Cooperativity, dynamic heterogeneity and the emergence of a growing length scale at the glass transition

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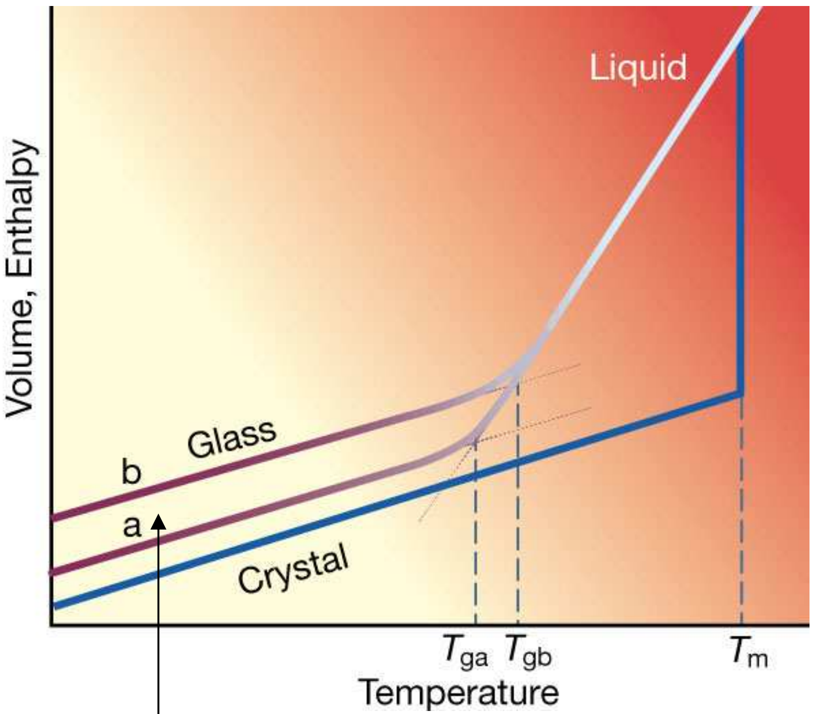
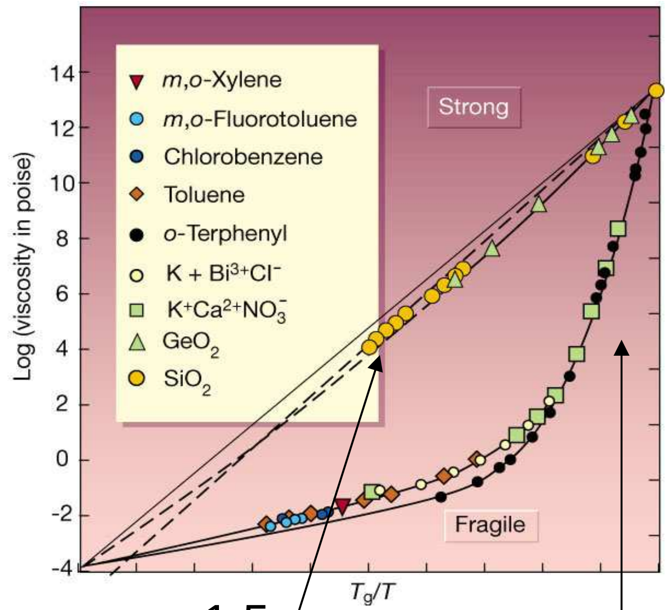
The Glass Transition





