

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

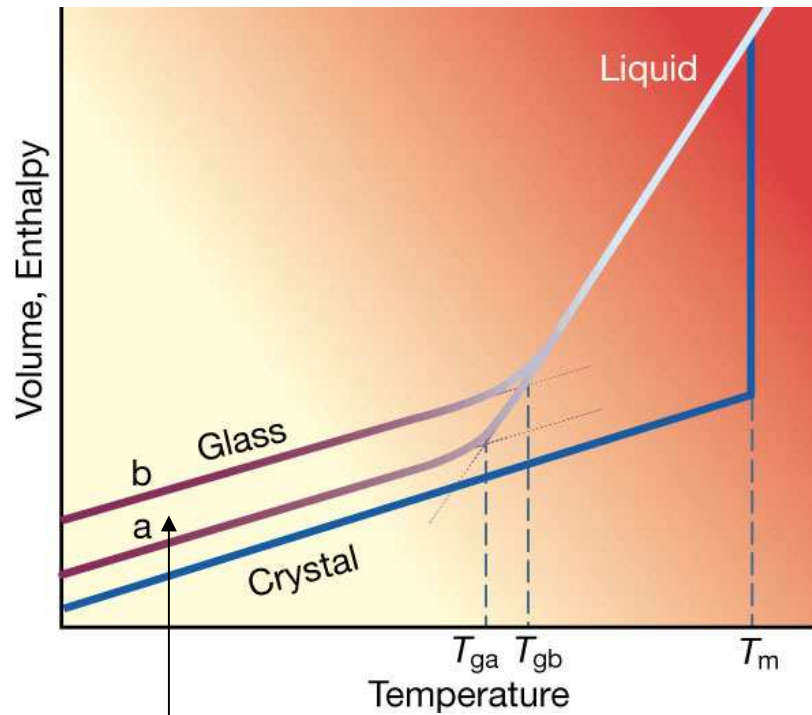
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SPhT CEA/Saclay

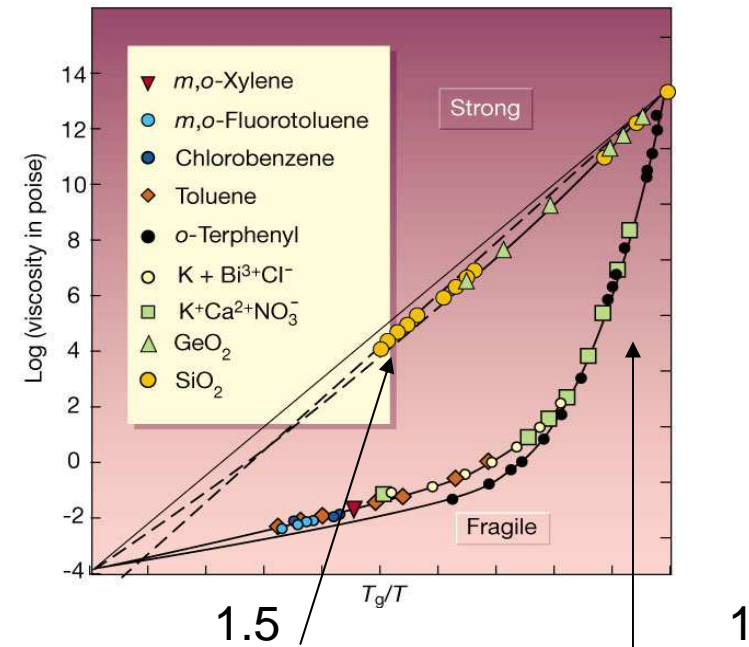
J.-P. Bouchaud

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Ladieu, A. Lefevre, K. Miyazaki, D. L'Hote, M. Pierno,
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The Glass Transition



