# Lengthscales in amorphous materials

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# Coworkers

- On-going work with:

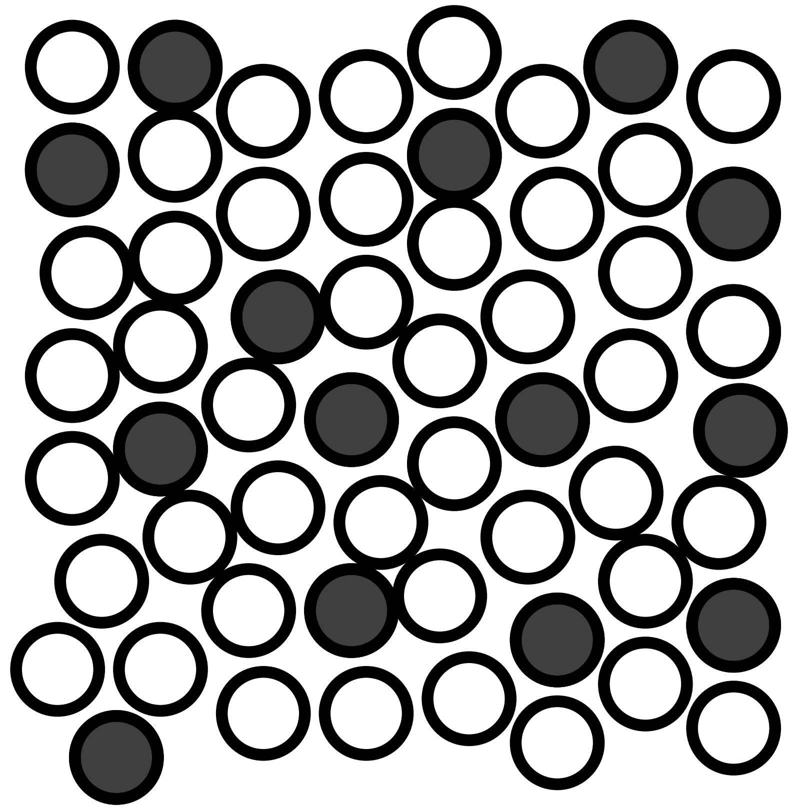
W. Kob (Montpellier)

- Related work with:

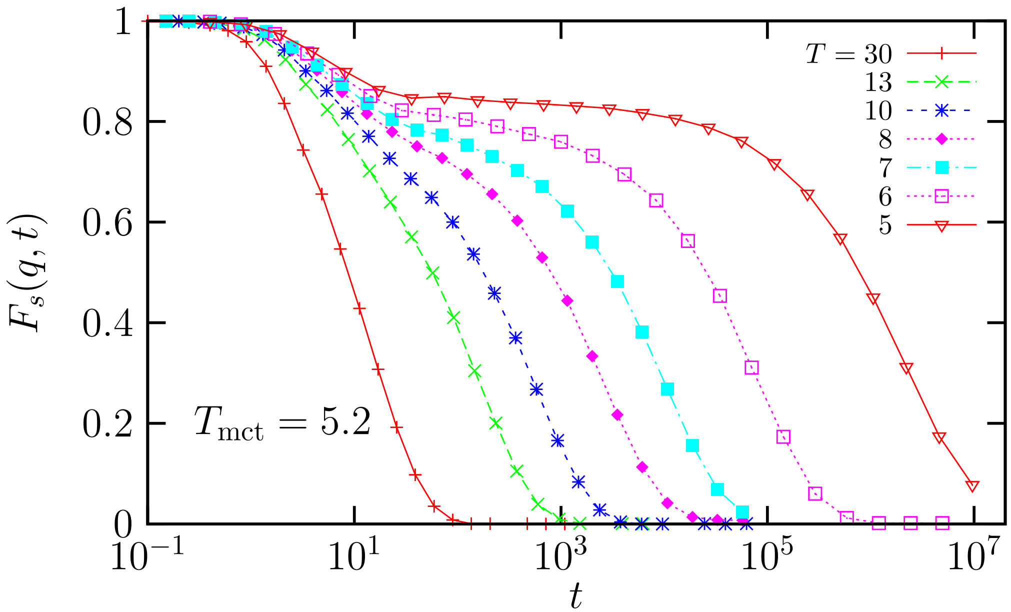
S. Roldan-Vargas (Granada)

- Discussions with:

C. Cammarota (Saclay)
G. Biroli (Saclay)



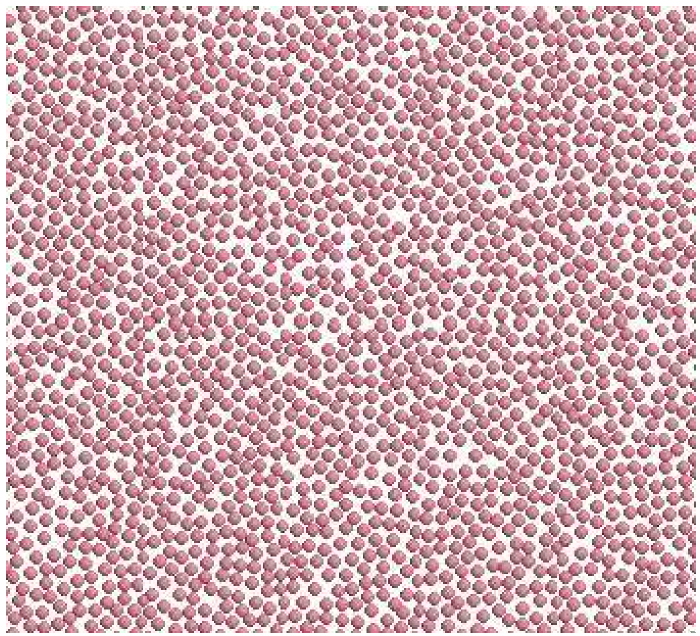
# Good old glass problem




- Glass transition: fluid - amorphous solid transition at low $T$.

- Rapid increase of viscosity, or relaxation time.