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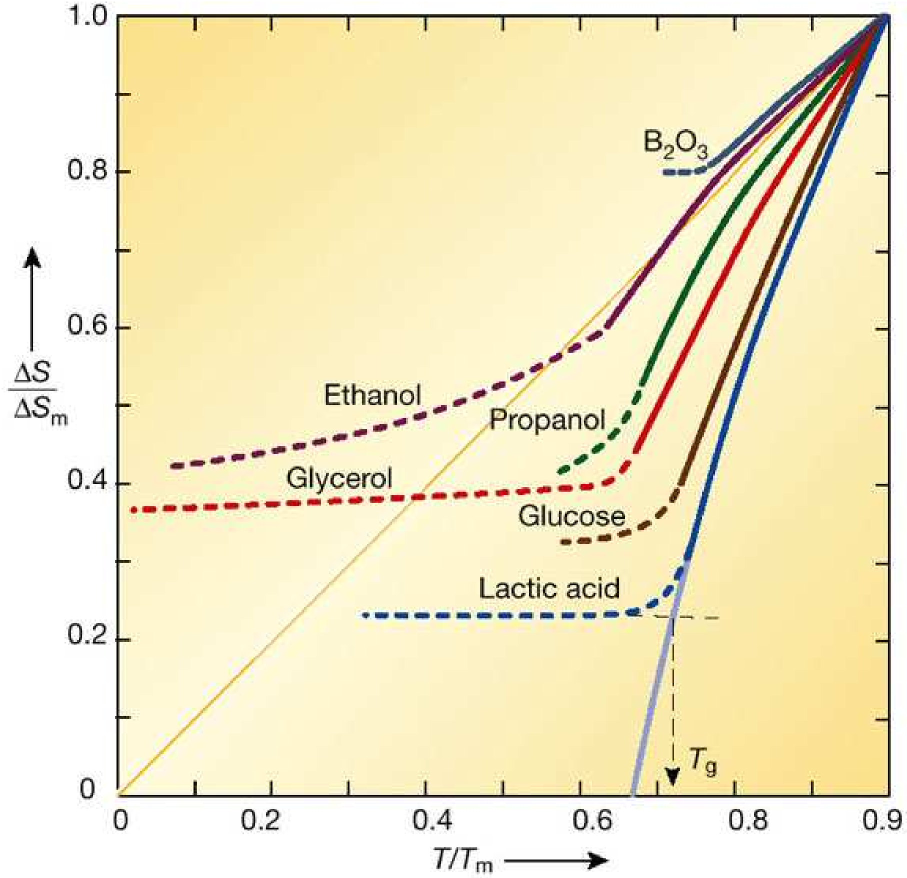
Amorphous rigid material : out of equilibrium system.

$$\tau \approx \exp\left(\frac{E_{act}}{K_B T}\right)$$

$$\tau \approx \exp\left(D\frac{T_0}{T-T_0}\right)$$

Configurational entropy: thermodynamic signature of a phase transition?

