Dynamical heterogeneity at the jamming transition of a colloidal suspension

Luca Cipelletti[1,2], Pierre Ballesta[1,3], Agnès Duri[1,4]

[1]LCVN Université Montpellier 2 and CNRS, France

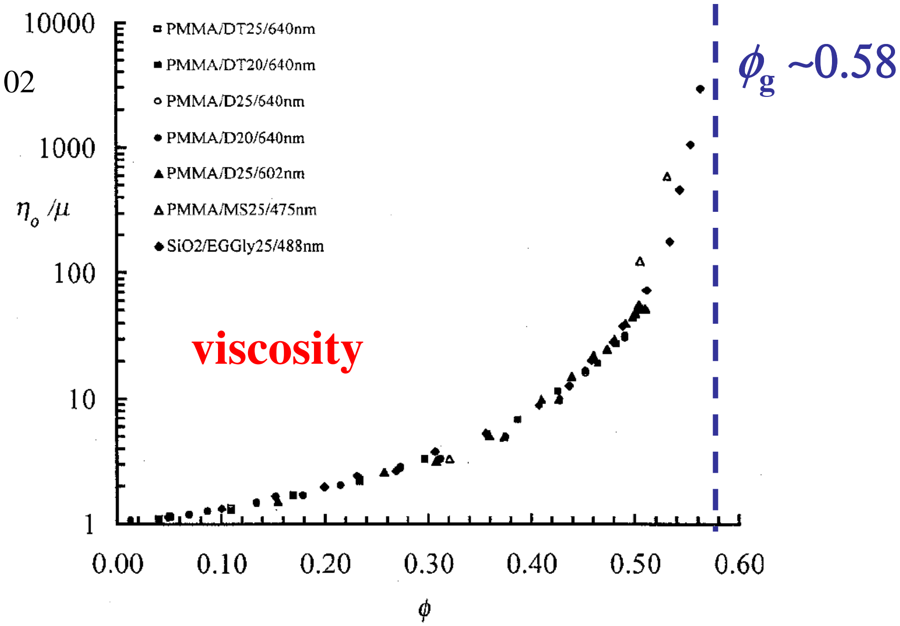
[2]Institut Universitaire de France

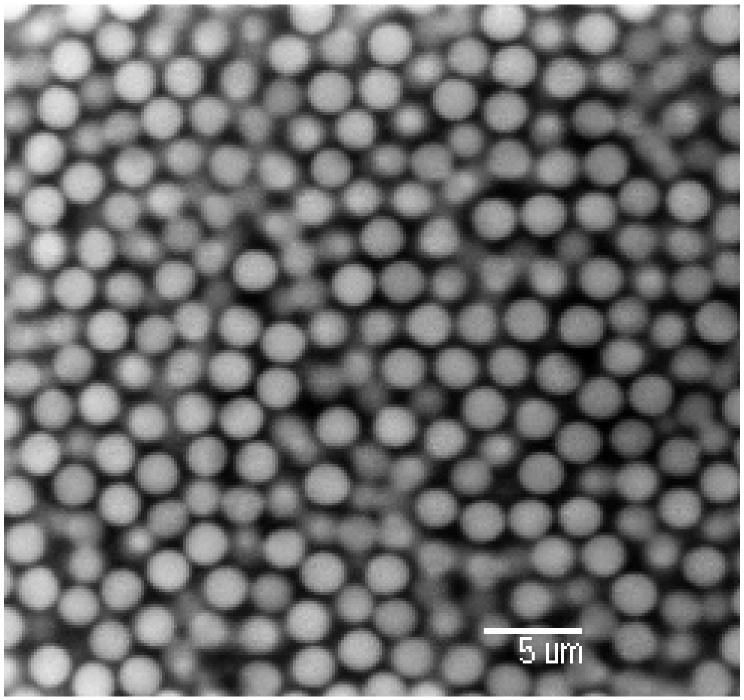
[3]SUPA, University of Edinburgh

[4]Desy, Hamburg

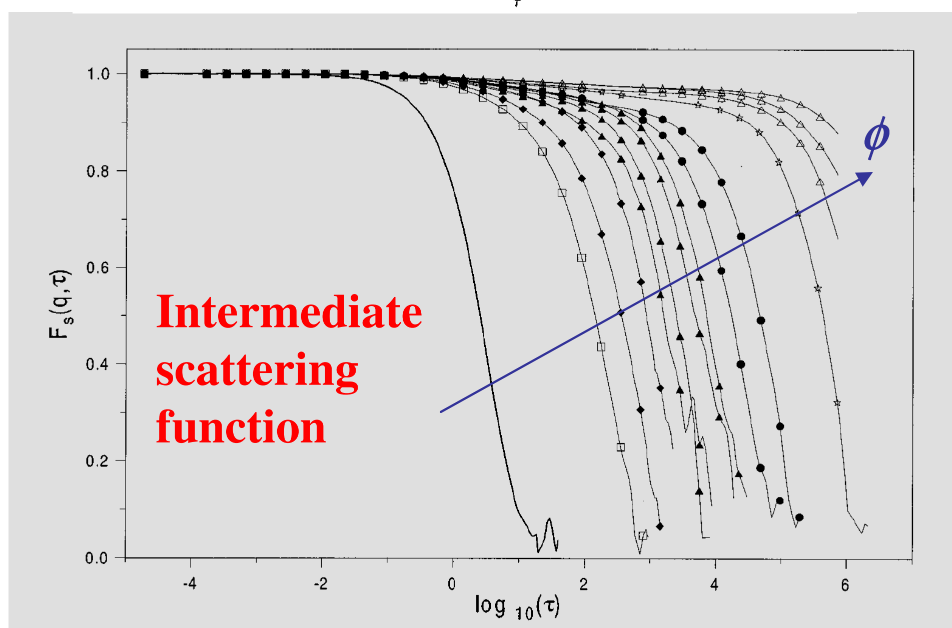
