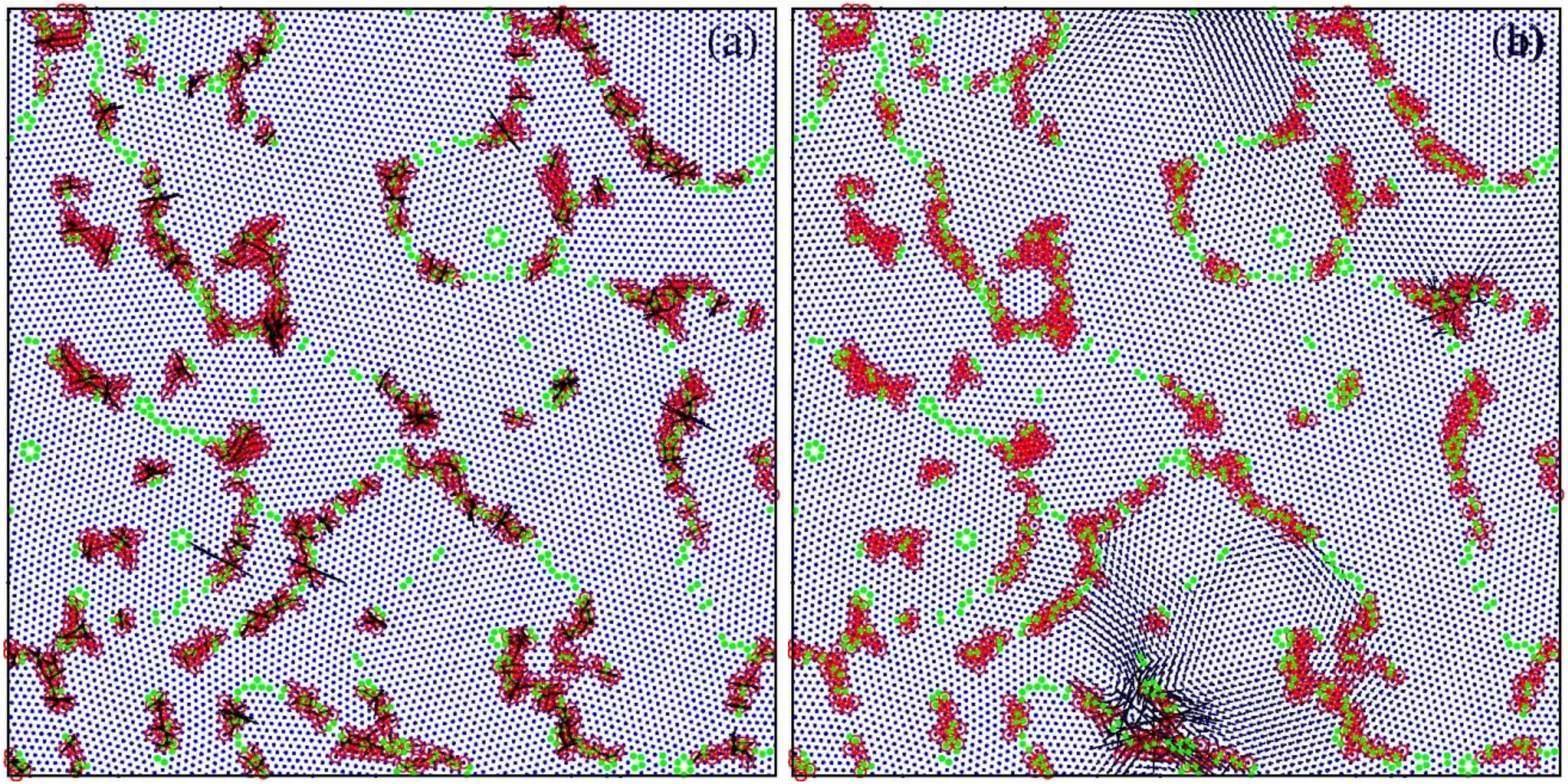
Displacement field resulted from partial dislocation glide upon 0.1% shear

Low energy mode localized at 3d partial dislocations

Dislocation core green, soft spot red

Polycrystal

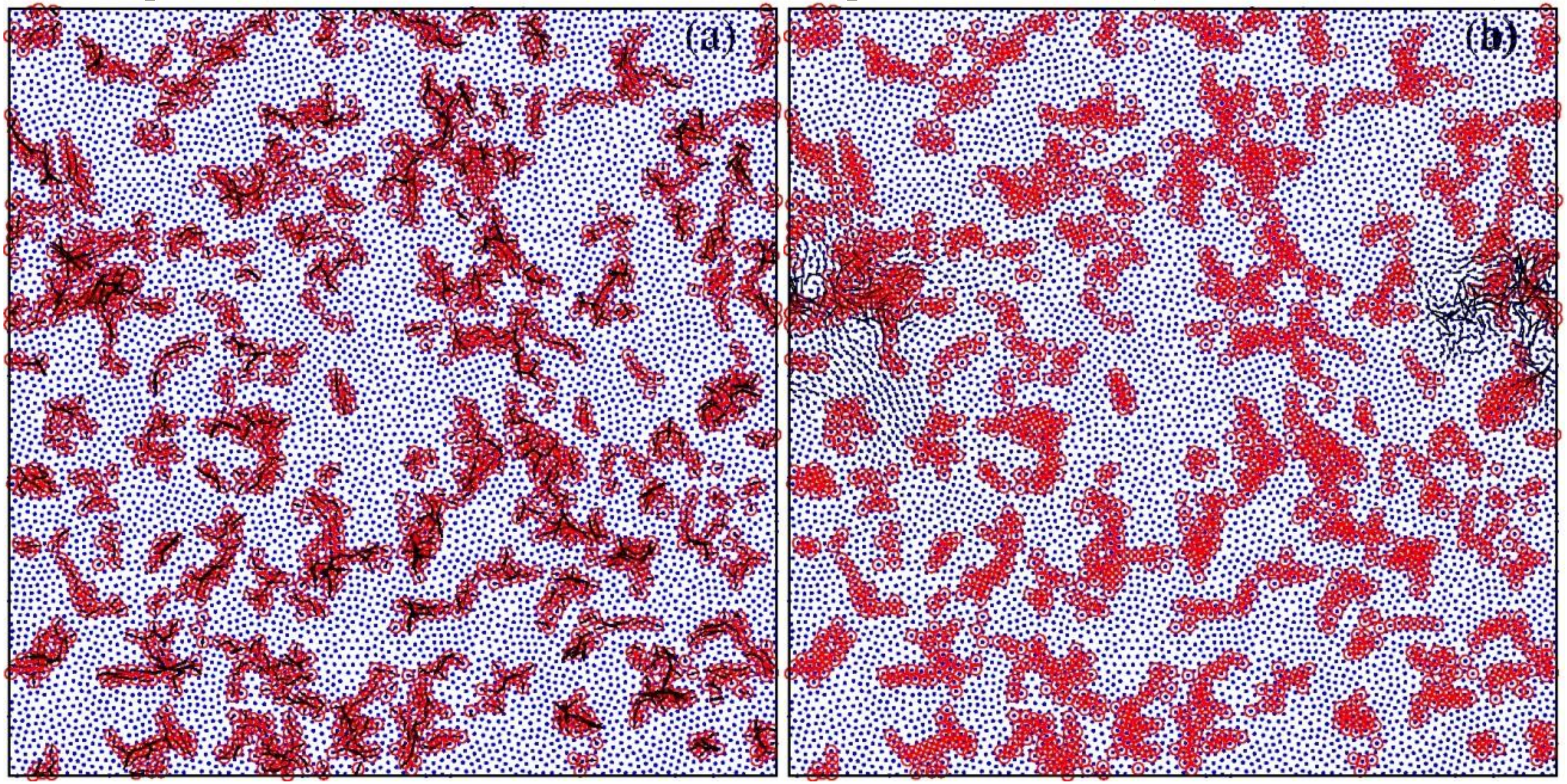
Soft spots with soft directions	Displacement field (0.1% shear at T=0)



- 10,000 Lennard-Jones particles, 300 modes, 30 particles → 9% soft spots
- Green: dislocations at gain boundaries, Red: soft spots

Glass

Soft spots with soft directions   Displacement field (0.1% shear at T=0)