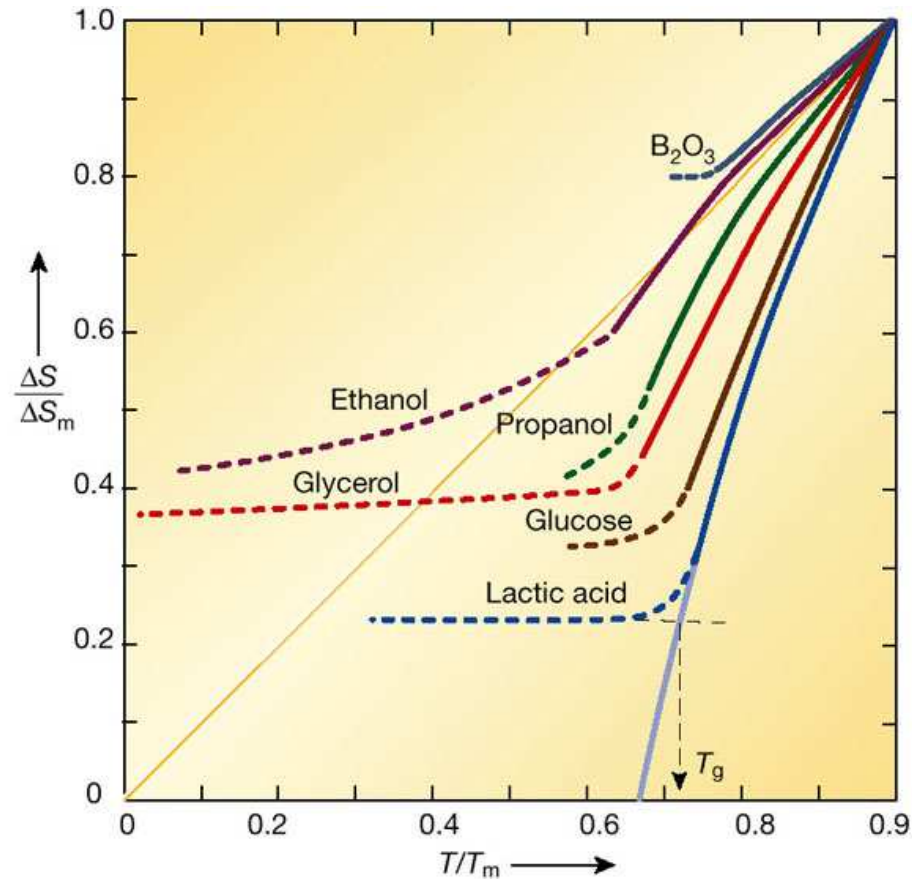
Amorphous rigid material : out of equilibrium system.



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

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

Configurational entropy: thermodynamic signature of a phase transition?