Soft glassy materials

Cheng et al. PRE 02



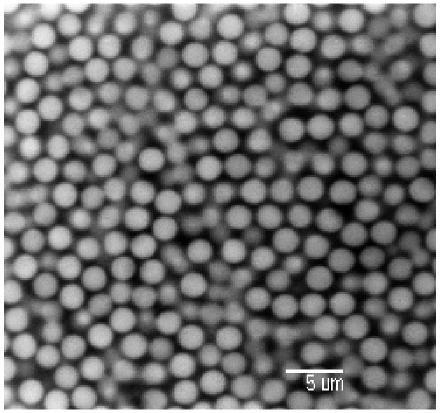


Confocal microscopy image by Eric Weeks



Van Megen et al. PRE 98

Soft glassy materials

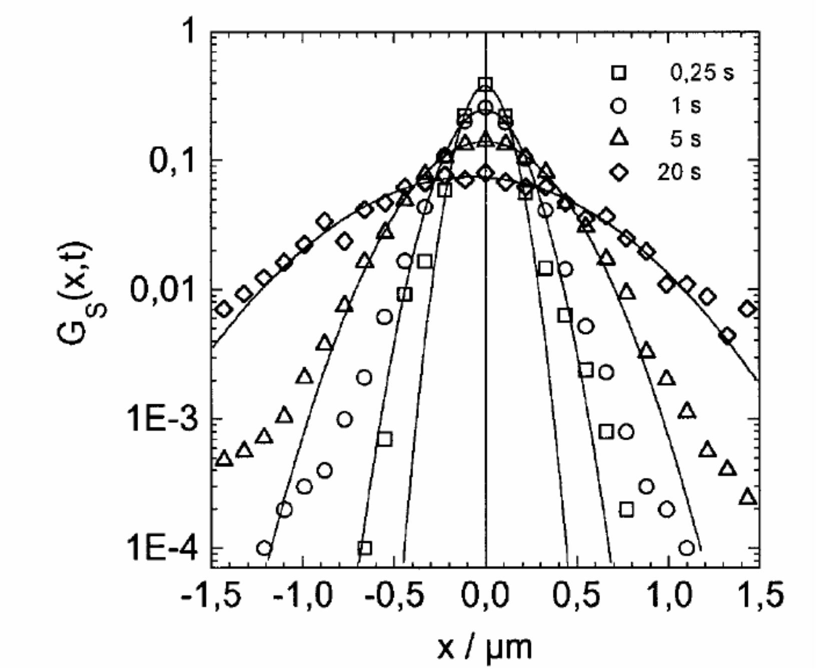


Outline

- **What** are dynamical heterogeneities ?
- **Why** should we care about DH ?
- **How** can we measure DH ?
- **DH** (very) close to jamming