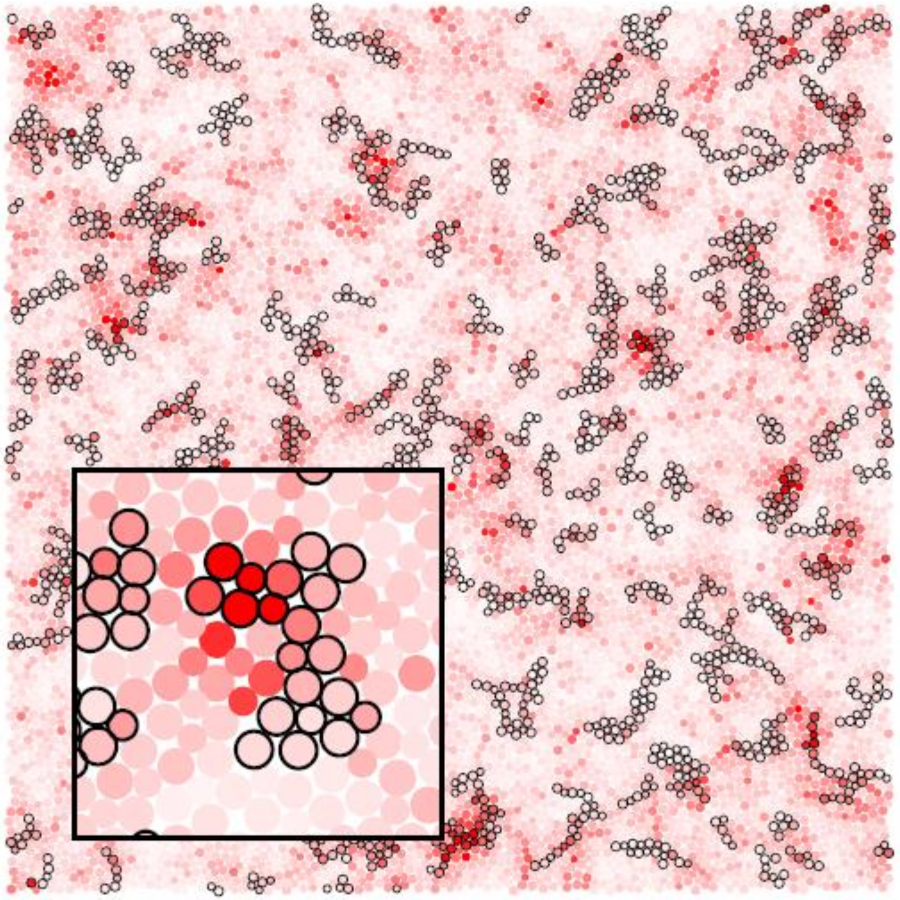


- 10,000 Lennard-Jones particles, 250 modes, 30 particles (~ 20 % soft spots)

Soft spot – plastic activity overlap (T=0)

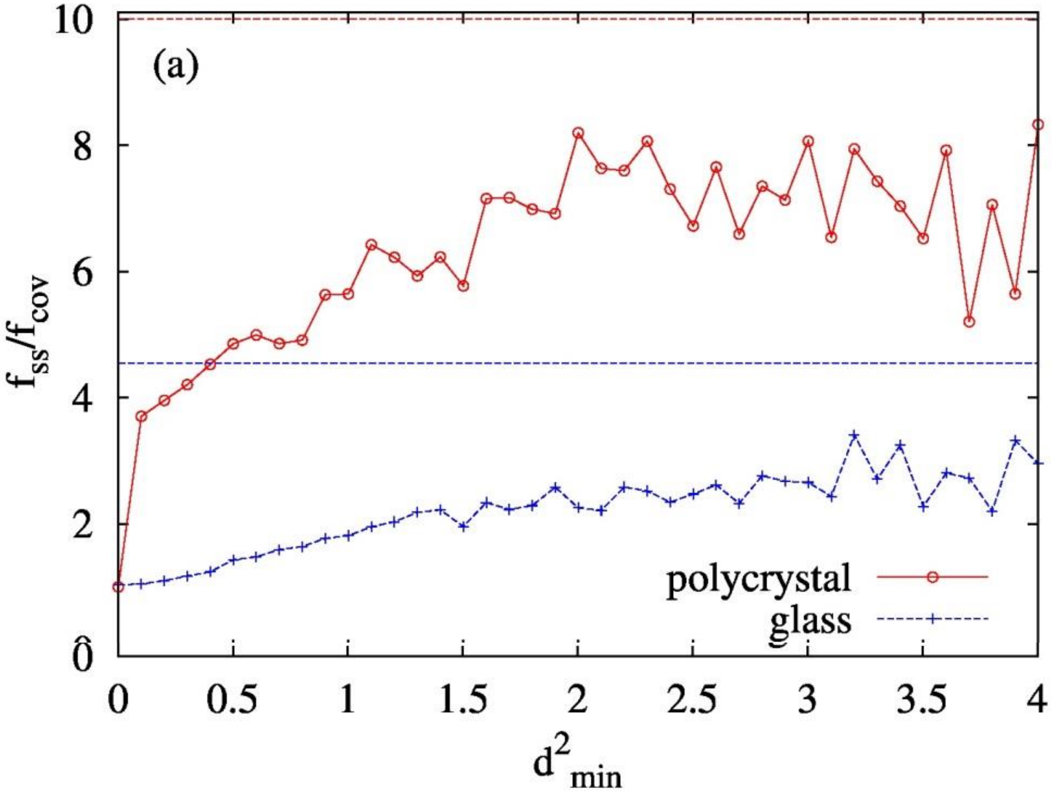
$$d^2_{min} = \sum_n \sum_{i=1}^{3} \left(r_n^i(t) - r_0^i(t) - \sum_j \left(\delta_{ij} + \varepsilon_{ij} \right) \left[r_n^j(t-dt) - r_0^j(t-dt) \right] \right)^2$$

"compute local strain tensor in neighborhood of particle, minimize mean squared difference between displacements of atoms relative to central one and those they would have if strain were uniform"



Color: local value of d^2_{min}, black circles: soft spots

Soft spot – plastic activity overlap (T=0)



→ Plastic event is 3 (glass) – 8 times (polycrystal) more likely at soft spot than anywhere else

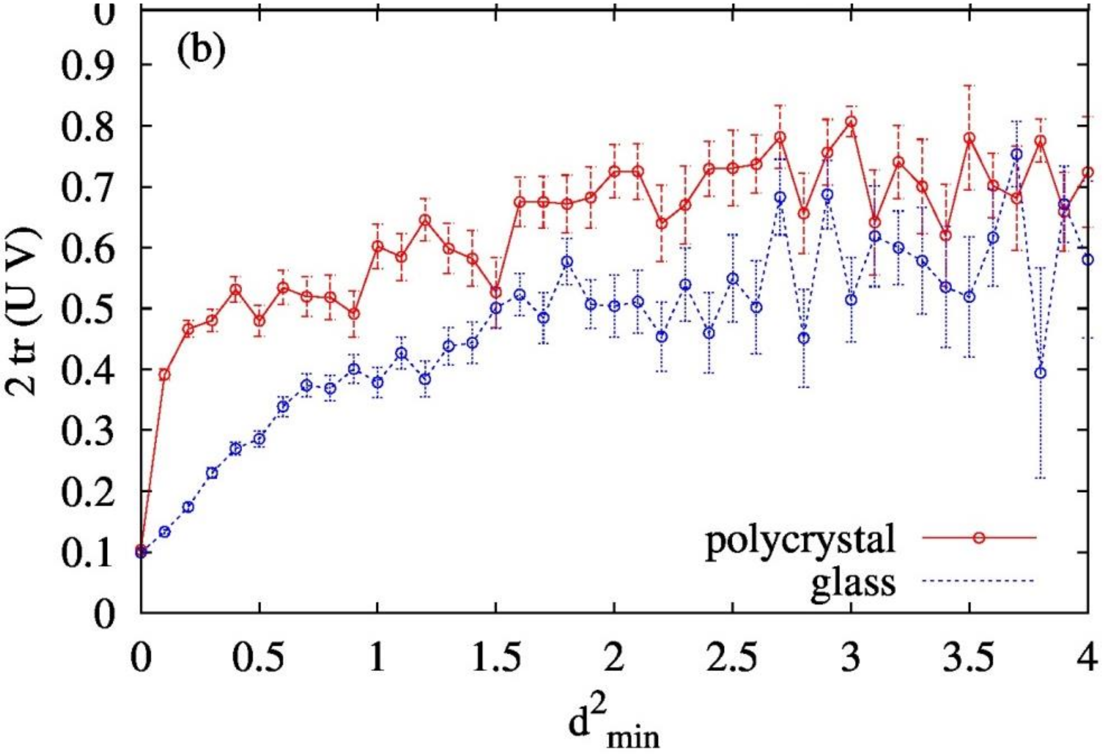
JR, S.Schoenholz, A.J. Liu, Phys Rev E (2014)

Question 2:

Can soft modes predict not only where structural relaxation or plastic events will occur, but also how?