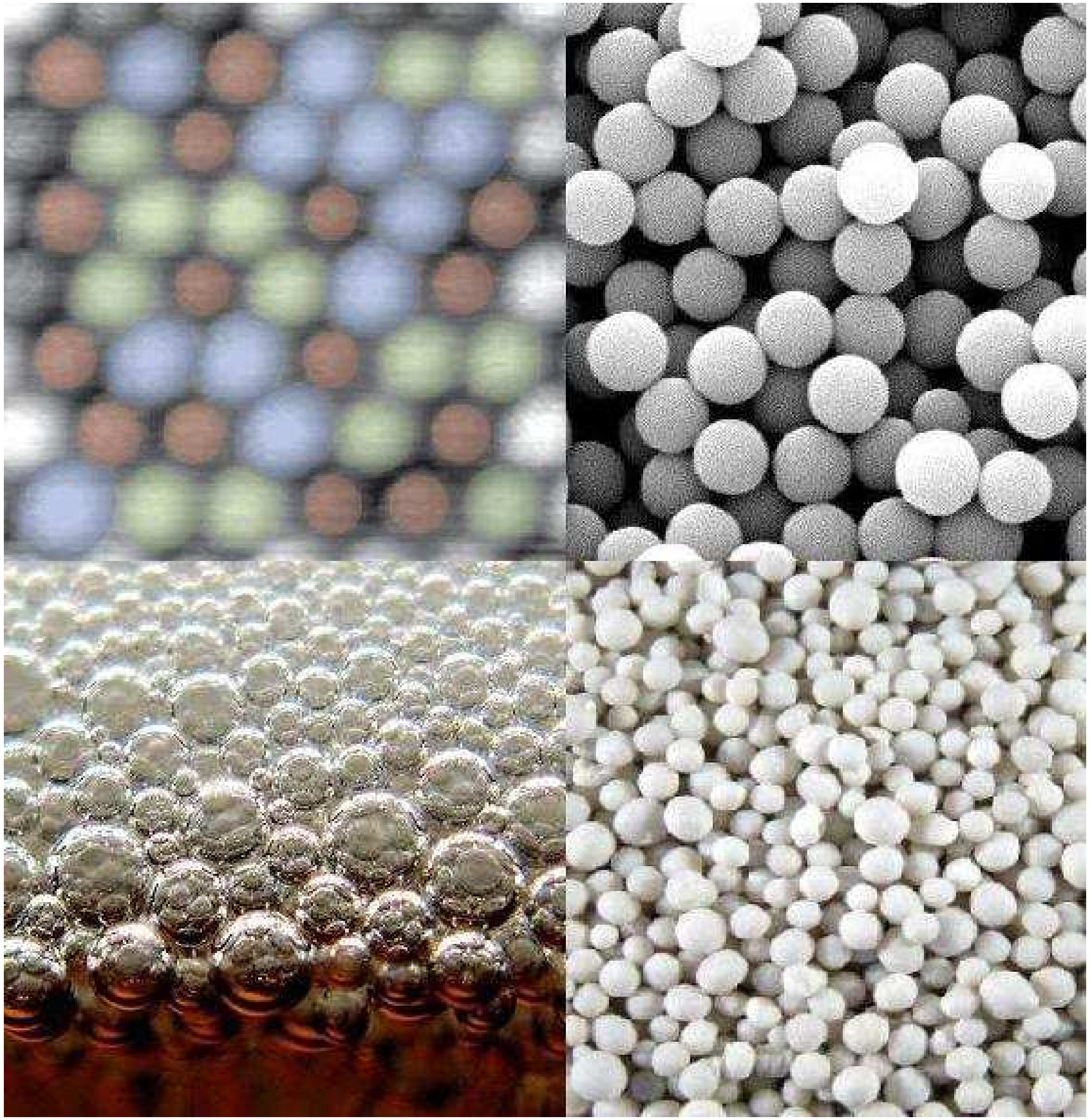
- Glass $=$ liquid "too viscous" to flow–ill-defined.



- Very little structural change at $g(r)$ level.

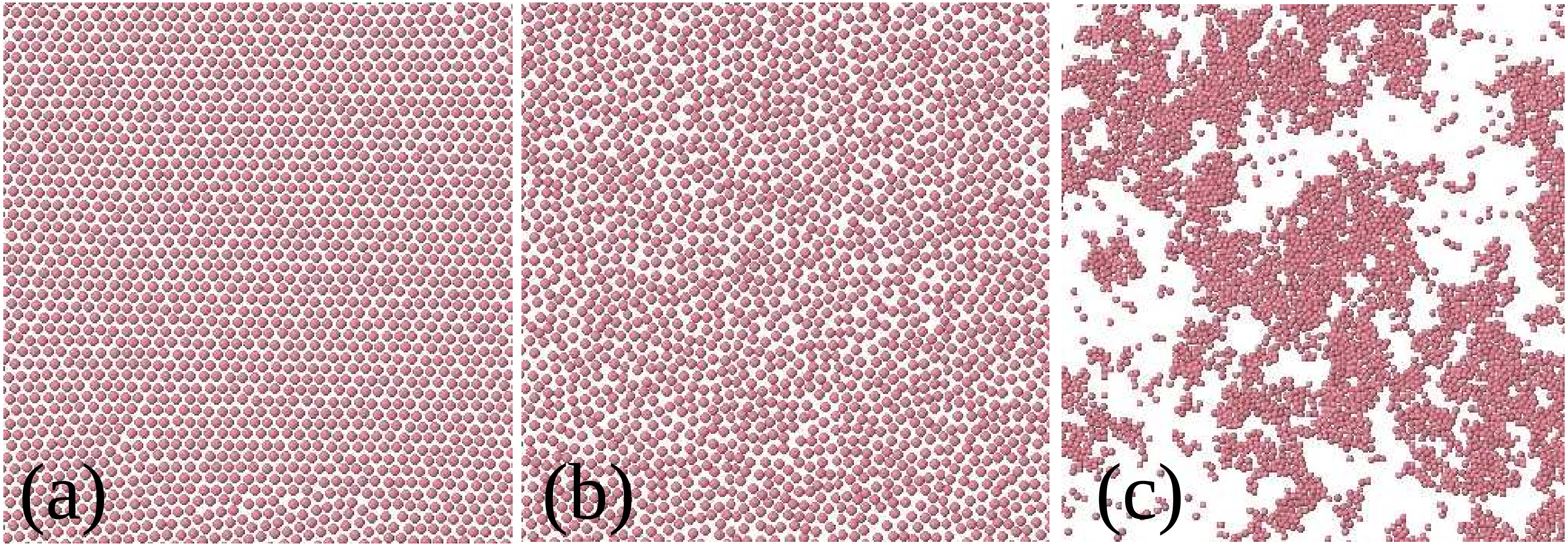
- Small cause big effects, or some new physics?

# Glasses—across the scales



[Berthier & Biroli, Rev. Mod. Phys. 2011]

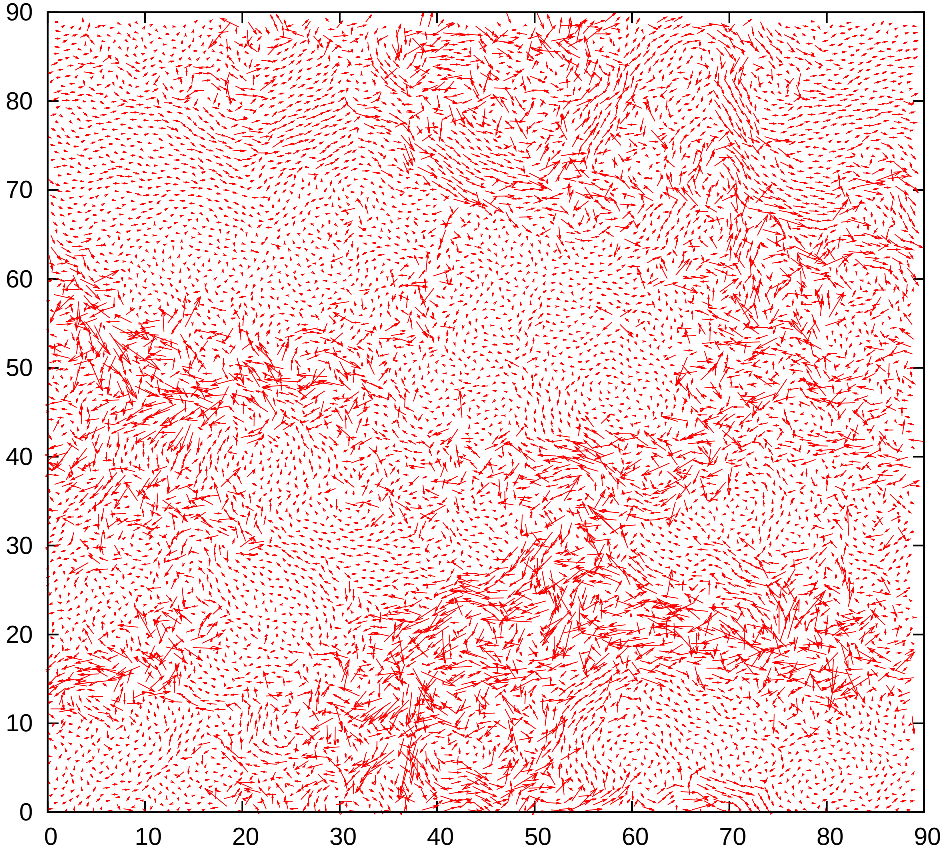
# Analogies don't work–or do they?