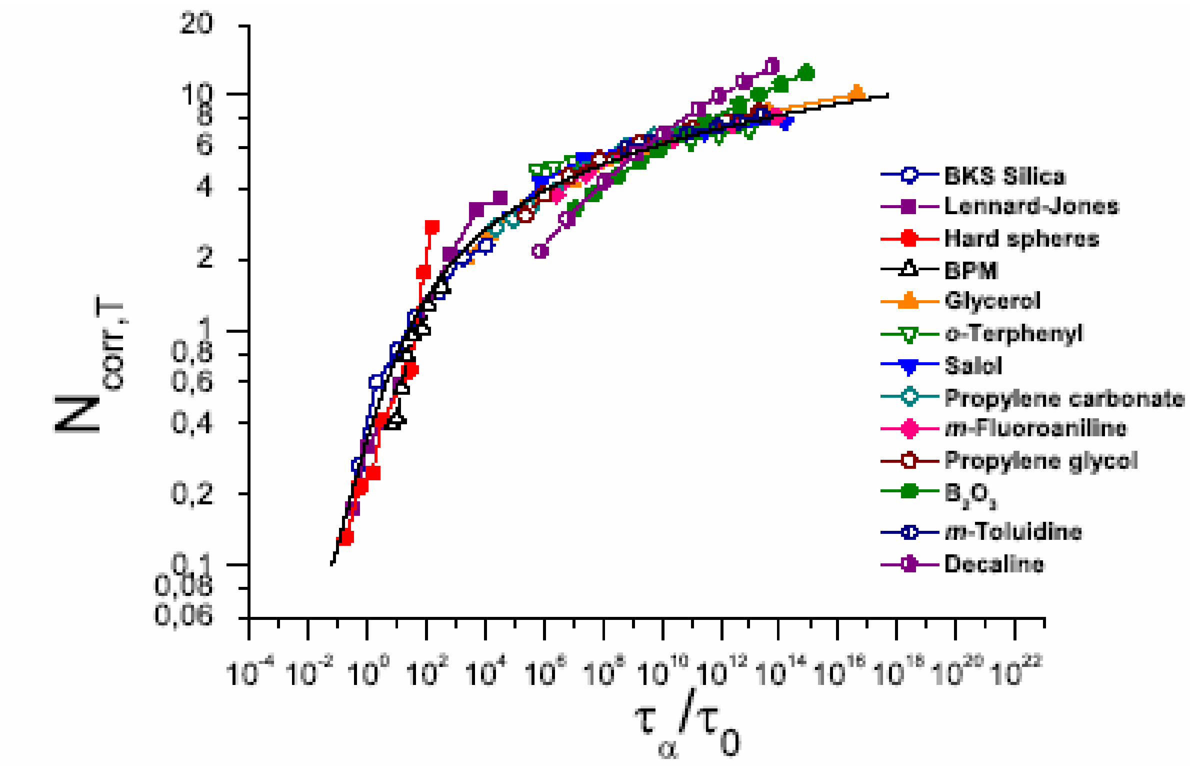


The temperature at which the extrapolated excess of entropy vanishes is very close to the temperature at which the extrapolated viscosity diverges with a Vogel-Fulcher law

Phase transition?

- Pro: extremely fast rise of the relaxation time
- Pro: entropy « vanishing » at To
- Pro: universal behavior independent of microscopic details
- Cons→Pro: growing correlation length?

Growing dynamical correlations approaching the glass transition



Number of correlated particles: one → hundreds

condmat 2007, Science 2005

Growing dynamical correlations approaching the glass transition II

- Detailed study of growing dynamical correlations in numerical simulations
- Critical power law behavior observed in simulations
- Experiments suggest a cross-over to activated dynamics

Dynamic growing correlations are probed by a 4 point functions (akin to spin glass susceptibility)

7378 J. Chem. Phys., Vol. 119, No. 14, 8 October 2003

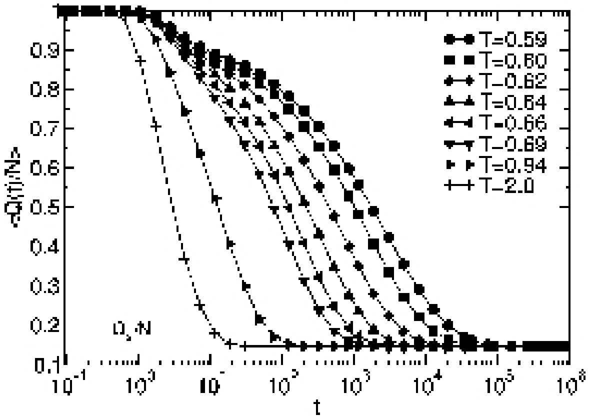
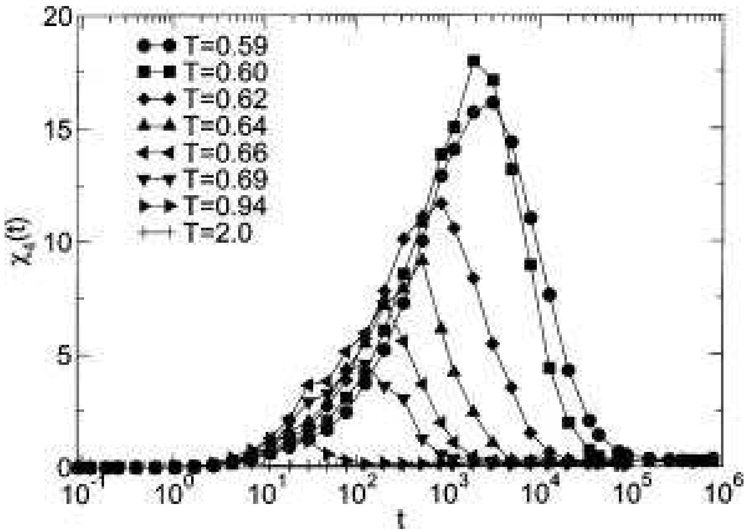


FIG. 3. Average time-dependent overlap "order parameter" $\langle Q(t)/N\rangle$ for each state point simulated. The solid line corresponds to the random value $Q_{ra}/N = 0.147$, as discussed in the previous section.