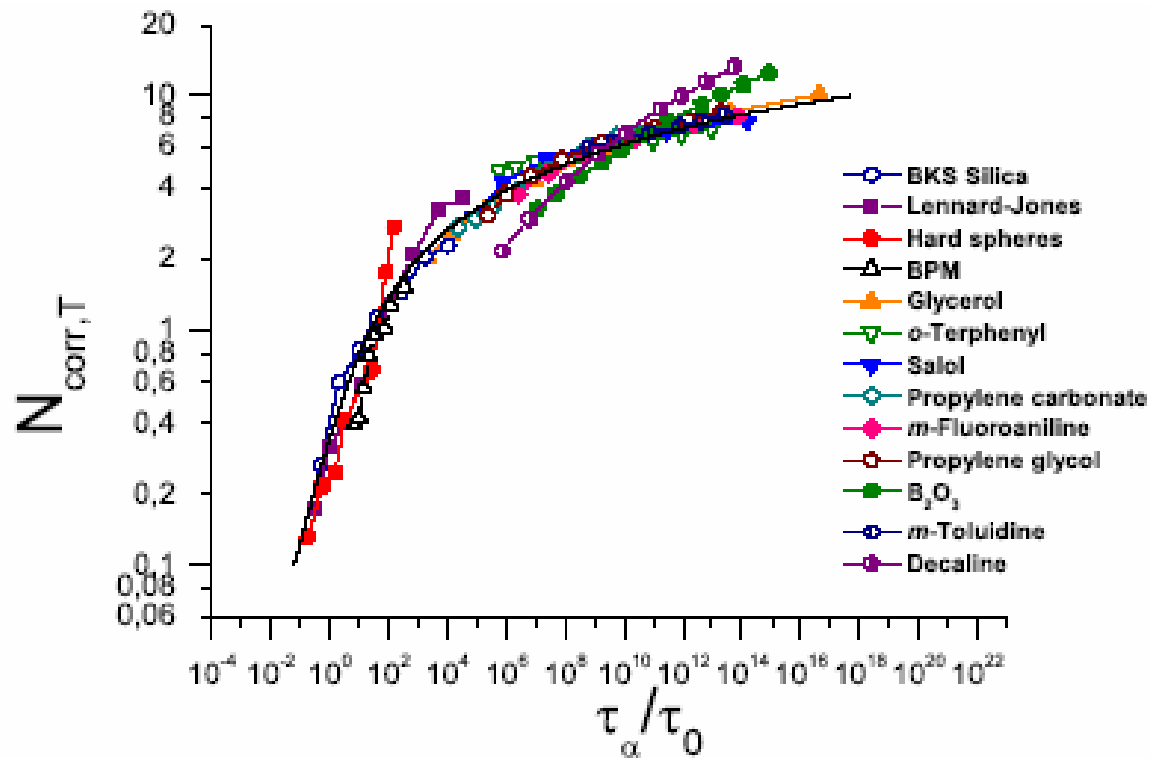


The temperature at which the extrapolated excess of entropy vanishes is very close to the temperature at which the extrapolated viscosity diverges with a Vogel-Fulcher law

Phase transition?

- Pro: extremely fast rise of the relaxation time
- Pro: entropy « vanishing » at T_0
- Pro: universal behavior independent of microscopic details
- Cons \rightarrow Pro: growing correlation length?

Growing dynamical correlations approaching the glass transition



Number of correlated particles: one \rightarrow hundreds

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)

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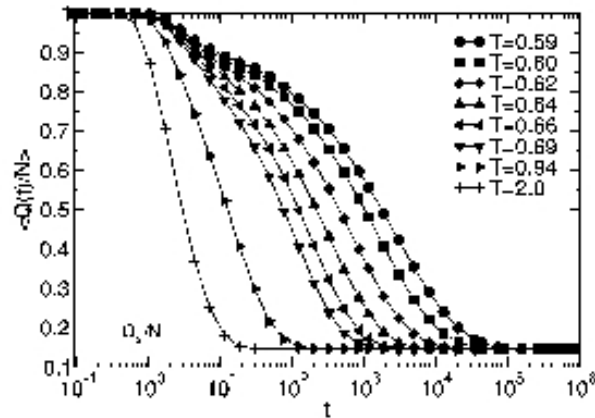
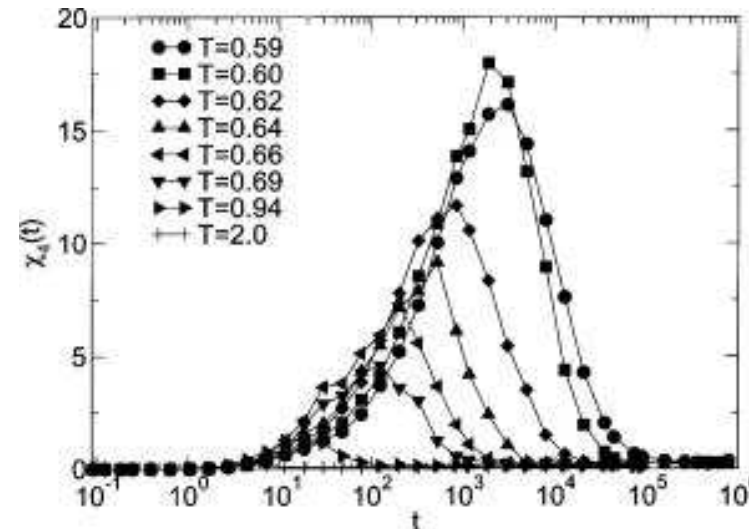


FIG. 3. Average time-dependent overlap "order parameter" $\langle Q(t)/N \rangle$ for each state point simulated. The solid line corresponds to the random value $Q_w/N=0.147$, as discussed in the previous section.