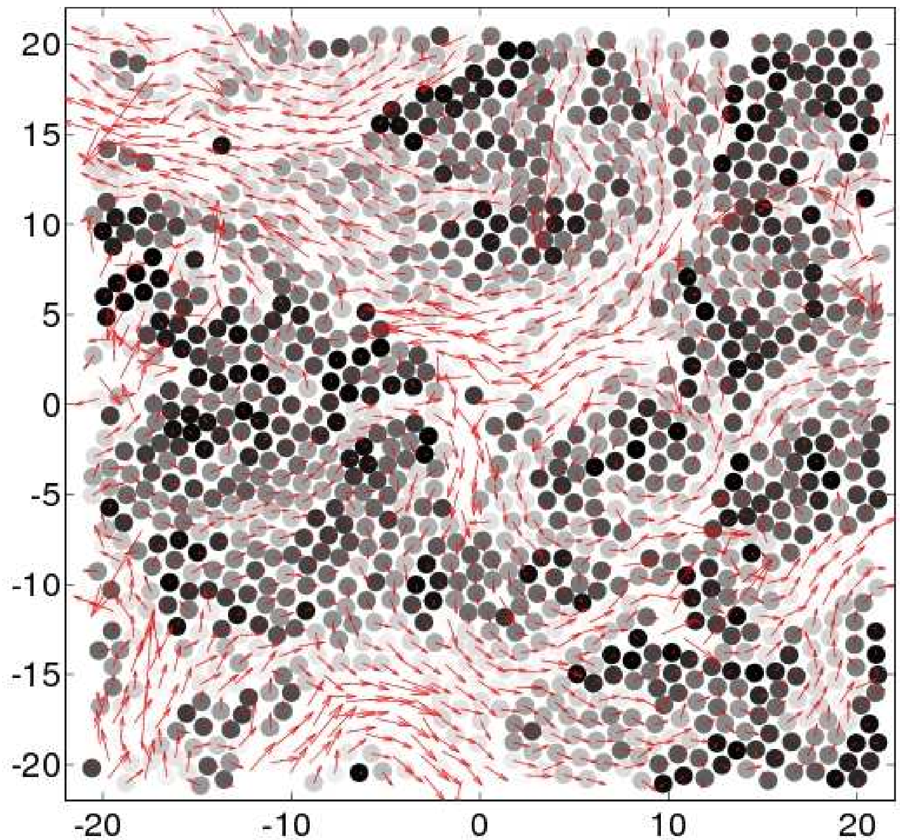



- (a) Crystals are solid because they are ordered: symmetry breaking.
- (c) Critical slowing if critical fluctuations: diverging length scale.
- (b) Why viscous liquids? "Hidden" criticality?
- (b) Why amorphous solids? "Hidden" symmetry breaking?

# Dynamic heterogeneity

- When density is large, particles must move in a correlated way. New transport mechanisms revealed over the last decade: fluctuations matter.



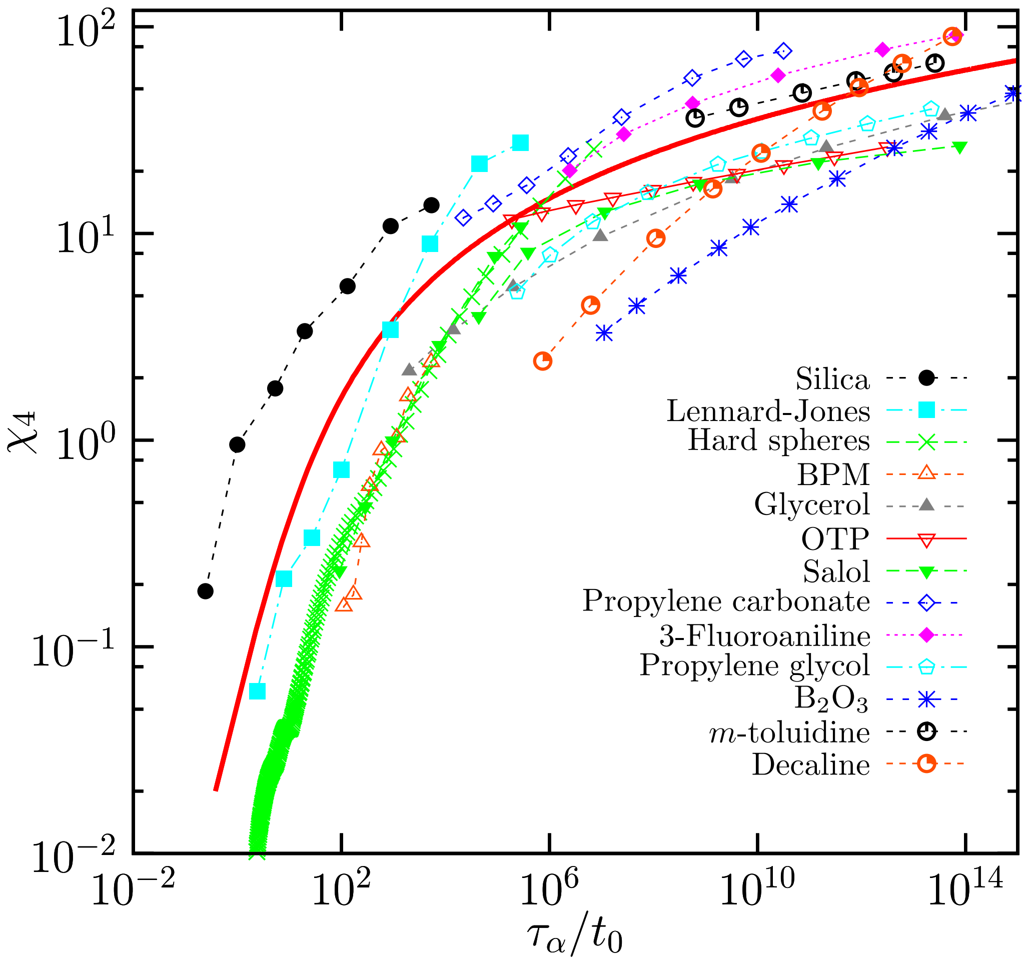




[Dauchot *et al.*, '05]

- Experimentally challenging in liquids—easier in soft matter & grains.

# “Dynamic criticality” in liquids

- Local dynamics is an “order parameter” with growing spatial fluctuations.




- Criticality revealed by multi-point dynamic susceptibilities.

- Theory suggests experimentally measurable fluctuations “$\chi_4$” to quantify typical correlation volume.

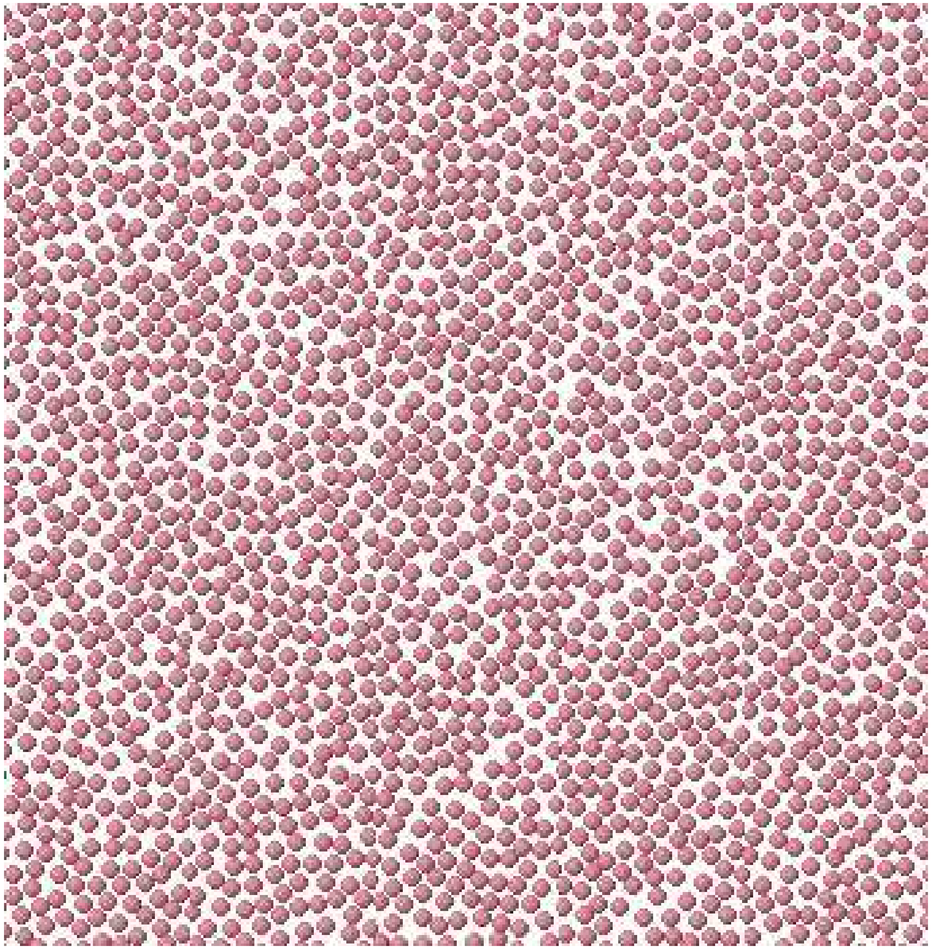
[Berthier *et al.*, Science ’05]