No growing length in any static correlation function or two point function

$$Q(t) = \int dr_1 dr_2 w(r_1 - r_2)\rho(r_1,0)\rho(r_2,t)$$

$$\chi_4 = N\left[< \left(\frac{Q(t)}{N}\right)^2 > - < \left(\frac{Q(t)}{N}\right) >^2 \right]$$



(for Binary Mixture Lennard-Jones)

Harrowell, Glotzer, Yamamoto-Onuki, Berthier,…

Dynamic field theory of dynamic correlations in glass-forming liquids

- Starting point: stochastic equations for slow degrees of freedom (in particular density), e.g.:

$$\partial_t \rho(x,t) = \nabla \cdot \left(\rho(x,t) \nabla \frac{\delta F}{\delta \rho(x,t)} \right) + \eta(x,t)$$

$$< \eta(x,t) \eta(x',t') >= 2T \rho(x,t) \nabla \cdot \nabla' \delta(x-x') \delta(t-t')$$

- Field Theory a la Martin-Siggia-Rose-DeDominicis-Janssen (classical limit of Keldysh formalism)

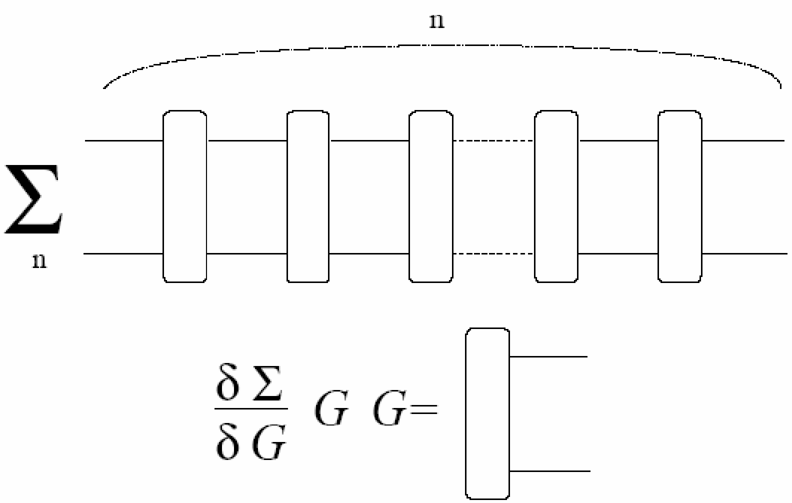
L. Berthier, GB, J.-P. Bouchaud, K. Miyazaki, D.R. Reichman JCP 2007

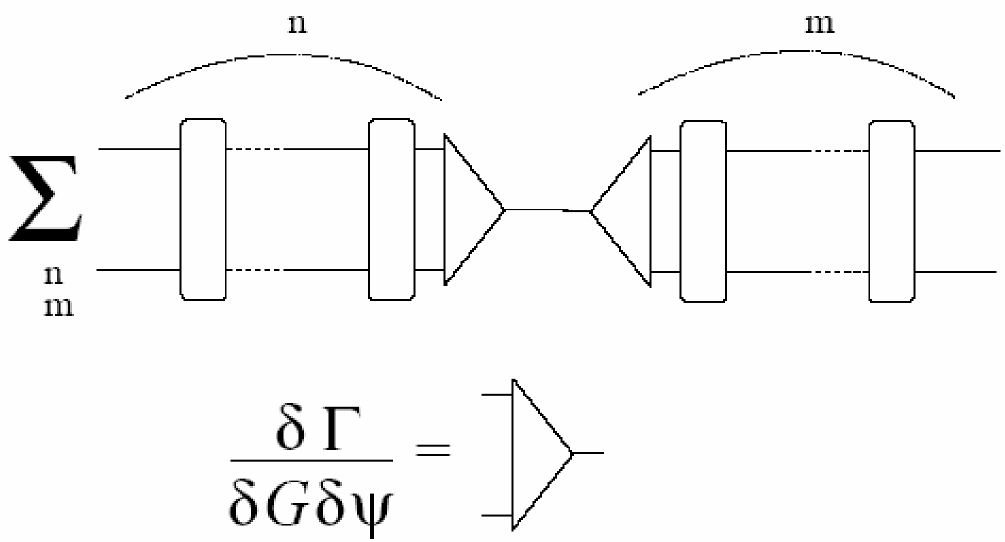
$$\langle \mathcal{A} \rangle = \int \mathcal{D}\rho \int \mathcal{D}\hat{\rho} \mathcal{A}[\rho] \; e^{S[\rho,\hat{\rho}]}, \tag{7}$$

with

$$\begin{aligned} S[\rho,\hat{\rho}] = \int d^3\mathbf{x} \int dt \, \Bigg\{ \hat{\rho}(\mathbf{x},t) \left[-\partial_t \rho(\mathbf{x},t) + \nabla \cdot \left(\rho(\mathbf{x},t) \nabla \frac{\delta \mathcal{F}[\rho]}{\delta \rho(\mathbf{x},t)} \right) \right] \\ + T \rho(\mathbf{x},t) (\nabla \hat{\rho}(\mathbf{x},t))^2 \Bigg\}, \end{aligned} \tag{8}$$