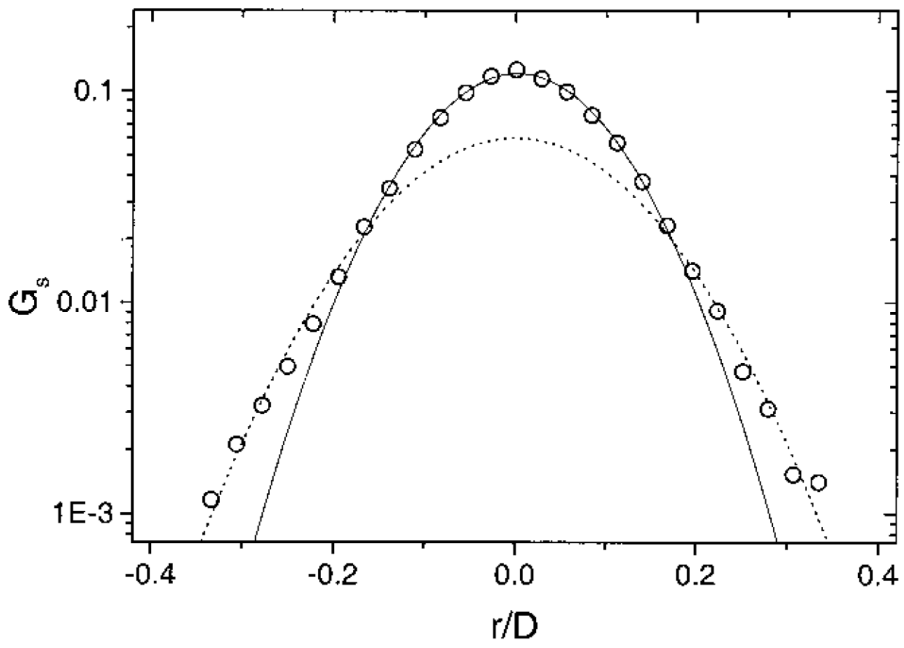
Dynamical Heterogeneity

PDF of particle displacements in a dense colloidal suspension



Optical microscopy

Kasper et al. Langmuir 98

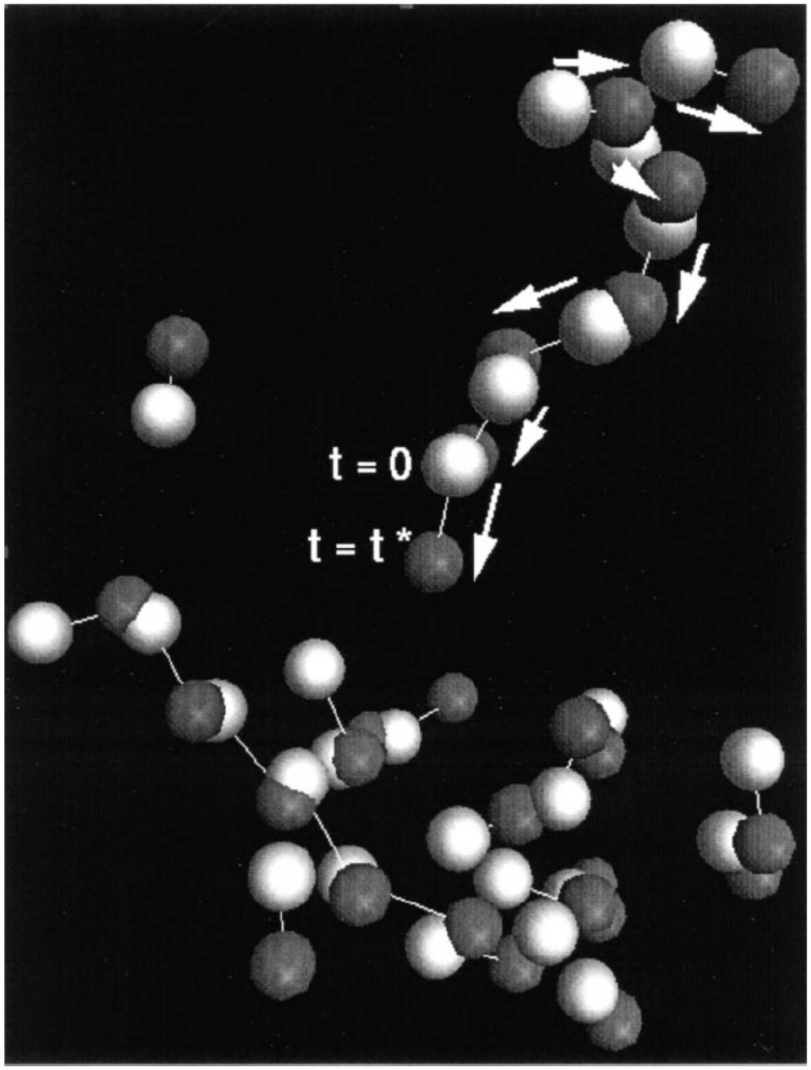


Confocal microscopy

Kegel et al. Science 00