Alignment soft direction - displacement

Order parameter for alignment btwn displacements $\mathbf{v}_i$ and soft directions $\mathbf{u}_i$: trace of product of nematic tensors



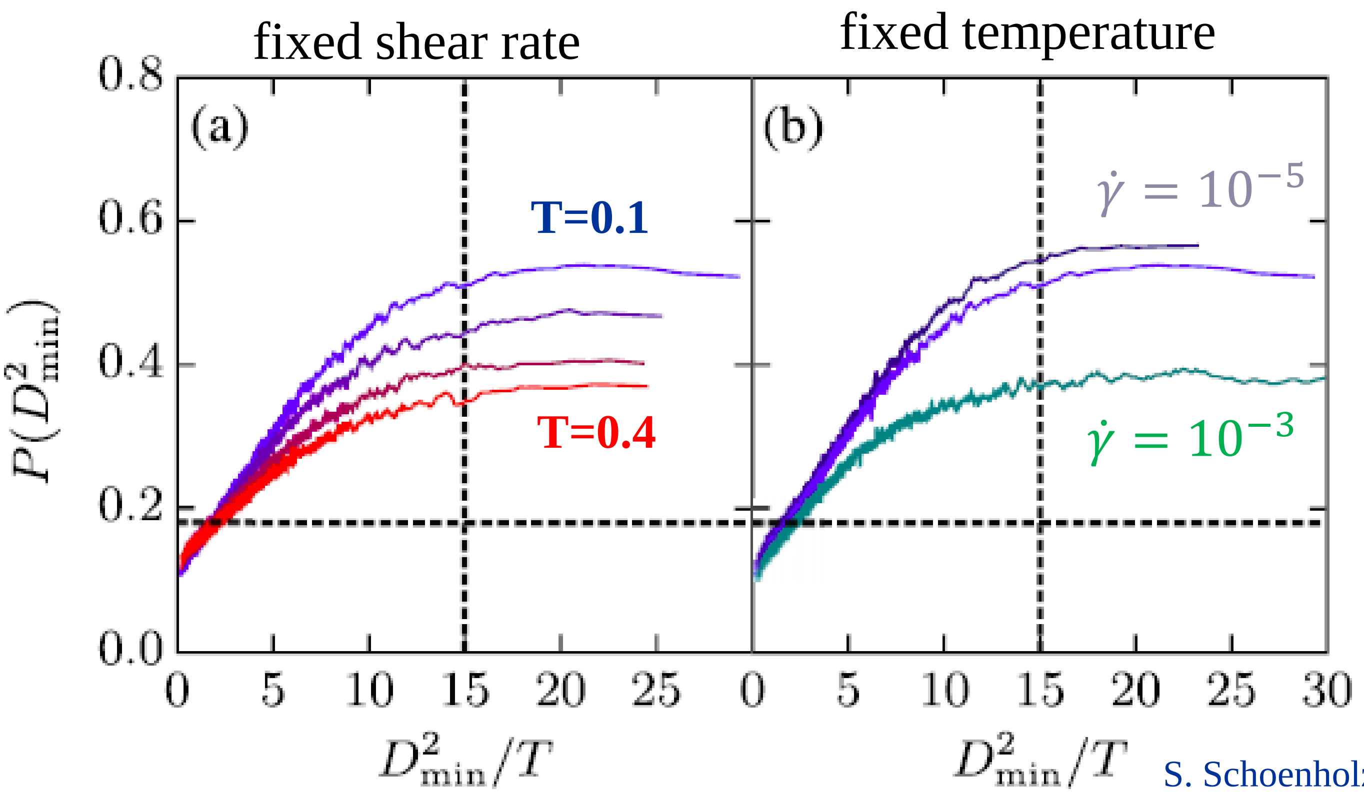
→ saturates between 0.55 (glass) – 0.7 (polycrystal)

JR, S.Schoenholz, A.J. Liu, Phys Rev E (2014)

Question 3:

What about finite temperature and finite shear rate?

Finite temperatures



S. Schoenholz et al, Phys Rev X (2014)

- Plastic intensity scales ~ T
- Overlap plastic event/soft spots comparable to T=0 even for a supercooled liquid!
- Overlap decreases with increasing shear rate

Question 4:

Are the soft spot maps long lived structural features of the glass?