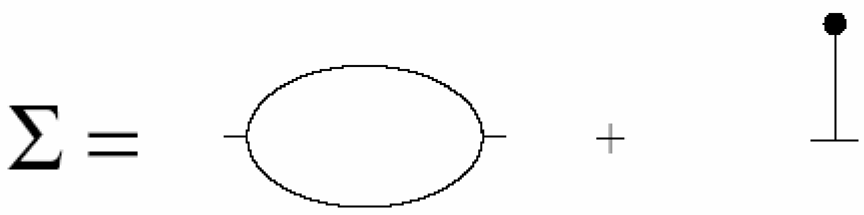
General expression for 4 point dynamical correlations: 2 contributions



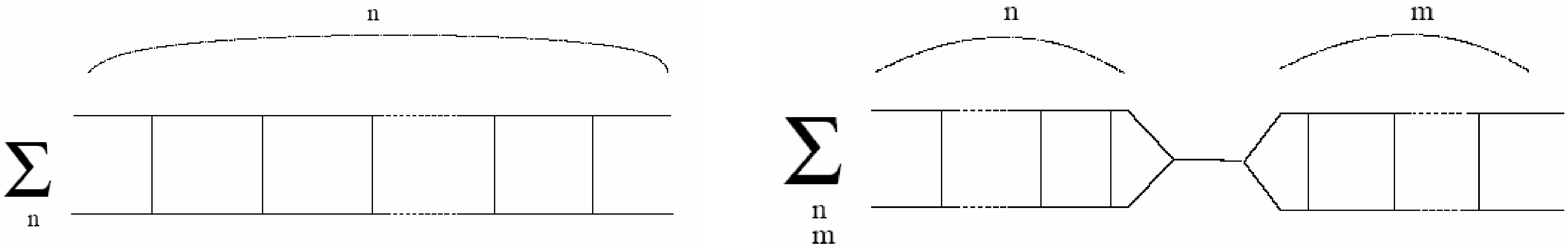


From this diagrammatic expression one can construct a theory of dynamical correlations and find experimentally accessible observables to probe dynamical correlations

One Loop Self Consistent Theory



- Self-consistent one loop equations on density-density correlation functions (Mode Coupling Theory of the Glass Transition)→dynamical glass transition



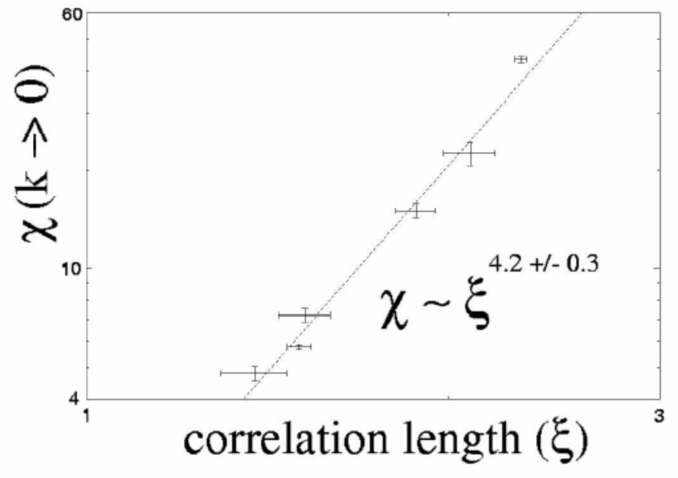
→ Divergent dynamical correlation at the transition