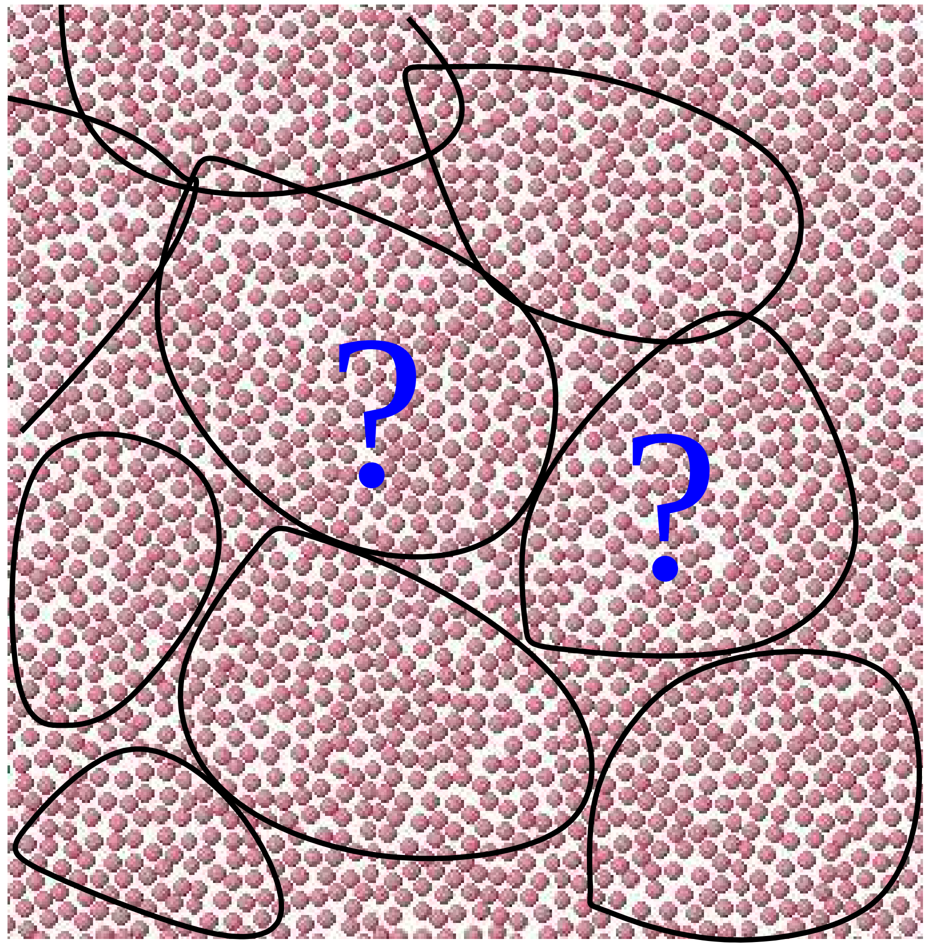
- Spatial fluctuations grow (modestly) near the glass transition.

*Dynamical heterogeneities in glasses, colloids and granular materials (Oxford, June 2011)*

Eds.: Berthier, Biroli, Bouchaud, Cipelletti, van Saarloos.

# Back to the local structure






- Are we missing something?
- E.g., specific geometric motifs? [Coslovich, Tarjus, Kurchan, etc.]

# Why confinement?

• Confinement is a way to probe static and dynamic lengthscales: perturb at $\mathbf{r}$, and measure effect at $\mathbf{r}+\mathbf{r}'$.

• Long history in supercooled liquids and polymers (interfaces, films, etc.)

$\rightarrow$ $n$-body static correlations: How does the position of $(n-1)$ particles influence the *position* of the $n^{\mathrm{th}}$ particle?

$\rightarrow$ $n$-body dynamic correlations: How does the position of $(n-1)$ particles influence the *dynamics* of the $n^{\mathrm{th}}$ particle?

• Confining the liquid "by itself". (1) Probe equilibrium correlations; (2) No "direct" effect on averaged fluid properties (e.g. no layering).

[Scheidler *et al.*, 90's]

# Point-to-set correlations

• Pin a 'set' of particles from equilibrium configuration at $t = 0$. Perform equilibrium average with pinning field at $t > 0$. [Montanari & Semerjian '08]

(1) How far does the system escape from $t = 0$ config?
Overlap $\langle Q \rangle = \langle q(t \to \infty) \rangle$ with $q(t) = \mathcal{N}^{-1} \sum_i n_i(t) n_i(0) \approx F(q,t)$.

Fluctuations of $q(t \to \infty)$: $P(Q)$, $\chi = N[\langle Q^2 \rangle - \langle Q \rangle^2]$.

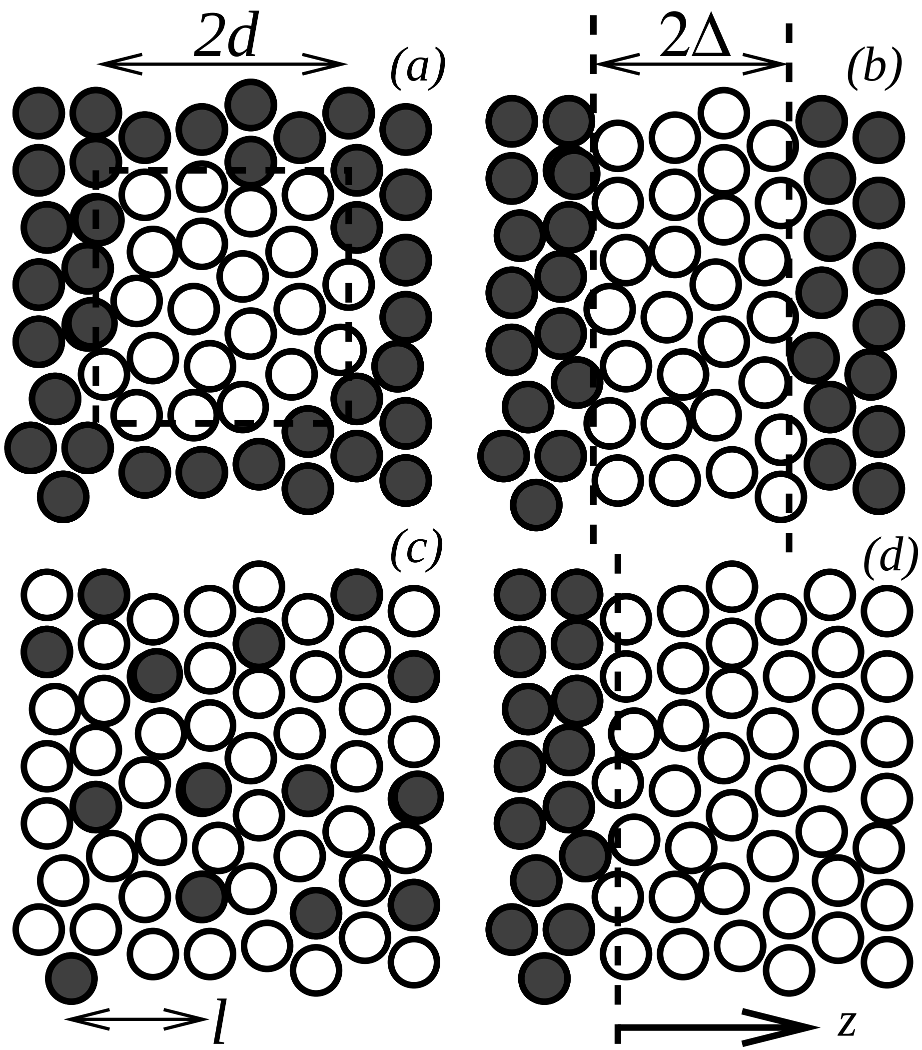
(2) How fast does the system escape? $Q_{\rm self}(t) \approx F_s(q,t)$.