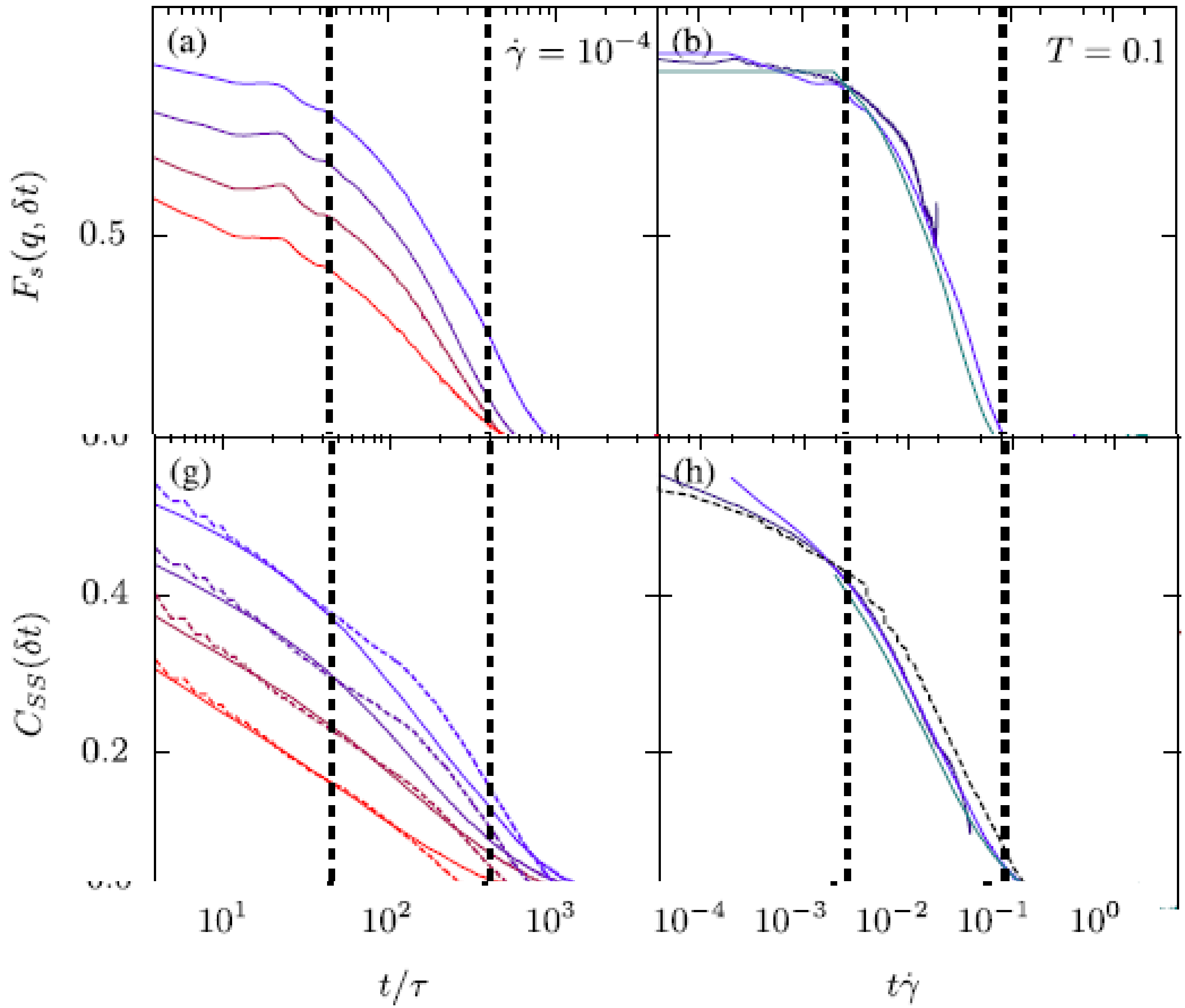
Correlations

Self-intermediate scattering function:
Measures particle mobility

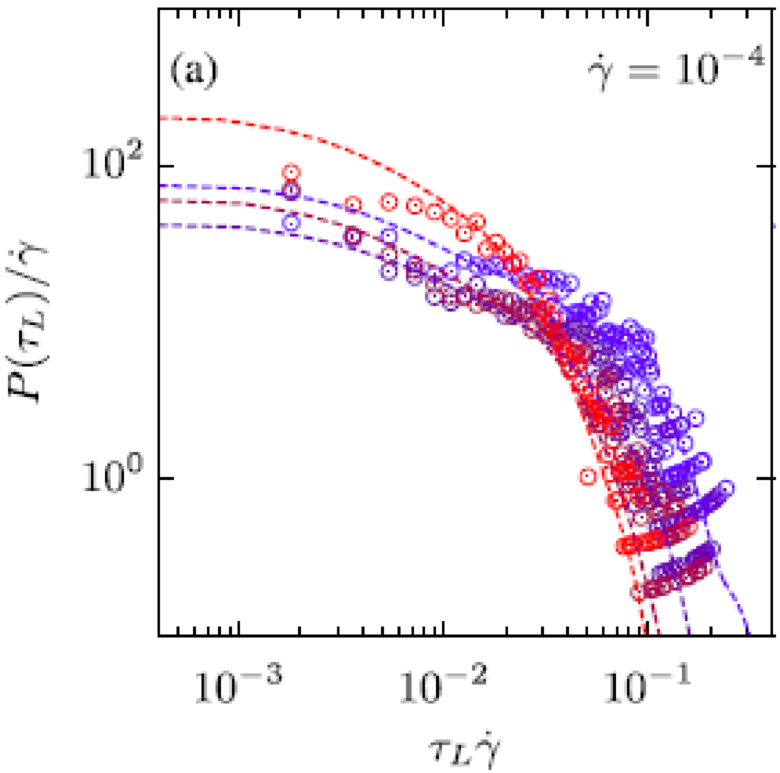
Soft spot autocorrelations:
Measures lifetime

→ slow decorrelation
→ collapse with strain at low T: driven regime
→ almost every particle needs to rearrange to decorrelate soft spots

vary temperature   vary strain rate



Soft spot lifetime distributions

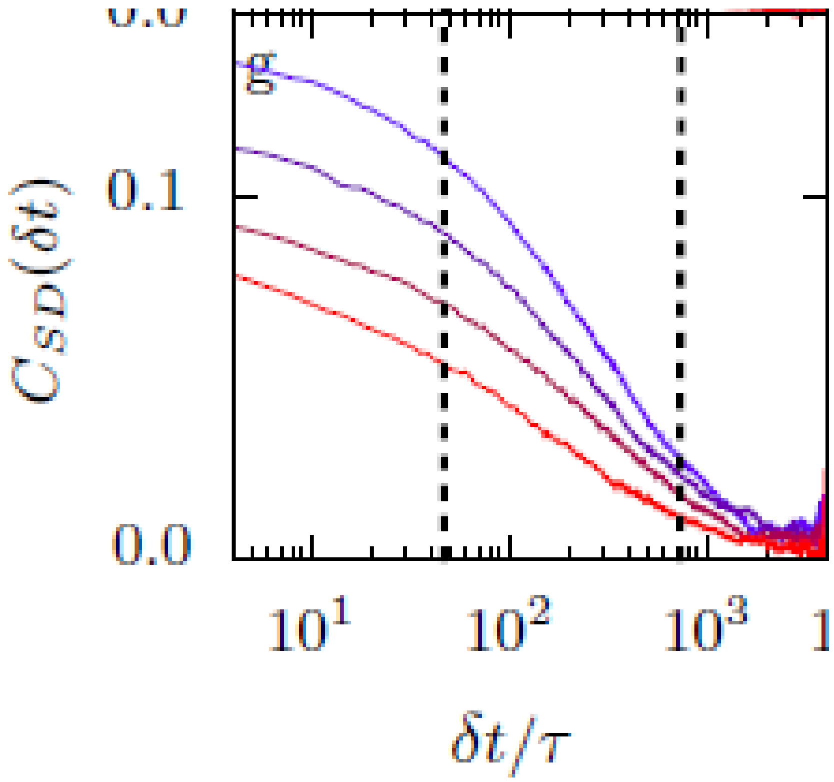


- Measure decay of acf of individual soft spots, extract lifetime distribution
- Power law with exp cutoffs
- $C_{ss}(\delta t)$ is probability that soft spot has not yet decayed, so

$$C_{ss}(\delta t) \sim 1 - \int_0^{\delta t} P(t)dt$$

→ Soft spot field behavior can be reduced to single soft spot dynamics
→ Soft spots survive multiple rearrangements before destruction

Soft spot – plasticity cross-correlations



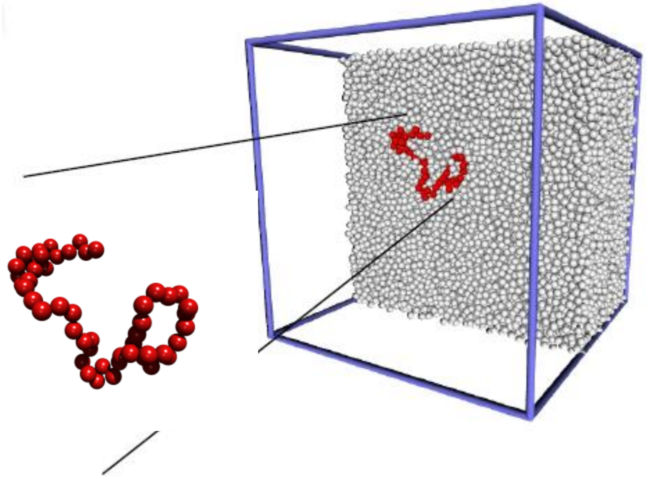
4 different temps.

- Logarithmic decay from overlap plateau value to random overlap
- Random overlap reached when strain ~ 0.1 (yield strain)

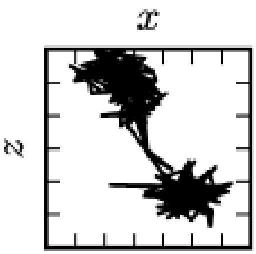
Question 5:

How robust are these findings in 3D glasses (most studies so far focused on 2D mixtures of discs)?

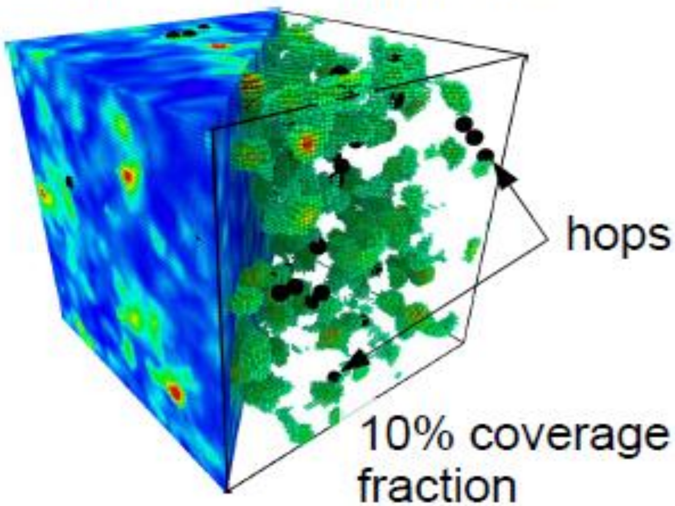
Softness map in polymer glasses



New (3D) glass former: bead-spring polymer chains, 10,000 particles



New measure of rearrangement: find particle hops from trajectories (cage escape)