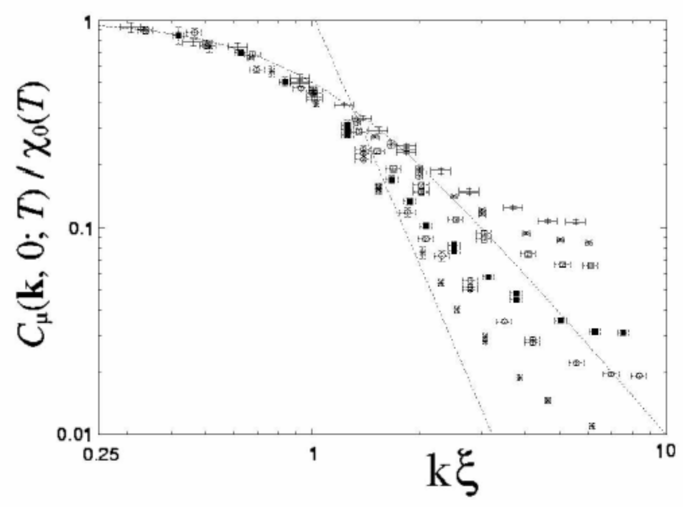
Results

- The sum of all ladders diverges as 1/(T-Td)
- There is a power law behavior in time with the exponent b.
- The peak takes place at $\tau_\alpha \propto (T - T_d)^{-\gamma}$
- There is a unique diverging correlation length $\xi \propto (T - T_d)^{-1/4}$
- $\eta = -2$