• Project config. at $t \to \infty$ onto config. at $t = 0$. Natural thermodynamic analog of dynamic heterogeneity studies ($\chi_4$).

• Note: Pinning field prevents the system from fully escaping the $t = 0$ configuration. We are probing metastability.

# What geometry?

- Qualitatively different choices for ‘set’ of pinned particles at at $t = 0$.




(a) Cavity. Probe one point (center), system is finite. [Biroli *et al.*, ’08]

(b) Sandwich. Probe an infinite plane (middle), $d-1$ dimensions.

(c) Pinned. Probe all free particles, homogeneous, infinite, $d$-dimensional.

(d) Wall. Probe profile; bulk recovered for $z \to \infty$. [Kob *et al.*, ’11]

- Standard critical point: one length to rule them all.

# Confinement & RFOT mosaic length

- RFOT theory: glass transition controlled by existence and evolution of metastable states, $s_c(T)$. [Kirkpatrick, Thirumalai, Wolynes '89]

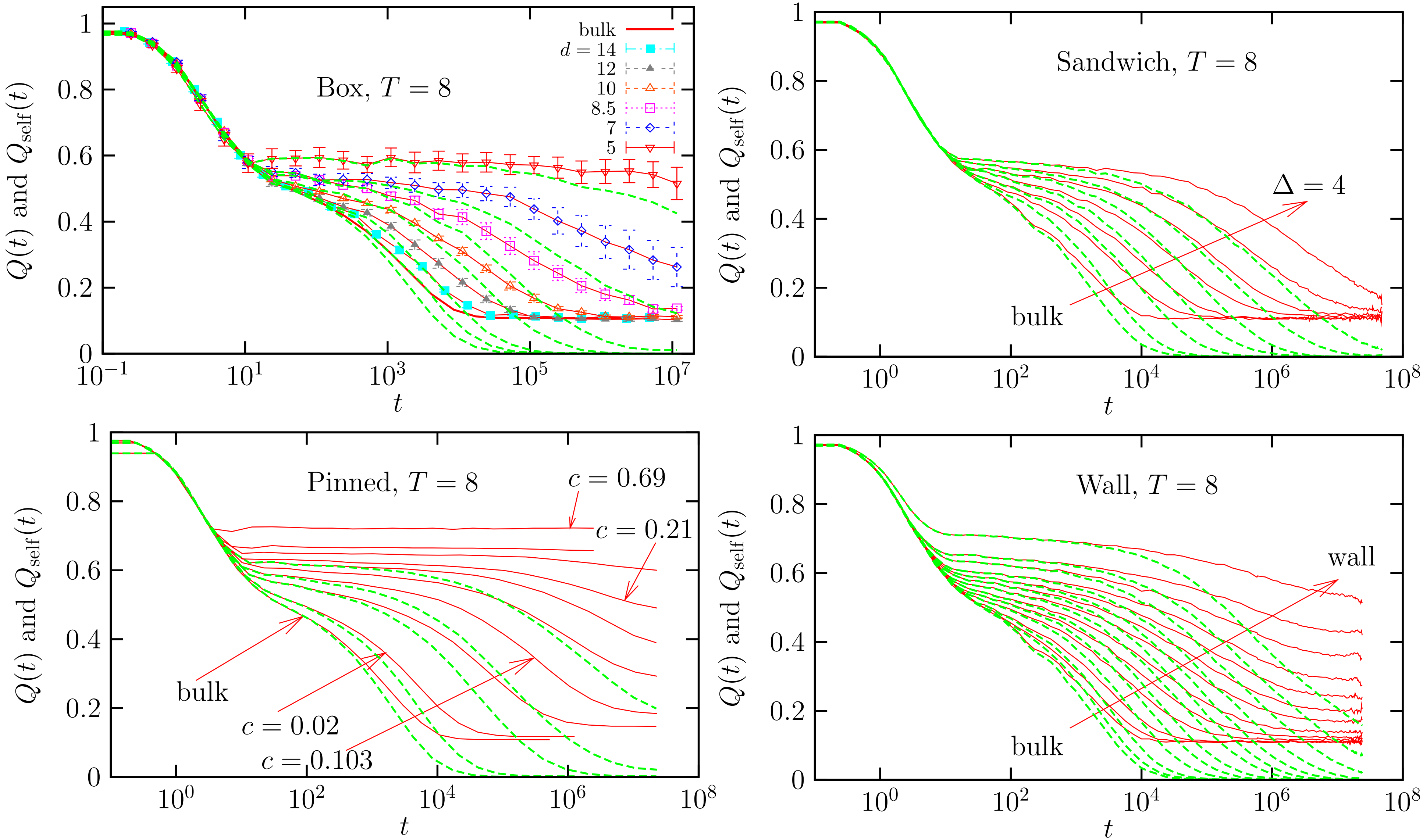
- The 'mosaic' length, $\ell \sim \sigma/(Ts_c)$, emerges as a competition between:
  -mismatch between states: $\sigma\ell^{d-1}$;
  -entropic gain due to state multiplicity: $Ts_c\ell^d$.

- Point-to-set correlation in closed cavity: crossover between low and high $\langle Q \rangle$ when $d \sim \ell$. [Bouchaud, Biroli JCP '04]

- Wall geometry: no surface/bulk competition, but thermal fluctuations near the wall of width controlled by $\ell$ (power law? log?)

- Sandwich and pinned geometries: The "crossover" at $\Delta \sim \ell$ and $c^{-1/d} \sim \ell$ concerns an infinite number of particles $\rightarrow$ ideal glass transition in confinement is predicted. [Cammarota, Biroli, '11]