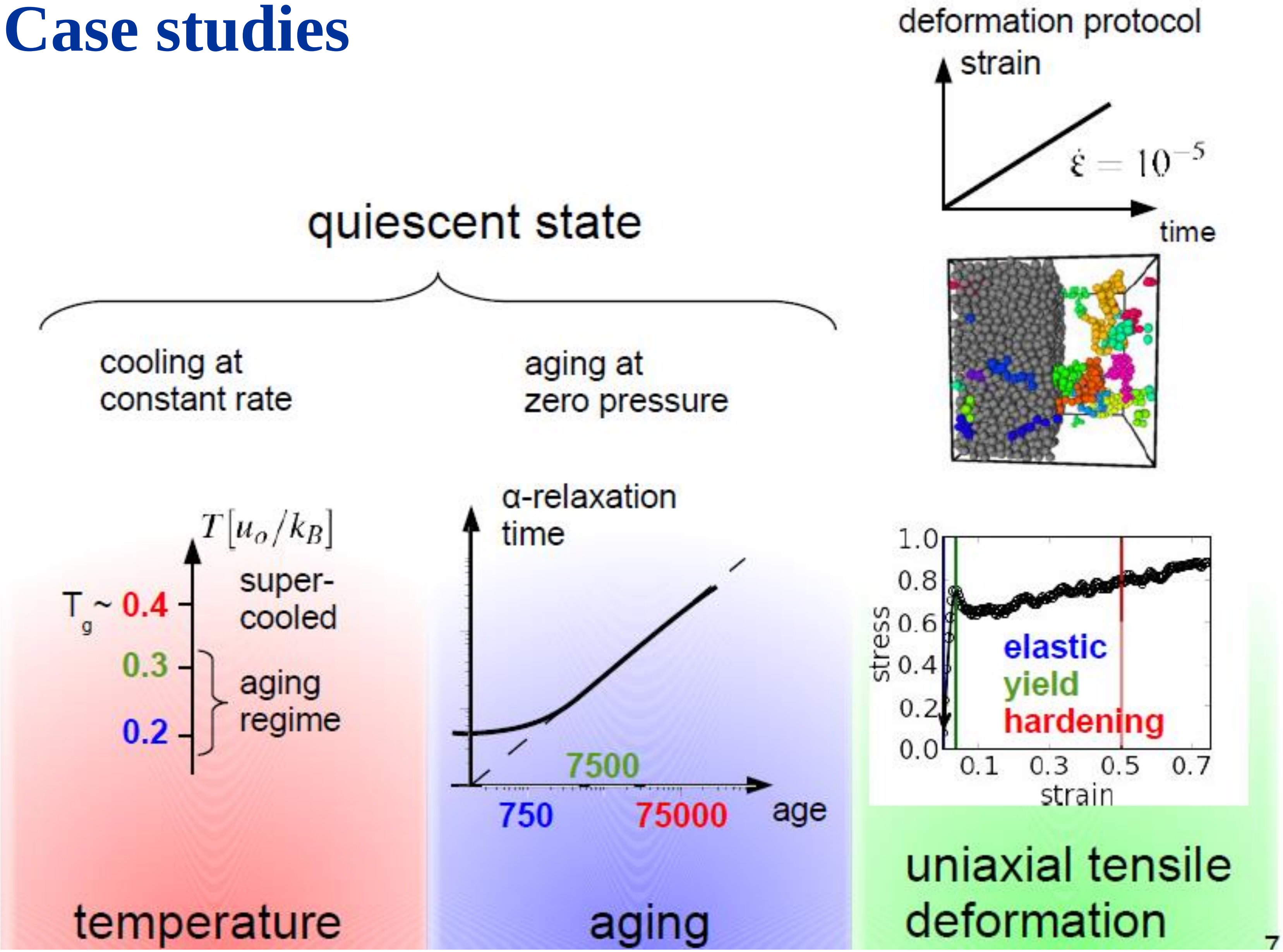


Consider continuous "softness field":

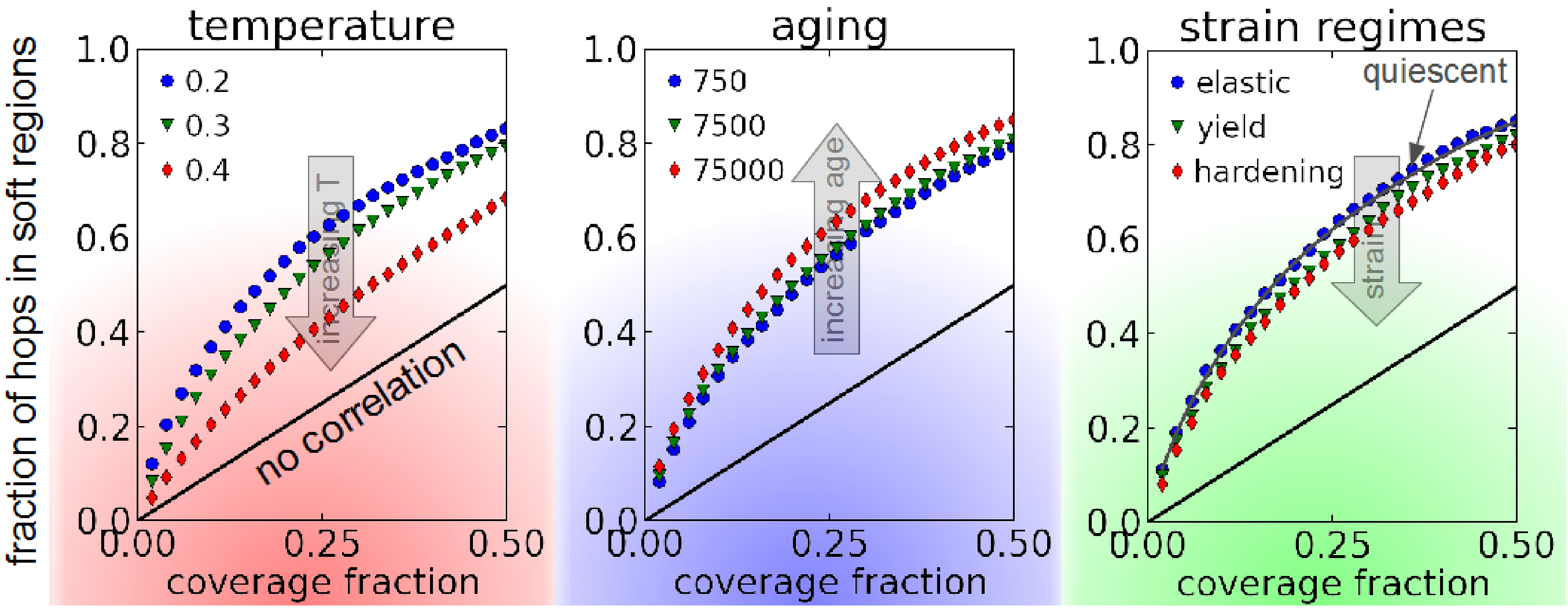
$$\phi_i = \frac{1}{N_m} \sum_{j=1}^{N_m} \left| \mathbf{e}_j^{(i)} \right|^2$$

A. Smessaert, JR, Soft Matter (2014)

Case studies



Softness map – particle hop overlap



- Overlap larger than random, maximum at ~ 25% coverage
- Aging: overlap increases (deeper traps)
- Deformation: overlap “rejuvenates” to as-quenched material

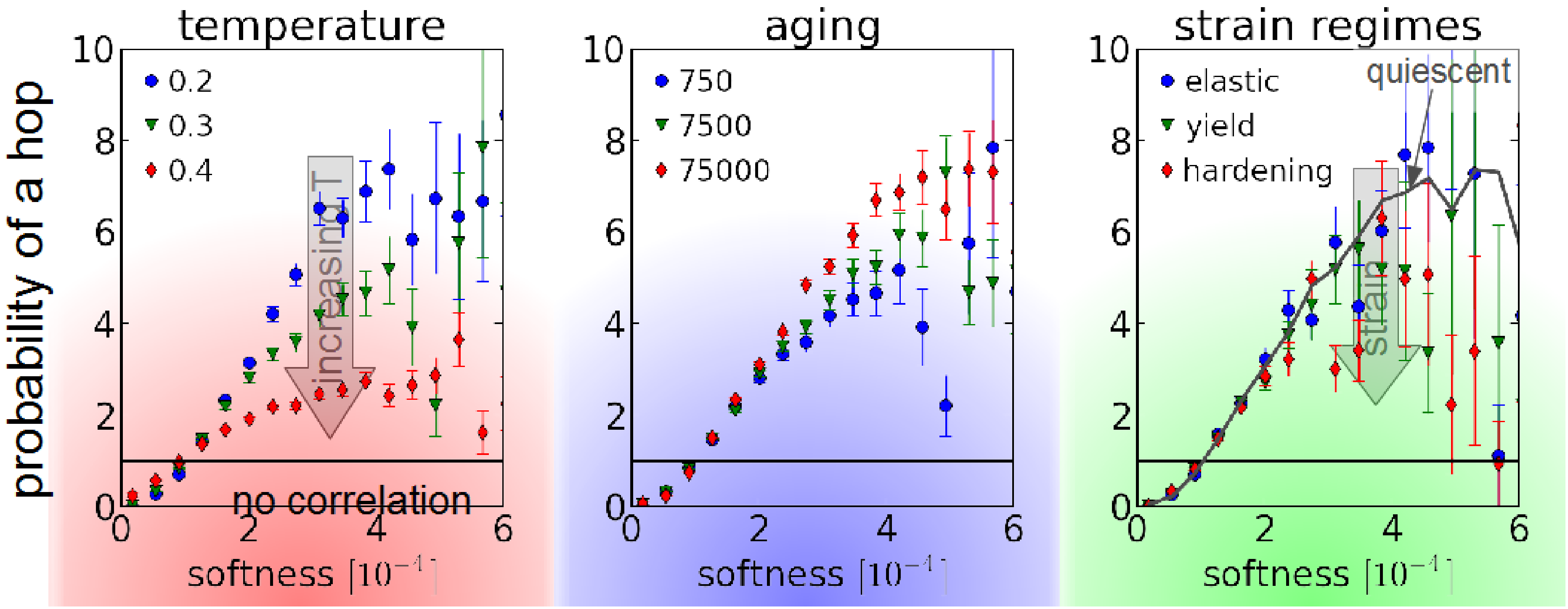
A. Smessaert, JR, Soft Matter (2014)