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

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



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

(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.:

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

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

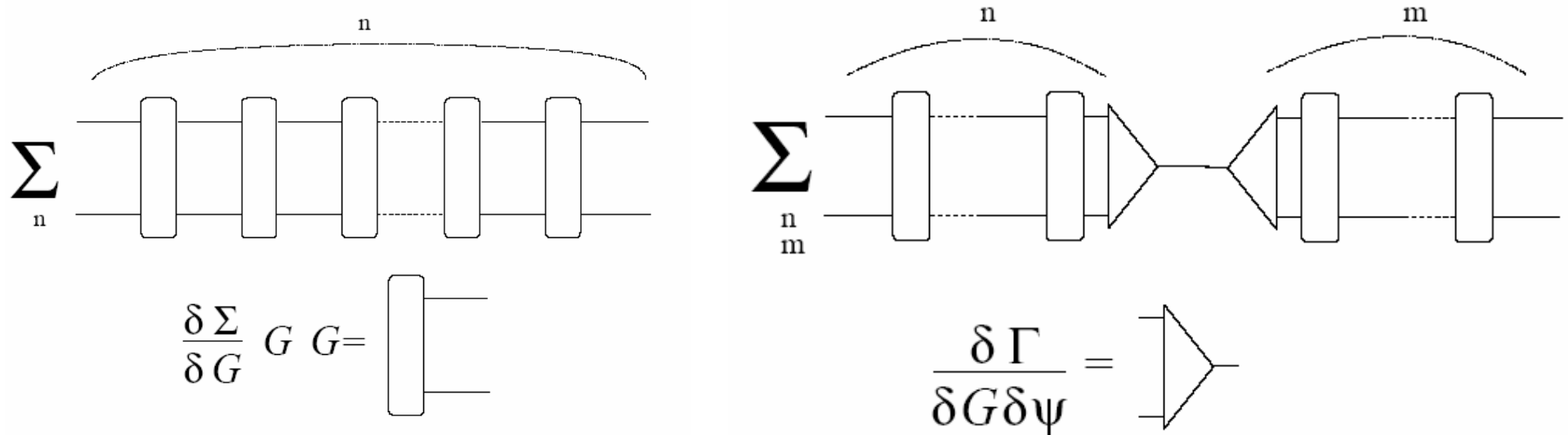
- Field Theory a la Martin-Siggia-Rose-DeDominicis-Janssen (classical limit of Keldysh formalism)

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

with

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

General expression for 4 point dynamical correlations: 2 contributions

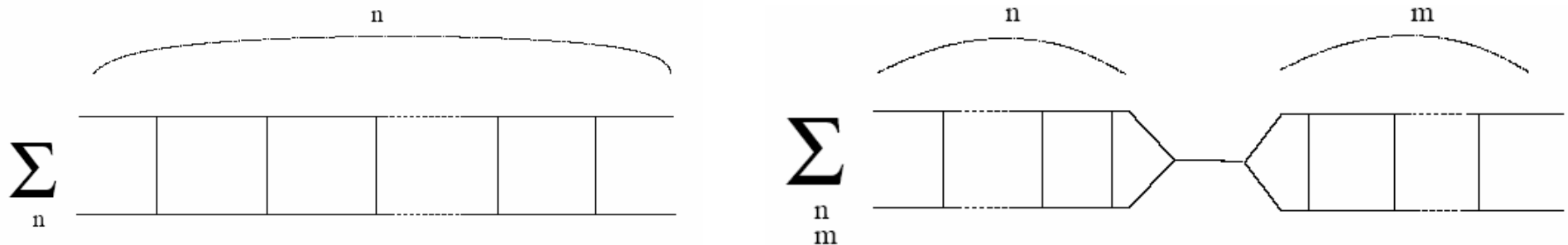


From this diagrammatic expression one can construct a theory of dynamical correlations and find experimentally accessible observables to probe dynamical correlations