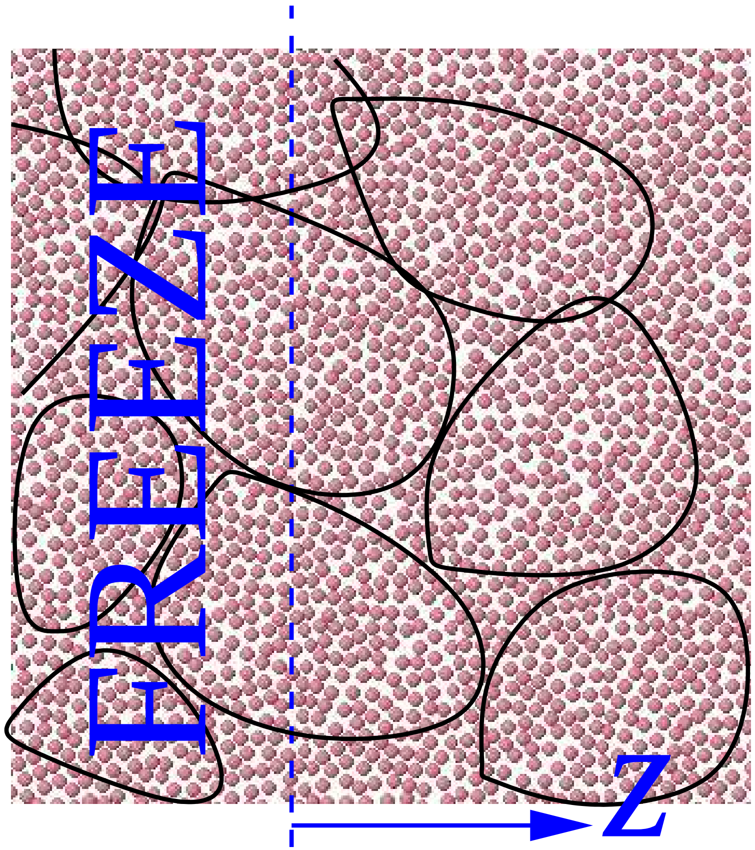
# Numerical comparison

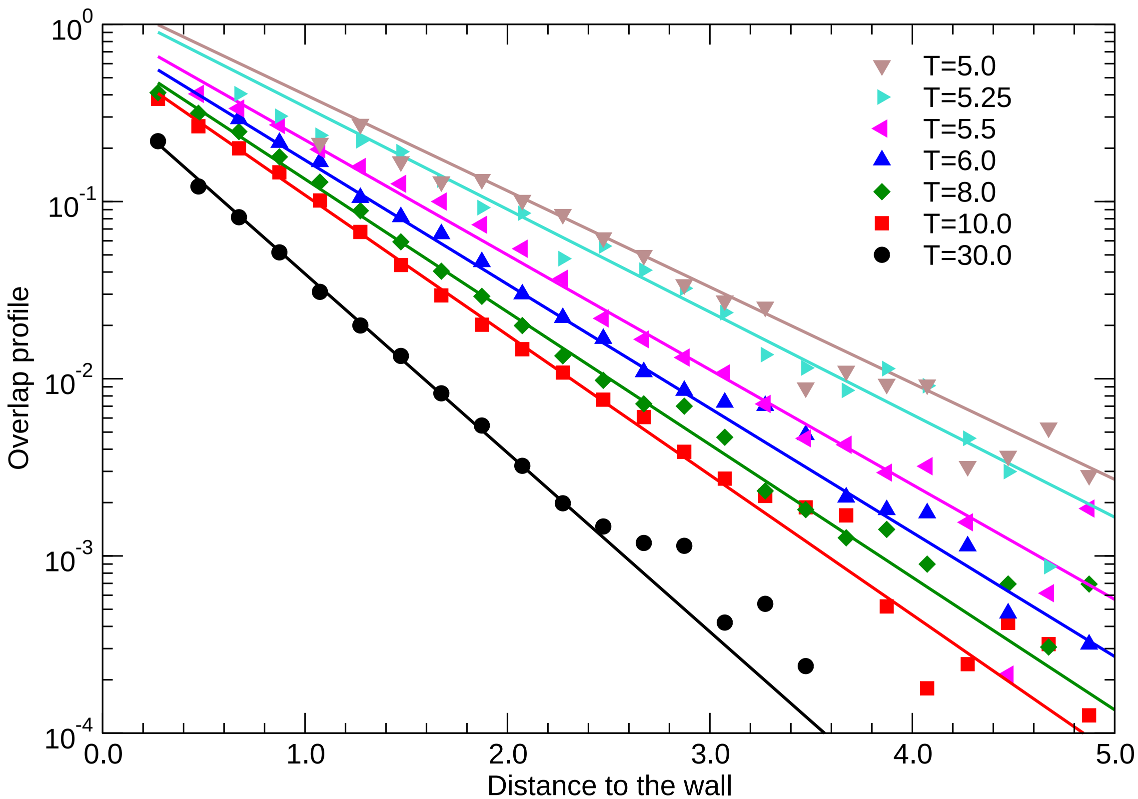



[Berthier, Kob, unpublished]

# The Wall

- We use the liquid as a template for itself. How far does “hidden order” propagate?



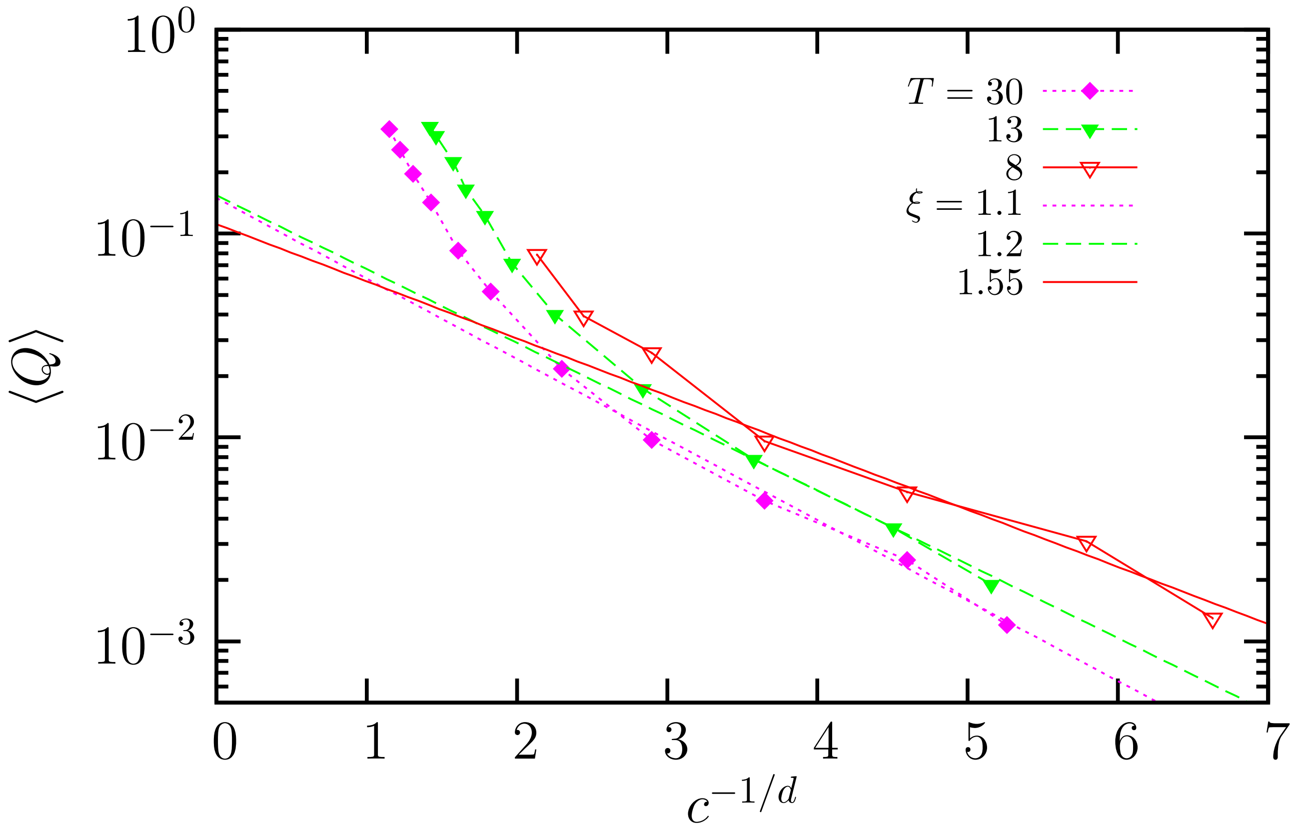




- “Amorphous order” propagates over larger length scales at lower $T$.