GB, J-P Bouchaud EPL 2004, GB, J-P Bouchaud, K. Miyazaki, D.R. Reichman PRL 2006

Simulations in BMLJ



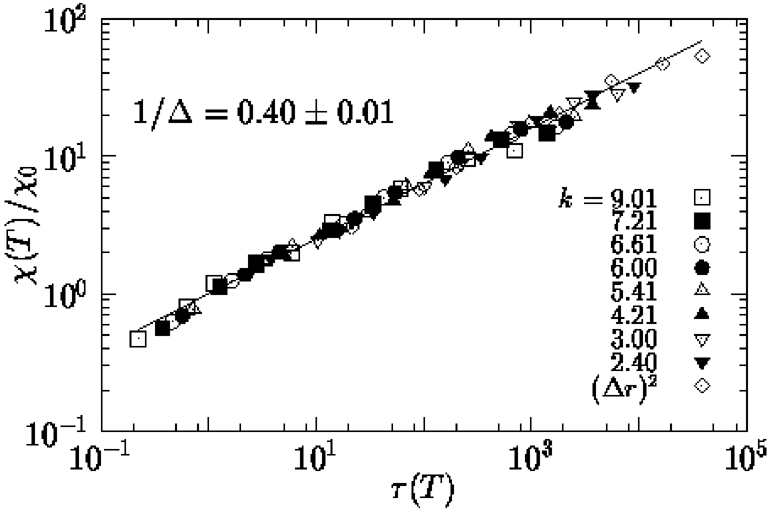


$$\nu_{MCT} = 1/4$$

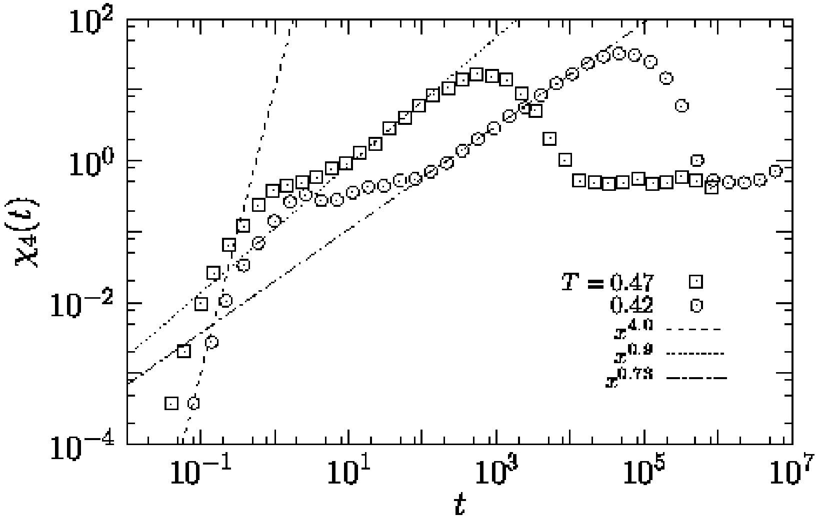
$$\eta_{MCT} = -2$$

Andersen & Stein

Simulations in BMLJ



$$\left(\frac{1}{\Delta}\right)_{MCT} = \frac{1}{\gamma} \cong 0.427$$



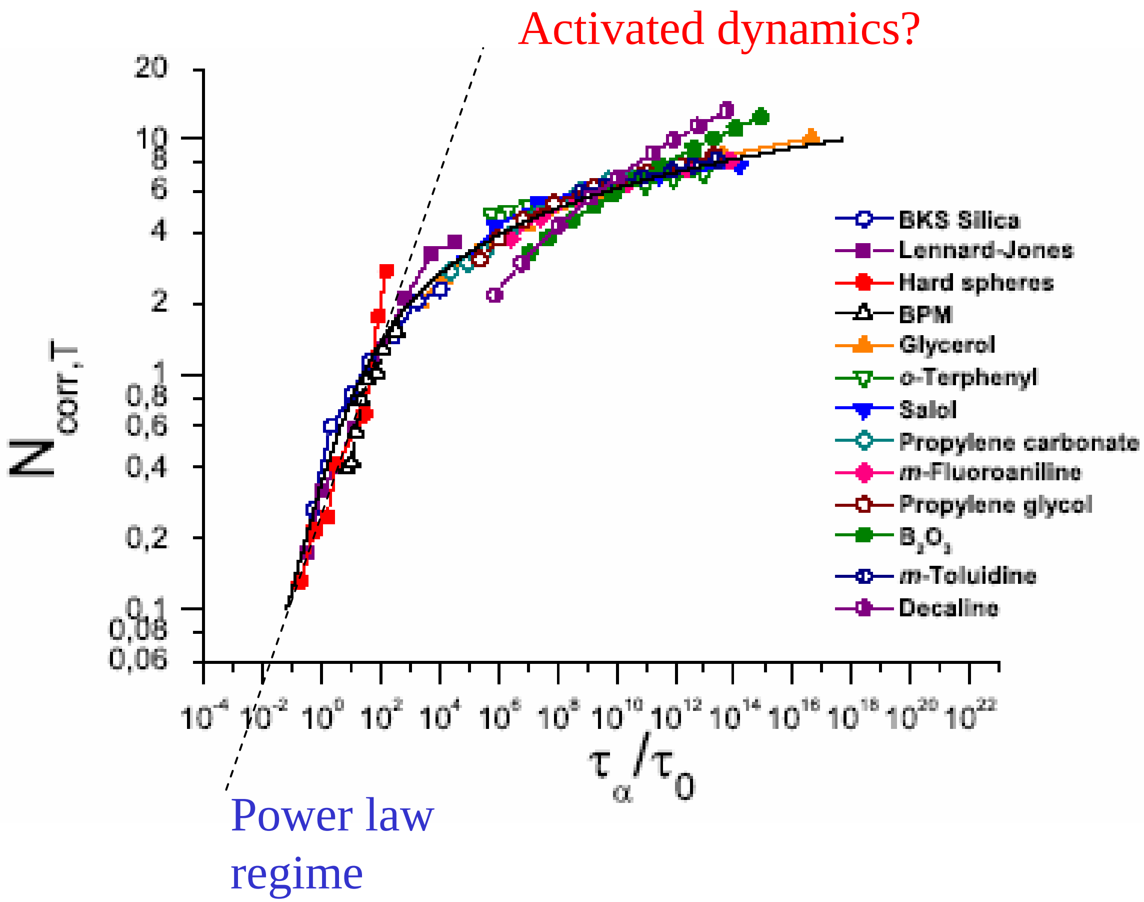
MCT: power law behavior in time before the peak with exponent $b \cong 0.63$

Berthier

Beyond one loop

- Results stable at any fixed self-consistent order above 8 dimensions.
- Upper critical dimension is 8 but….
- One has to determine the Ginzburg region…
- …and compare it to the region where non-perturbative processes transform the transition into a cross-over