Dynamical Heterogeneity

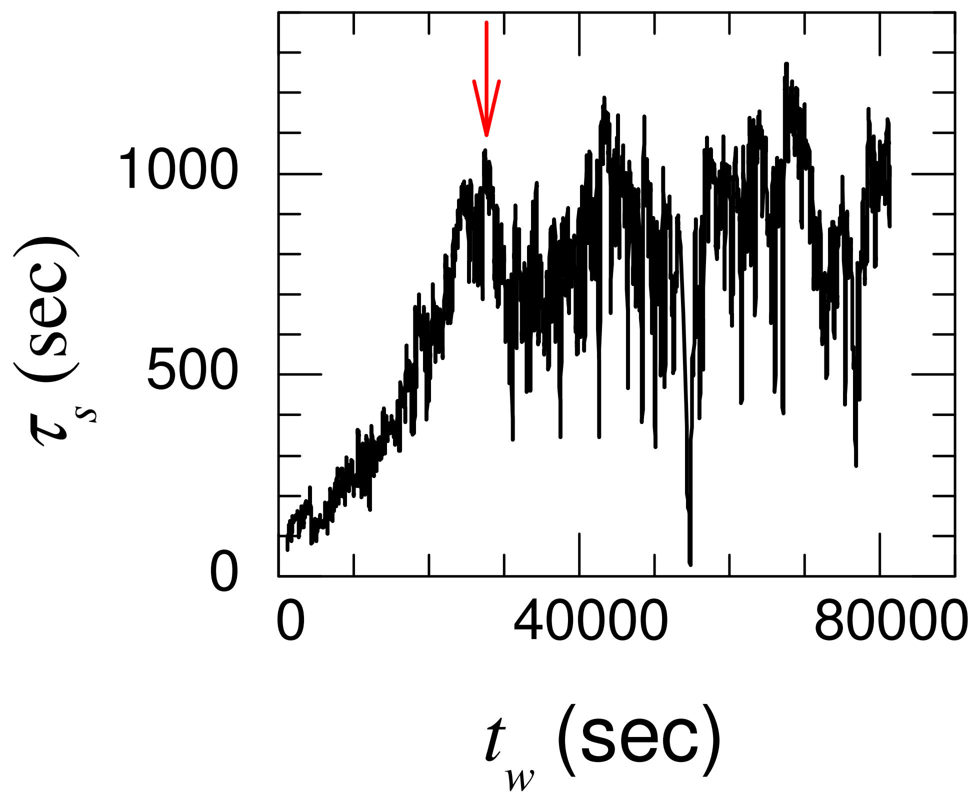
String-like motion in a LJ supercooled fluid



Donati et al. PRL 98

Dynamical Heterogeneity

Relaxation time in a colloidal suspension near close packing



Why are DHs important?