[Kob, Soldan-Vargas, Berthier, submitted ’11]

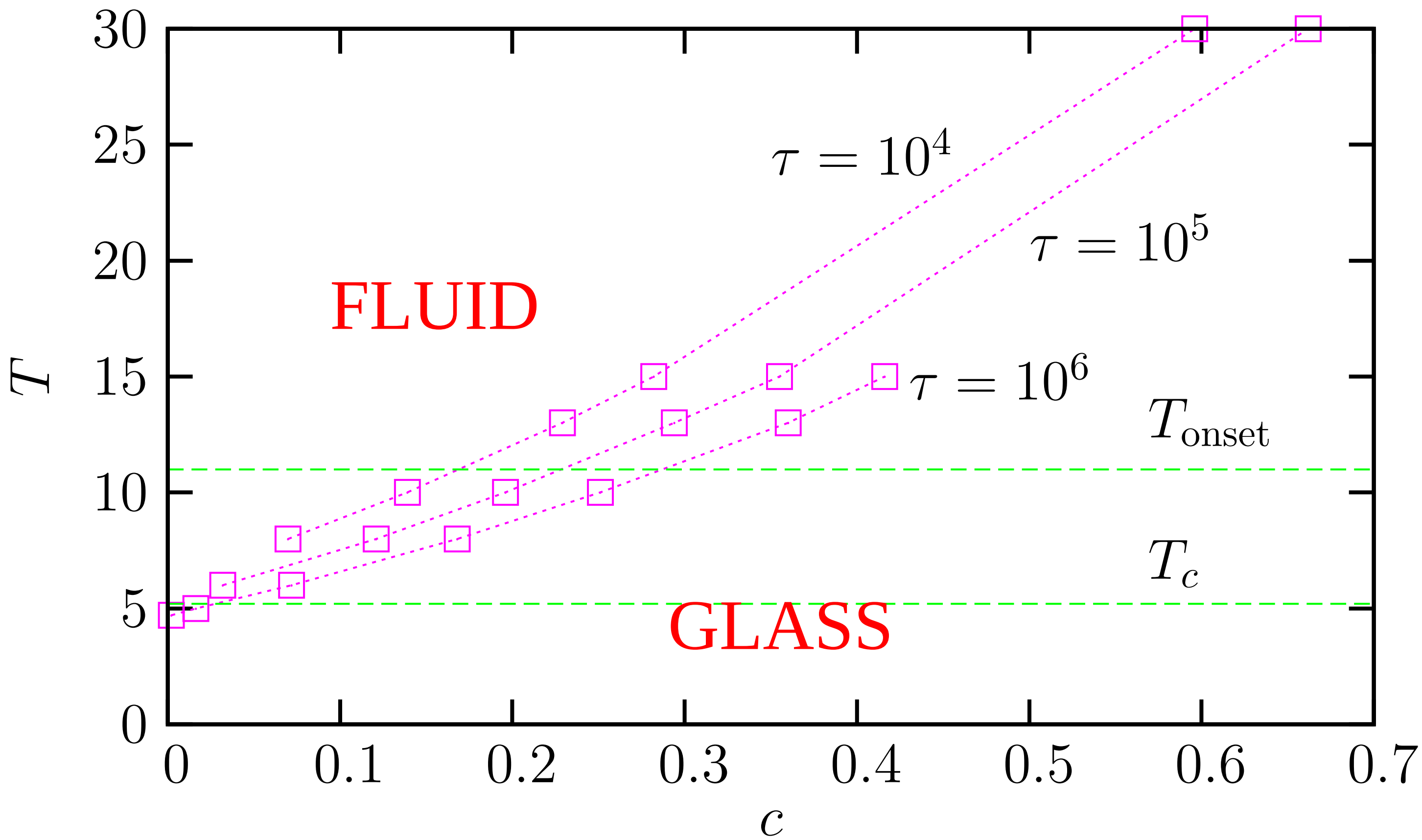
# Pinned particles: statics

• Temperature evolution: $\langle Q \rangle$ increases when $T$ decreases $\rightarrow$ less particles are needed to pin the initial configuration.




• Limited data again suggests slowly increasing static lengthscale. Consistent with wall (Kob *et al.*) and cavity (Biroli *et al.*) geometries.