A. Andreanov, GB, J-P Bouchaud

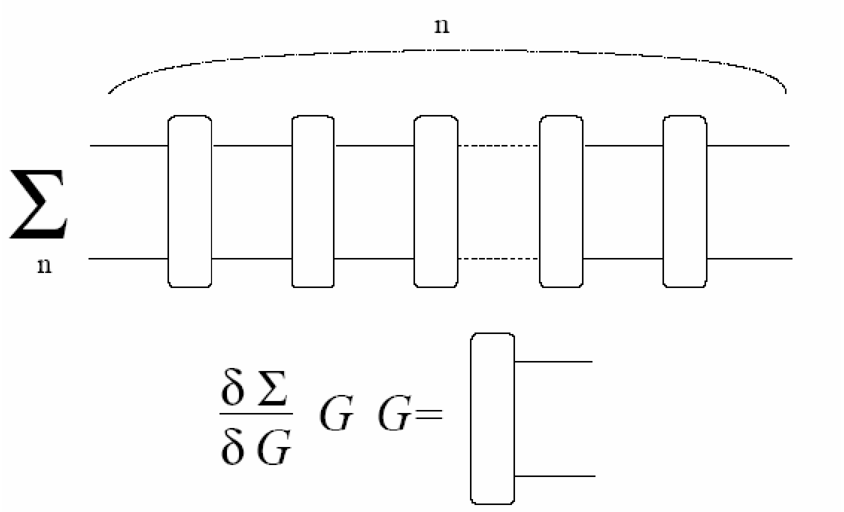


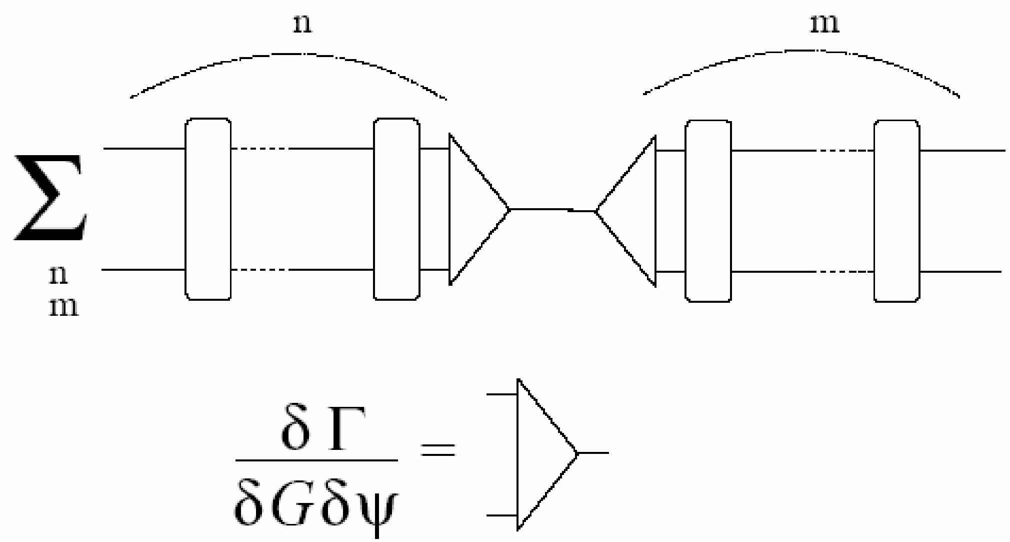
Close to the glass transition temperature → activated dynamics

$$(\xi^*)^{d-\theta} = \gamma / s_c(f) \propto \gamma/(T-Tc) \qquad \tau \propto \exp \Delta\beta(\xi^*)^{\psi} = \exp[K/(T-T_c)^{\psi/(d-\theta)}]$$

Kirkpatrick, Thirumalai, Wolynes; GB and J-P. Bouchaud

New theoretical tools to measure dynamic correlations





$$\chi_4(t)_{NPT} = \chi_4(t)_{NVE} + \left(\sqrt{\frac{k_B}{c_P}}T\,\frac{d < c(r,t) >}{dT}\right)^2 \geq \left(\sqrt{\frac{k_B}{c_P}}T\,\frac{d < c(r,t) >}{dT}\right)^2$$

$\left(\sqrt{\frac{k_B}{c_P}}T\,\frac{d < c(r,t) >}{dT}\right)$ can be measured directly in experiments→direct access to dynamical correlations

L. Berthier, GB, J-P Bouchaud, L. Cipelletti, D. El-Masri, D L'Hote, F. Ladieu, M. Pierno, Science 2005.

Conclusion & Perspectives

- Mounting evidence that the glass transition is a real phase transition.

- Mode Coupling Theory is successful to explain the growing of dynamic correlations for the first 6-7 decades of dynamical slowing down.

- Open problem (theoretical): have a firm theory of the dynamical slowing down close to the glass transition temperature. Activated dynamics?

- Open problem (experimental): measure directly the spatial correlations of the dynamics.

Example of diagram changing the critical exponent below eight dimension