One Loop Self Consistent Theory

$$\Sigma = \text{loop} + \text{pole}$$

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

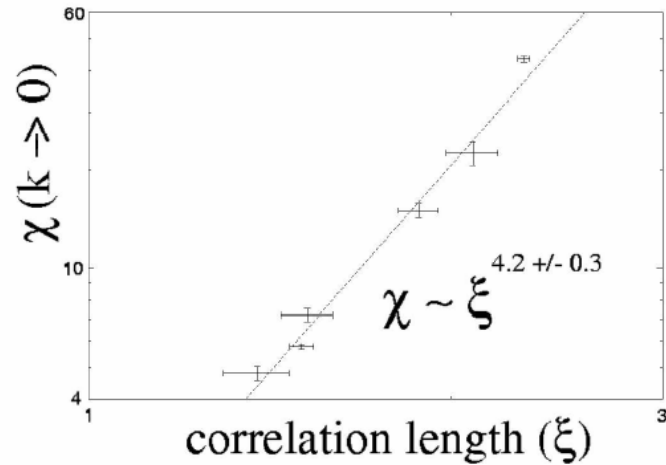


→ Divergent dynamical correlation at the transition

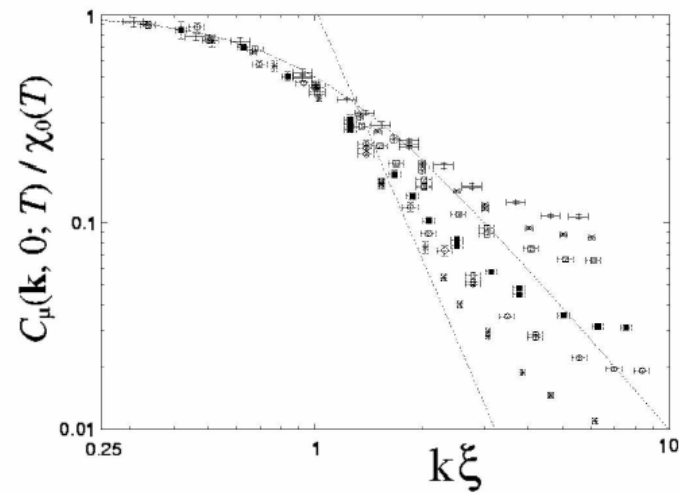
Results

- The sum of all ladders diverges as $1/(T-T_d)$
- 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 $\xi \propto (T - T_d)^{-1/4}$
- $\eta = -2$