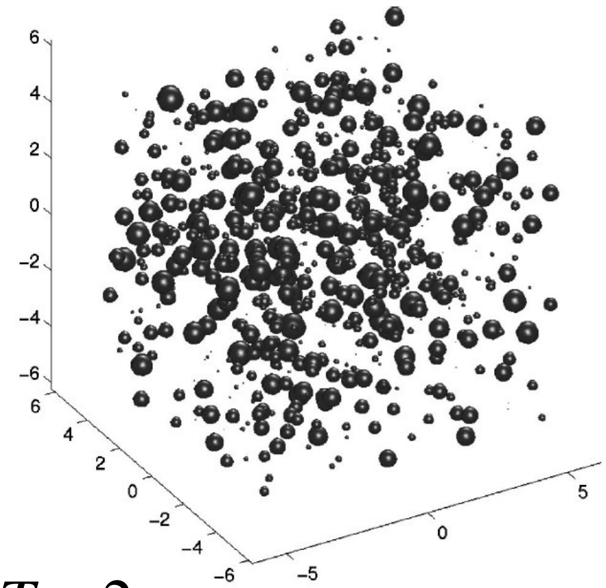
Crucial role in the slowing down of the dynamics close to the glass transition

- Adam-Gibbs: relaxation through **cooperatively rearranging regions.** Their size **increases** approaching the glass transition.

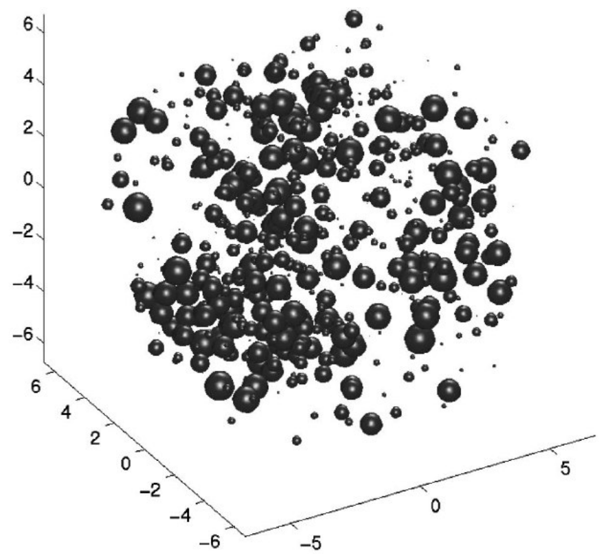
- Glass transition as a (dynamical) **critical phenomenon** ?

- DHs may allow one to **discriminate between competing theories**

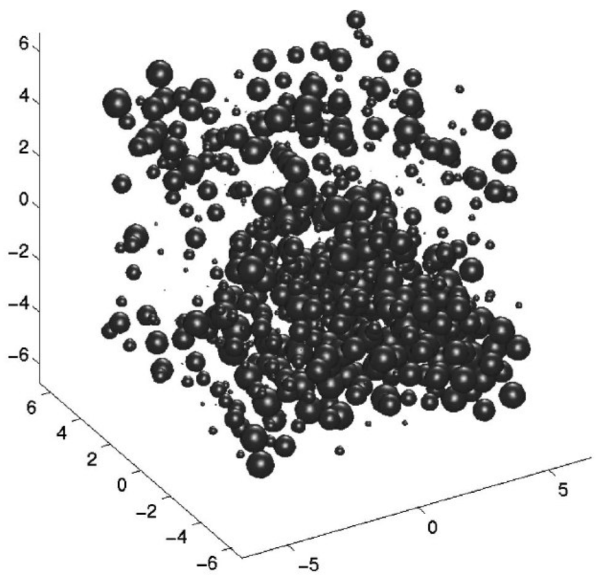
Size of DH: simulations



$T = 2$



$T = 0.6$



$T = 0.45$

Less mobile particles in a LJ supercooled fluid (T_c ~ 0.435)