Hop probability grows with softness

- what is the probability of a particle to hop given its softness?
- in terms of the average hop probability

$$p_{hop}(\phi) = \frac{N_h(\phi)}{N(\phi)} \Big/ \frac{N_h}{N}$$

hop of soft particle up to 7x more likely than average

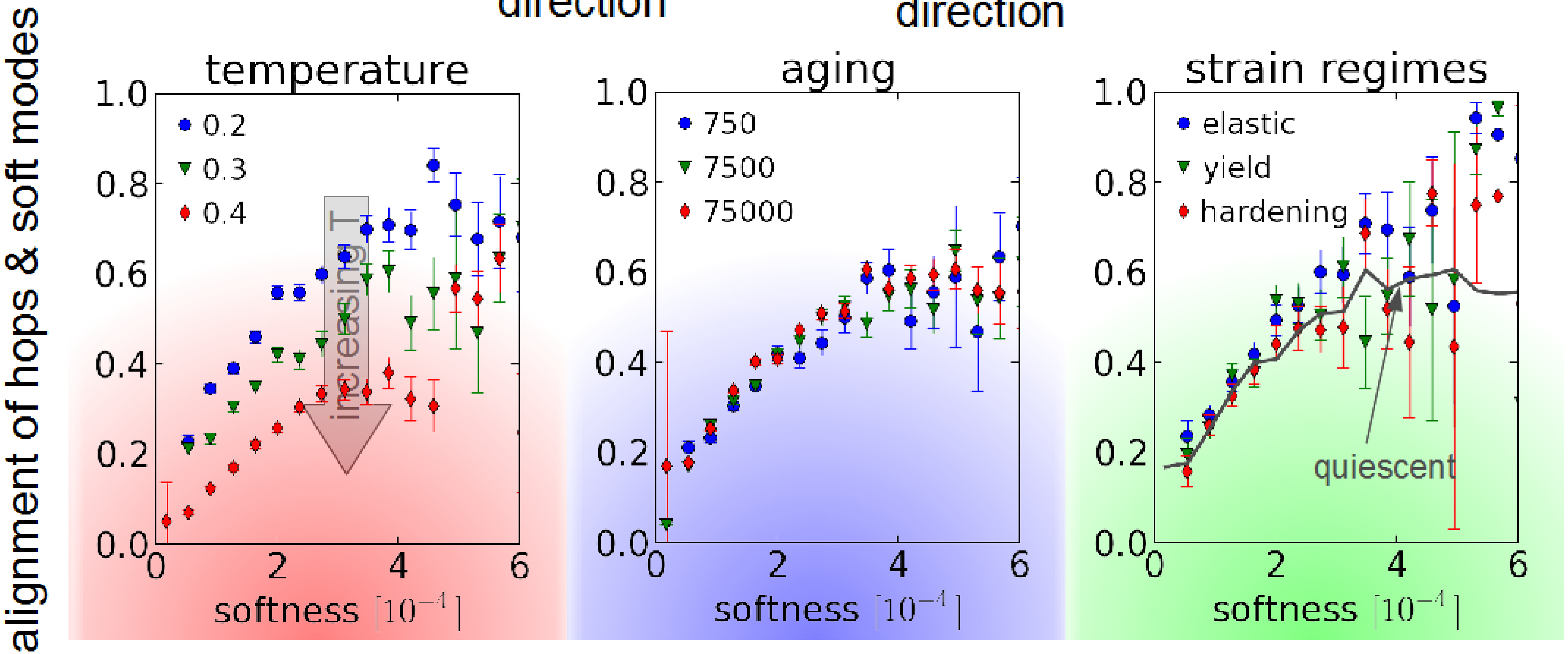


Hops occur preferentially along soft directions

$$C_d = \left\langle \frac{3}{2}\left(\hat{\mathbf{d}}_{hop}\cdot\mathbf{e}_{\phi}\right)^2 - \frac{1}{2}\right\rangle$$

hop direction

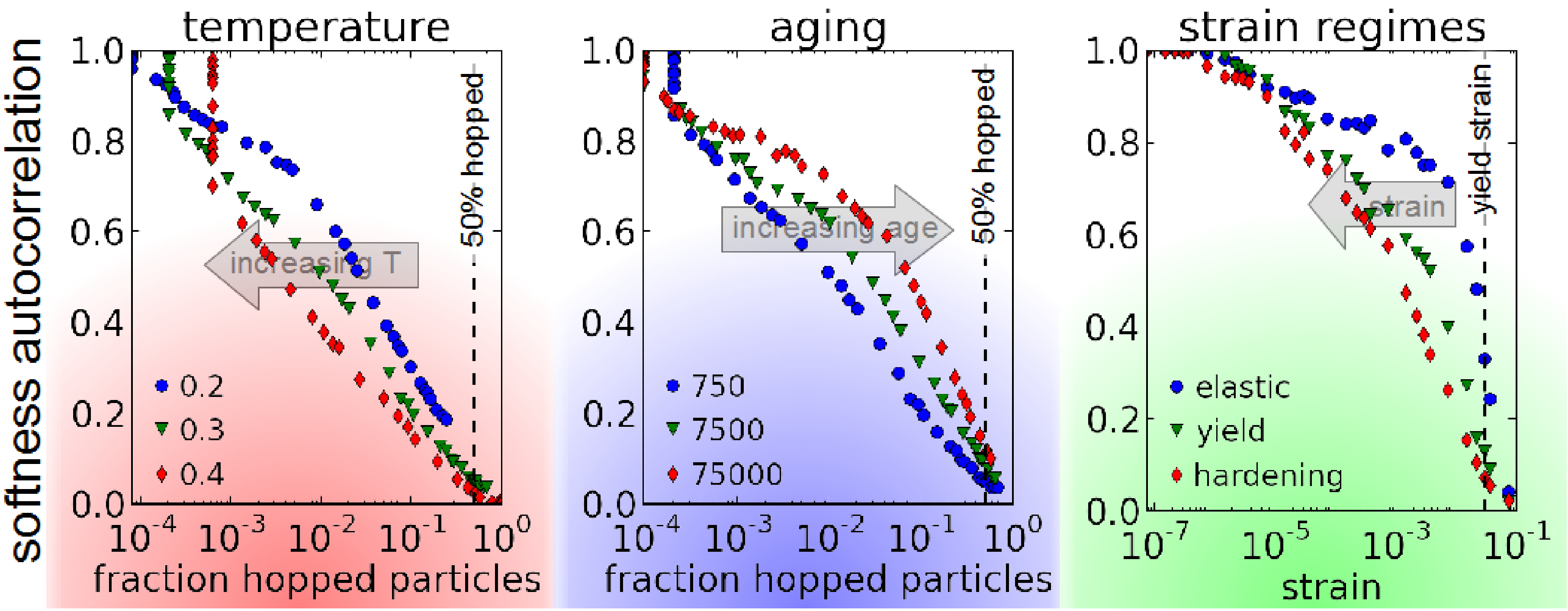
mean soft mode direction



Temporal stability of soft spot map

- autocorrelation measures longevity of softness
- quiescent systems: decorrelates after ~50% of particles have undergone rearrangements
- during deformation: decorrelates after yield strain is reached

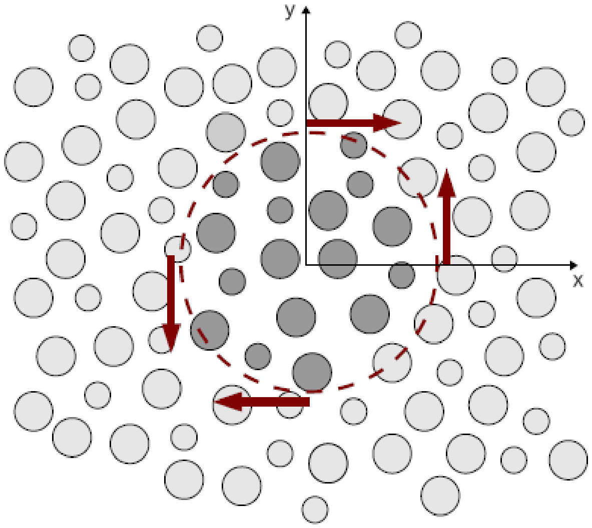
lifetime of soft regions of order alpha-relaxation time

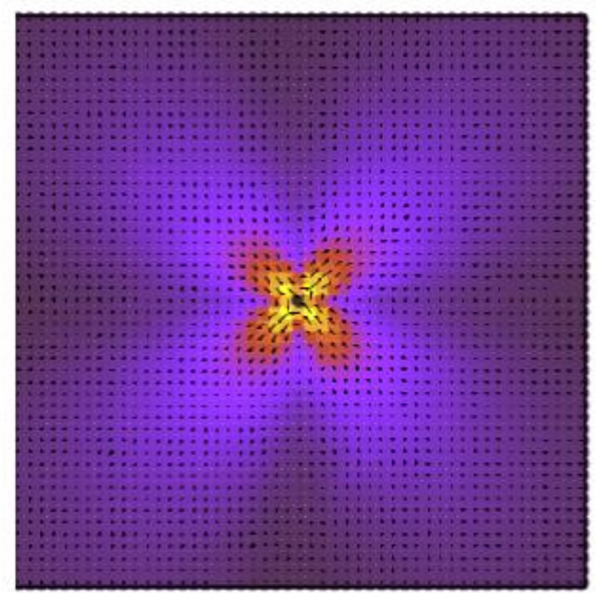


Question 6:

How do plastic events influence each other?