Simulations in BMLJ



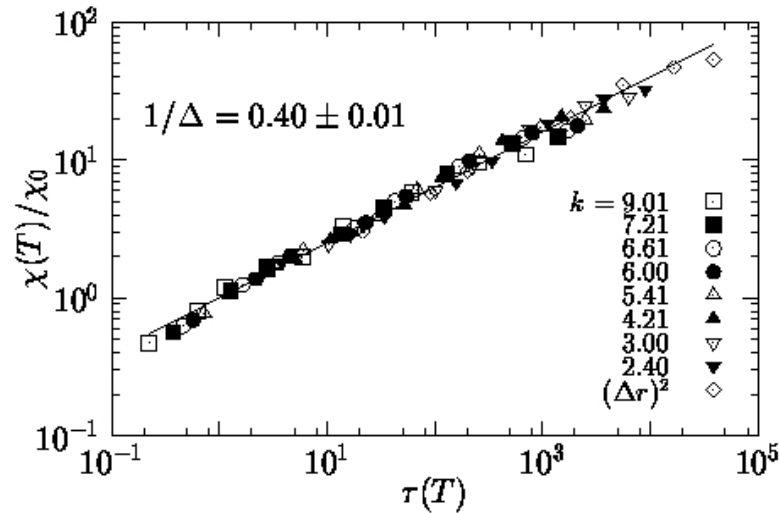
$$v_{MCT} = 1/4$$



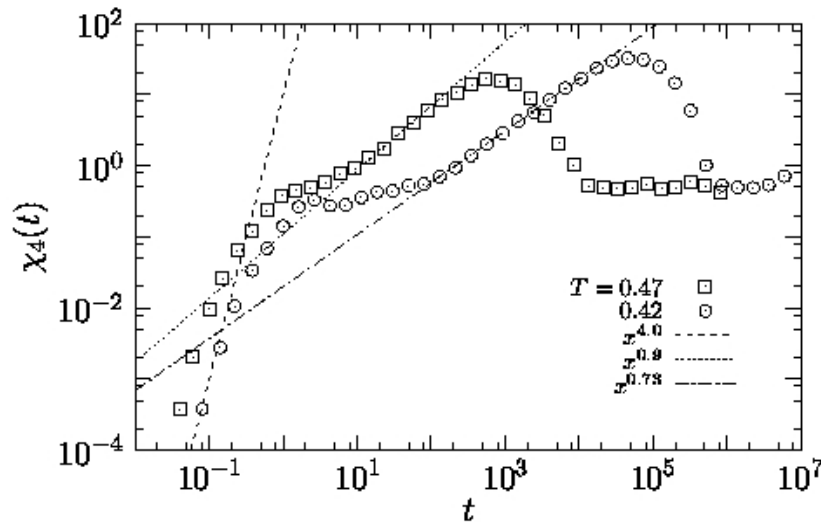
$$\eta_{MCT} = -2$$

Andersen & Stein

Simulations in BMLJ



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

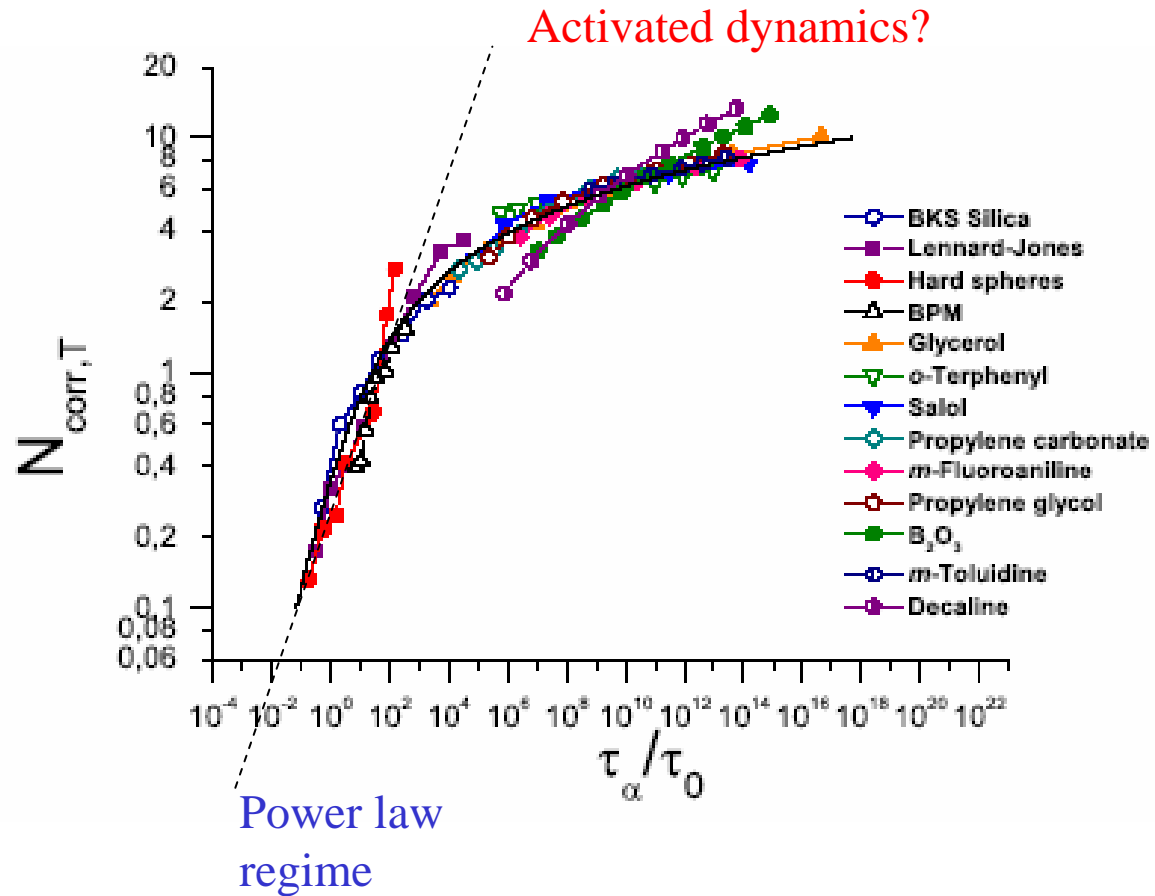


MCT: power law behavior in time before the peak with exponent $b \cong 0.63$

Berthier

Beyond one loop

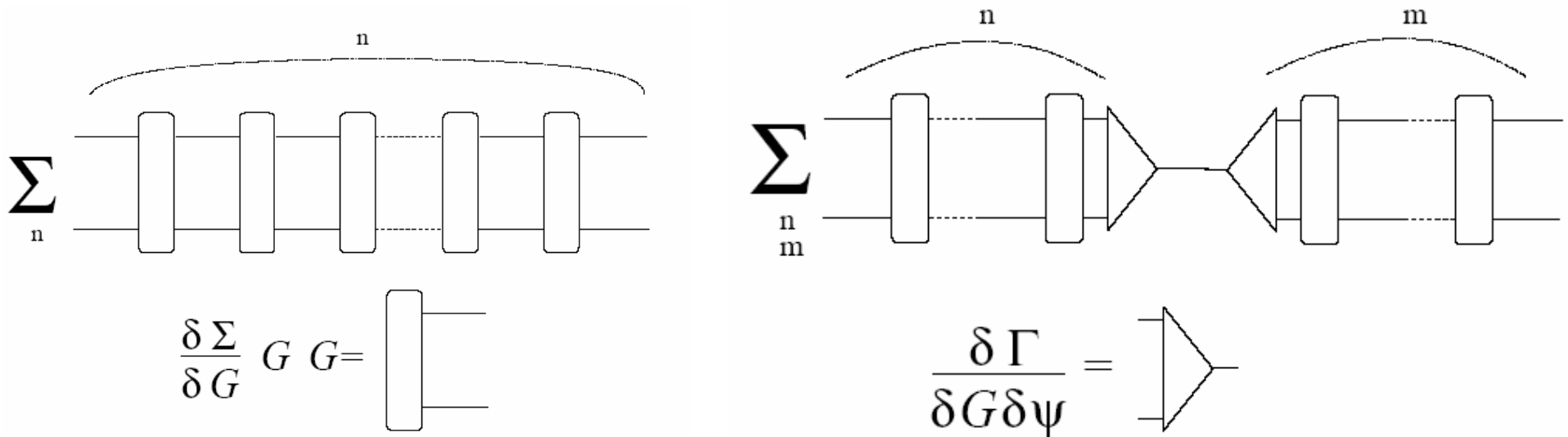
- Results stable at any fixed self-consistent order above 8 dimensions.
- Upper critical dimension is 8 but....
- One has to determine the Ginzburg region...
- ...and compare it to the region where non-perturbative processes transform the transition into a cross-over



Close to the glass transition temperature → activated dynamics

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

New theoretical tools to measure dynamic correlations



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

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

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