Berthier PRE 04

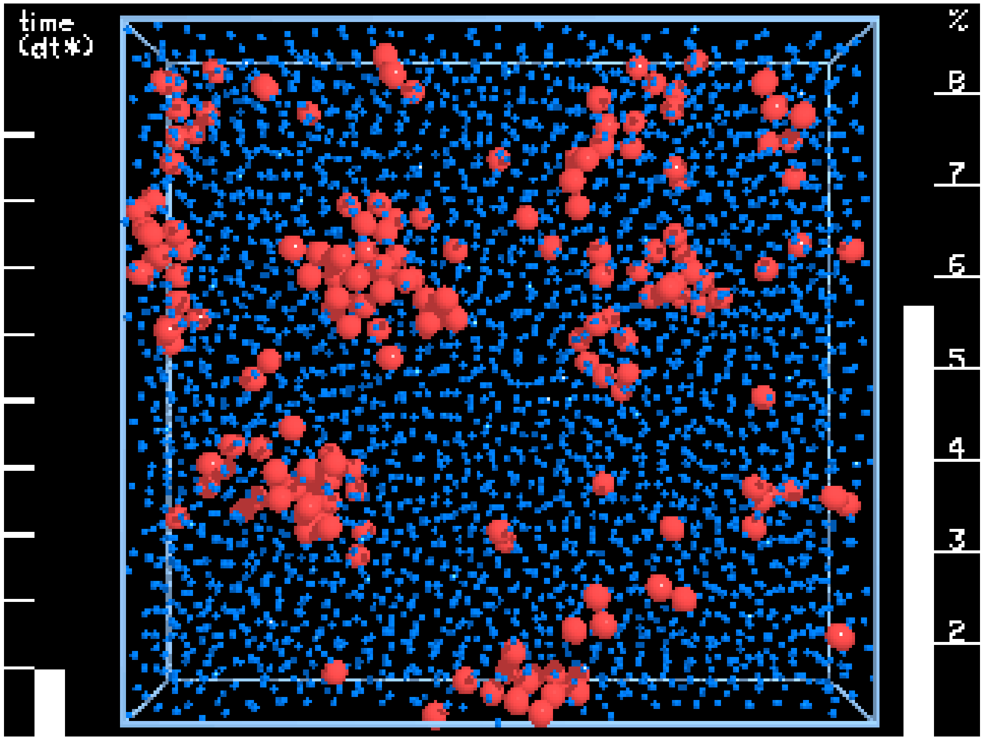
What quantities should we measure?

Space and time-resolved correlation functions $f(t,t+\tau,\mathbf{r})$ or particle displacement

- **Simulations** (far from T_g!)
- (Confocal) **microscopy** on colloidal systems
- **Granular** systems (2D, athermal, see Dauchot’s talk)