Elastic response to a local shear transformation





strain circular inclusion into ellipse

long time mean displacement field

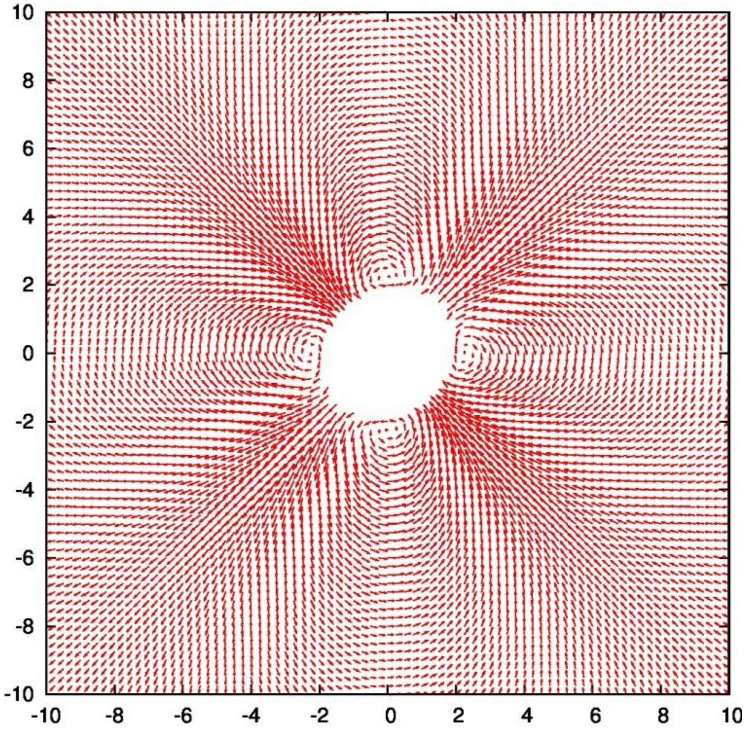
F. Puosi, JR, J-L. Barrat, PRE (2014)

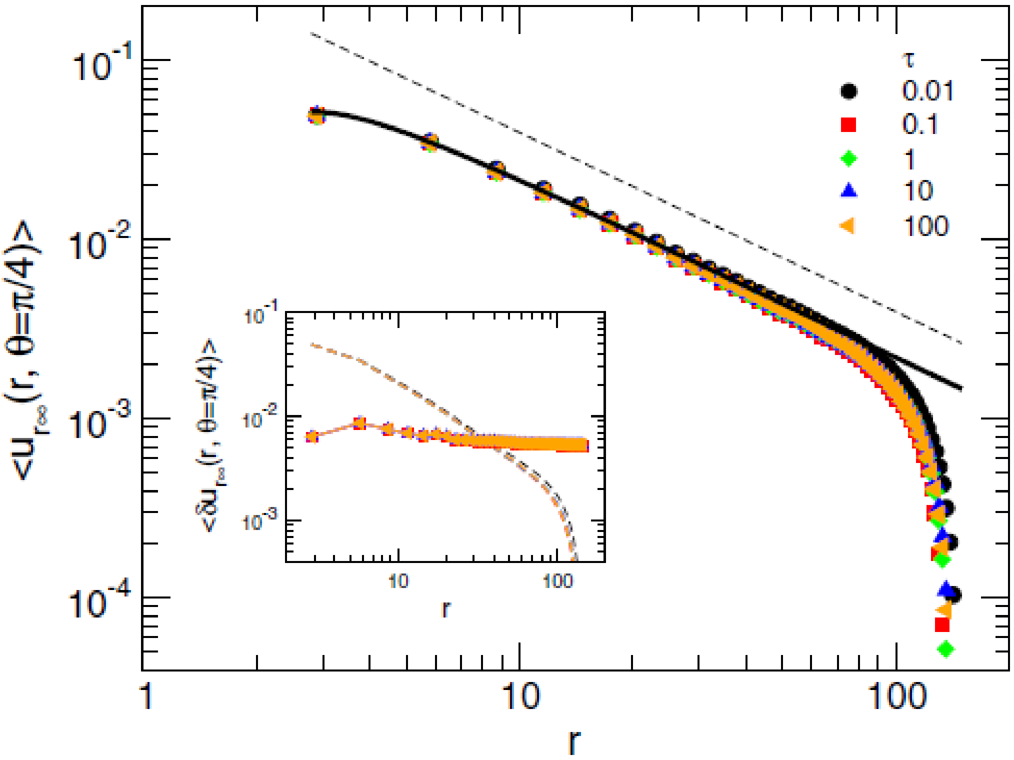
- perturbation: two force dipoles at the origin
- elastic Green's tensor:

$$G(r,\vartheta) \sim \frac{\cos(4\vartheta)}{r^2}$$

cf. Eshelby inclusion problem (1957)

Elastostatic solution (Eshelby inclusion)

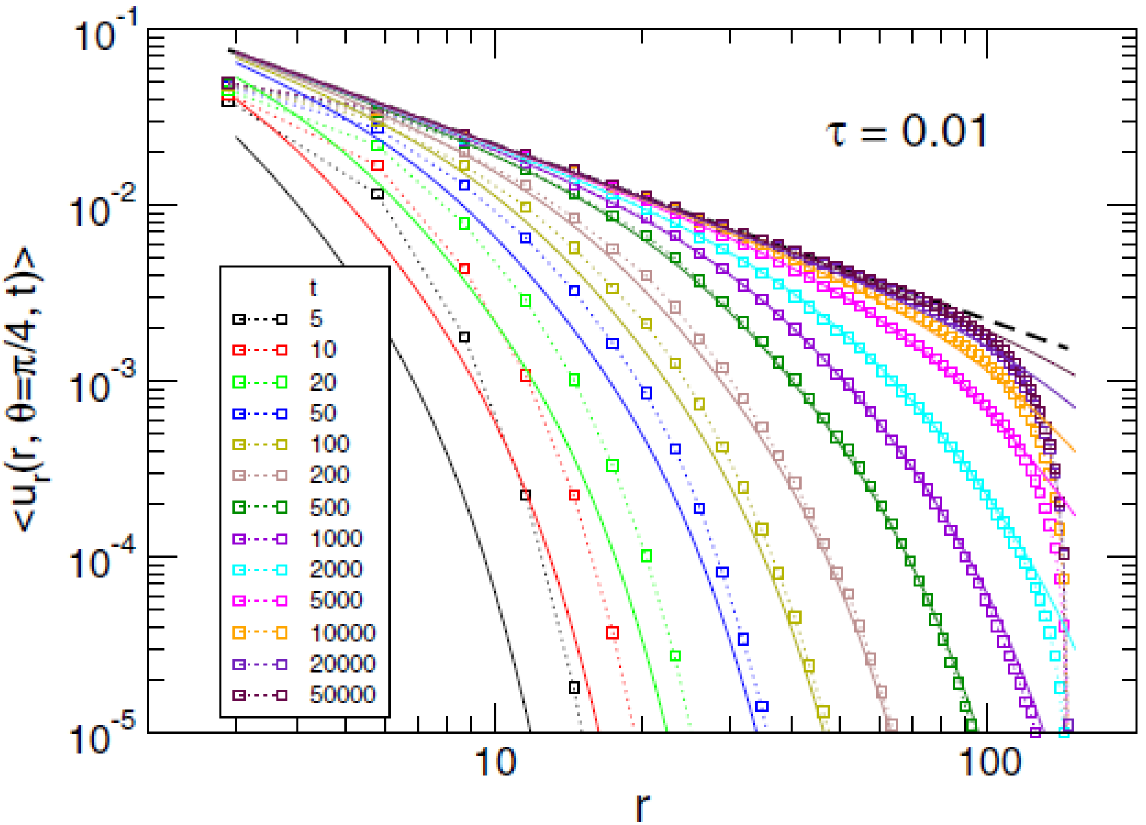




$$\mathbf{u}(\mathbf{r}) = \frac{\epsilon^*}{4(1-\nu)}\left(\frac{a}{r}\right)^2 \left\{\left[2(1-2\nu)+\left(\frac{a}{r}\right)^2\right]\left[2\hat{\mathbf{n}}\,(\hat{\mathbf{n}}\cdot\mathbf{r})-\mathbf{r}\right]+2\left[1-\left(\frac{a}{r}\right)^2\right]\left[\frac{2\,(\hat{\mathbf{n}}\cdot\mathbf{r})^2}{r^2}-1\right]\mathbf{r}\right\}$$