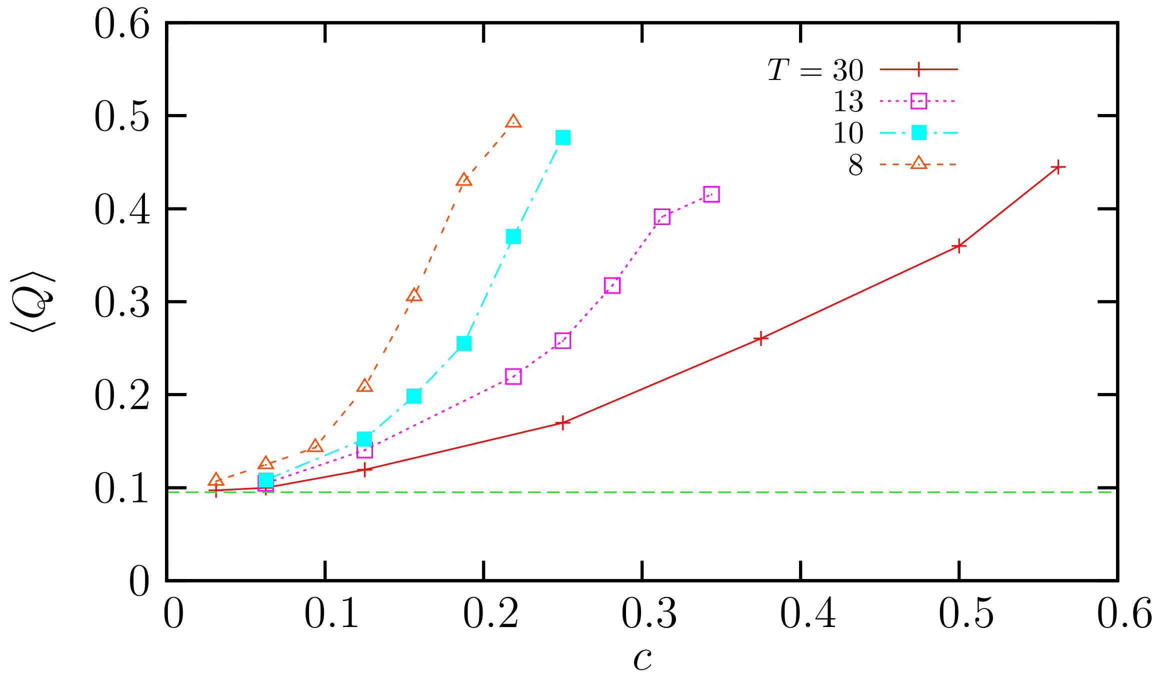
# Pinned particles: dynamics

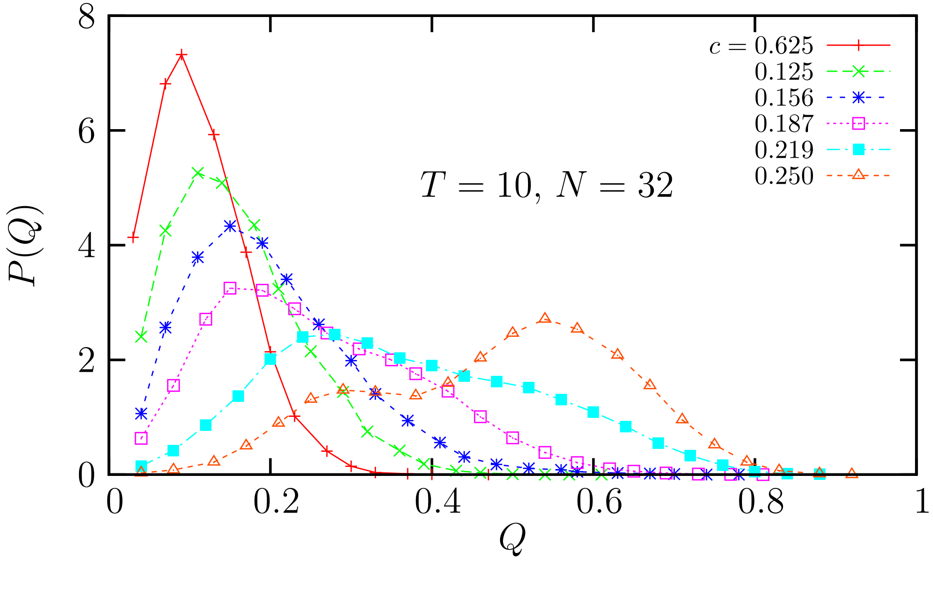


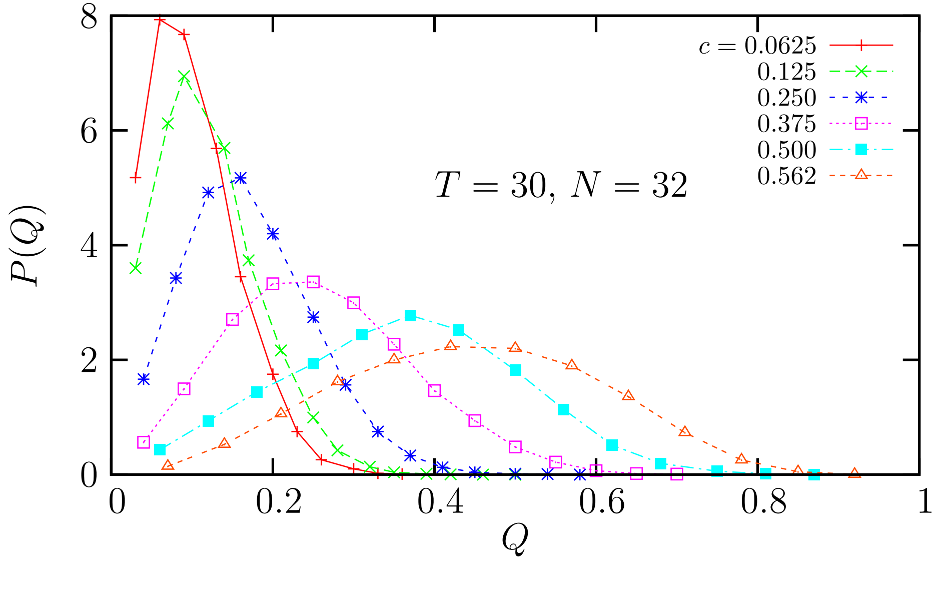

- It is easy to prepare equilibrium glass configurations.
- For some parameters ($N$, $T$, $c$), equilibrium can be maintained.

→ We believe we can study the ideal glass transition for the first time.

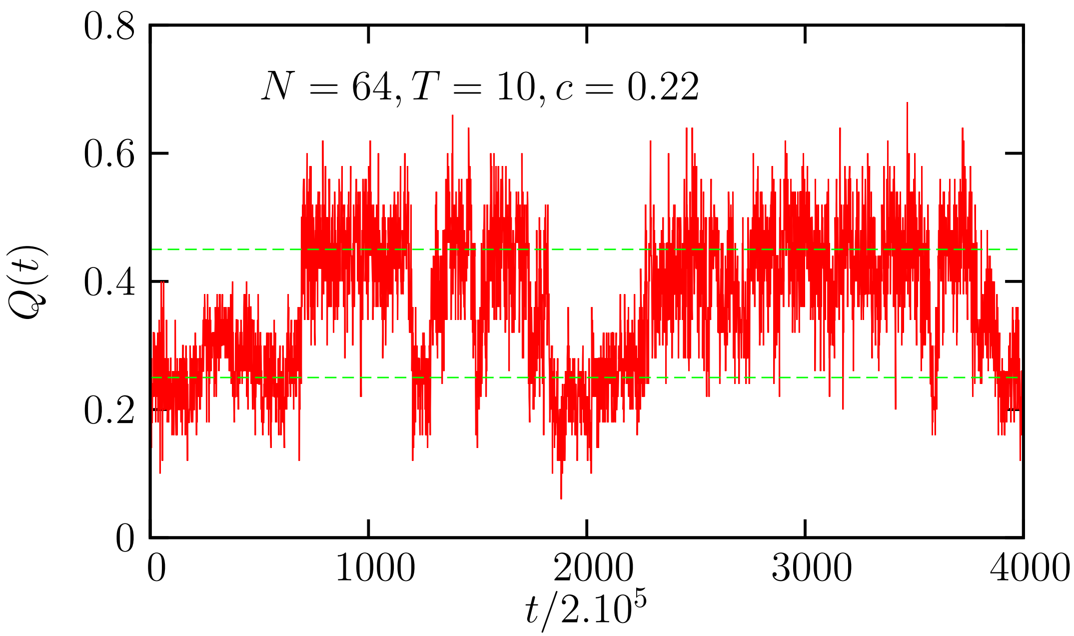
# First results for (small) systems

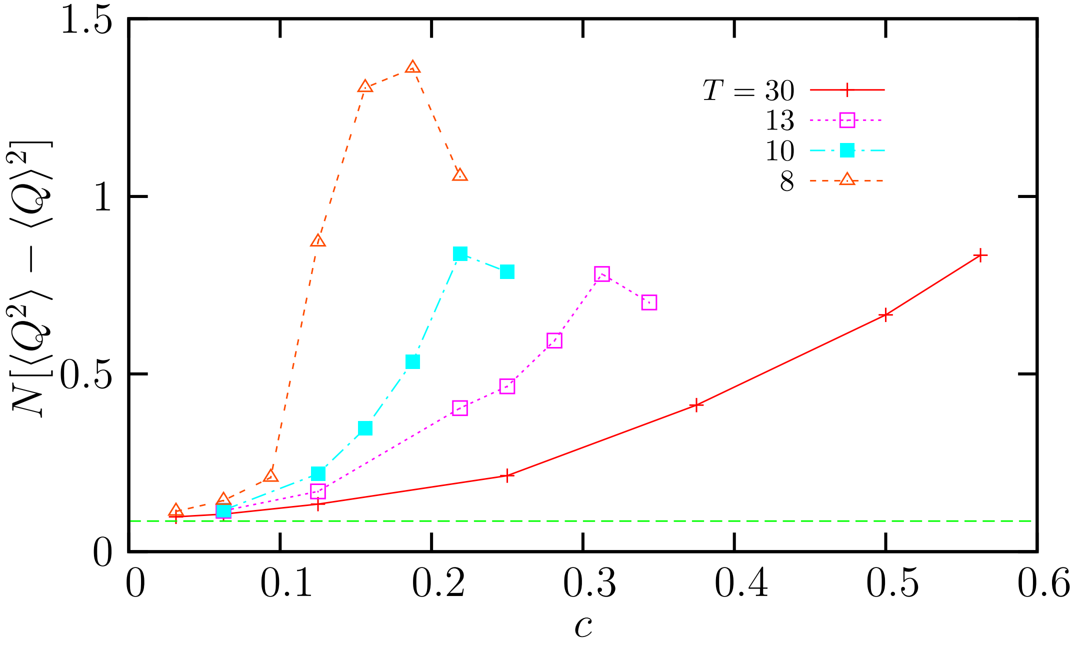


# Random first order transition?




- Average overlap becomes more abrupt at low $T$.

- $P(Q)$ becomes bimodal, time series reminiscent of first order transitions.

- The susceptibility $\chi$ develops a maximum.

These data suggest we might be able to establish the presence of an ideal glass transition in $(c, T)$ plane.
$\rightarrow$ Finite size scaling.

# Conclusions

- On-going effort to measure relevant thermodynamic spatial correlations in glass-forming liquids–beyond pair correlation level.

- Point-to-set correlations measured by confining the liquid by itself, various geometries.

- Pinned particles: seems an ideal geometry to measure static lengthscales and probe directly a microscopic mechanism of the glass transition.

- Will the random first order transition resist finite size scaling and larger $N$ studies?