Long time response averages out to continuum solution despite large fluctuations

Time evolution of the displacement field



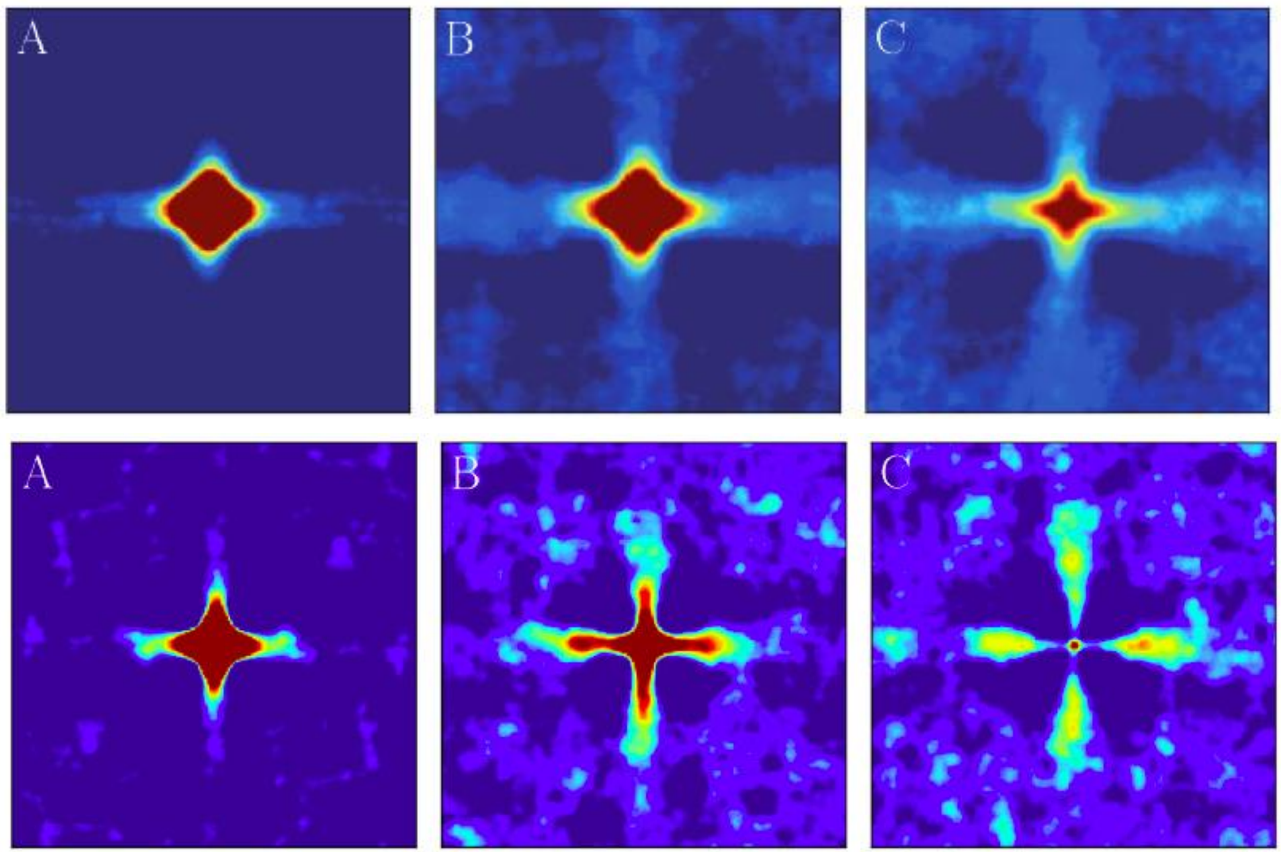
Full time dependent response (transients) agrees well with exact solution of diffusion equation for displacement in elastic medium.

F. Puosi, JR, J-L. Barrat, PRE (2014)

Plastic correlations in driven athermal solid

"probability for plastic event to occur at point r+Δr if plastic event was active at position r some time Δt ago"

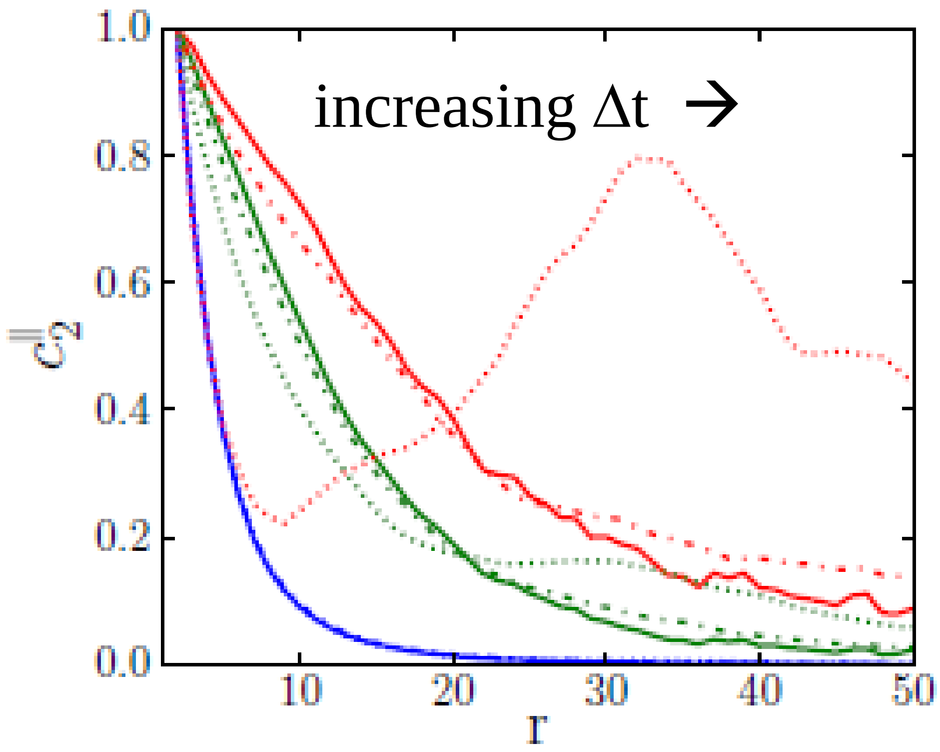
$$\left\langle \left[d^2_{min}(r,t) - \left\langle d^2_{min}(r,t) \right\rangle \right] \cdot \left[d^2_{min}(r+\Delta r, t+\Delta t) - \left\langle d^2_{min}(r, t+\Delta t) \right\rangle \right] \right\rangle$$



increasing strain rate ↓

increasing Δt →

Decay of plastic correlations along flow direction



A. Nicolas, JR, J-L. Barrat, EPJE (2014)

- 3 different strain rates
- Approximately exponential decay (can depend on dissipation scheme (Varnik et al. PRE (2014))

- Gradual buildup of correlations as elastic signal propagates through material
- Correlations spread approx with sound velocity

→ Construction of mesoscopic models: talk by J.-L. Barrat (Wednesday)

Conclusions

1. Flow defects (carriers of plasticity) are most efficient scatterers of sound waves in solids (dislocations, grain boundaries, glass)
2. In glasses, thermal and plastic rearrangements are several times more likely at soft spots (or high softness). Particles move preferentially along soft directions.

   → Linear (harmonic) theory predicts (a large part of) nonlinear response!

3. Correlations survive at finite strain rate and temperatures
4. Soft spot maps are long lived structural features, fully decorrelate only at yield strain or after α-time
5. Correlations robust in 3D glasses (eg. polymers)

Acknowledgements

- **Soft spots in amorphous materials:**
  Anton Smessaert,
  University of British Columbia

  Prof. Andrea J. Liu,
  Sam S. Schoenholz,
  University of Pennsylvania

- **Elastoplastic correlations and mesoscopic models:**
  Prof. Jean-Louis Barrat,
  Dr. Alexandre Nicolas,
  Dr. Francesco Puosi,
  Université Joseph Fourier, Grenoble, France