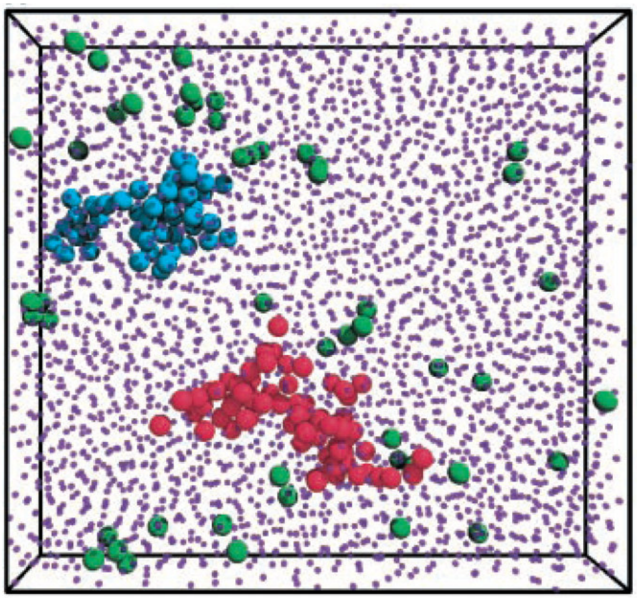
Confocal microscopy on colloidal HS

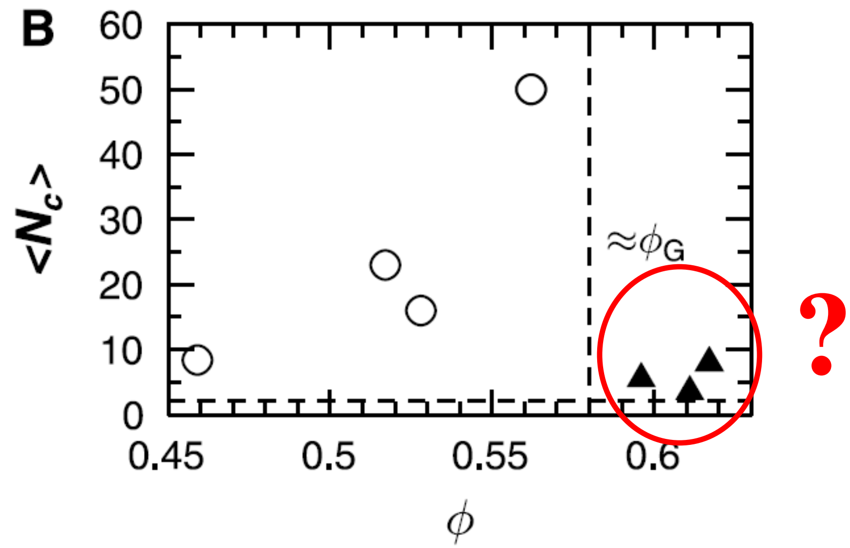
From E. Weeks web page

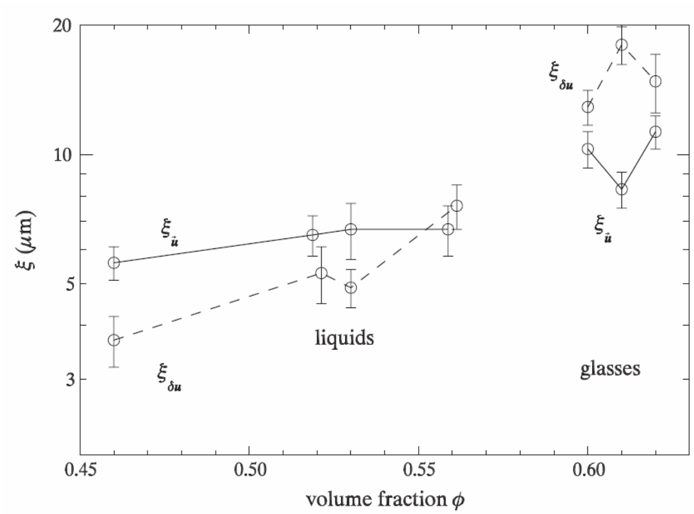


Confocal microscopy on colloidal HS

Weeks et al. Science 00







Weeks et al.,
J. Phys. Cond.
Mat 07

What quantities should we measure?

Space- and time-resolved correlation functions $f(t,t+\tau,\mathbf{r})$ or particle displacement

- **Simulations** (far from T_g!)
- (Confocal) **microscopy** on colloidal systems

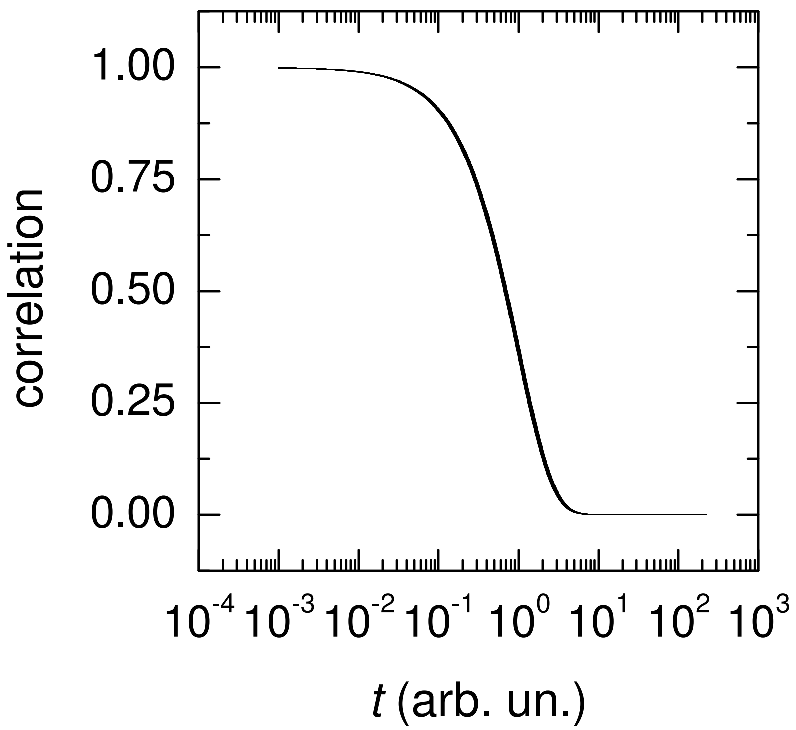
(limited statistics, stringent requirements on particles (size, optical mismatch…))

- **Granular** systems (2D, athermal, see

Dauchot's talk)

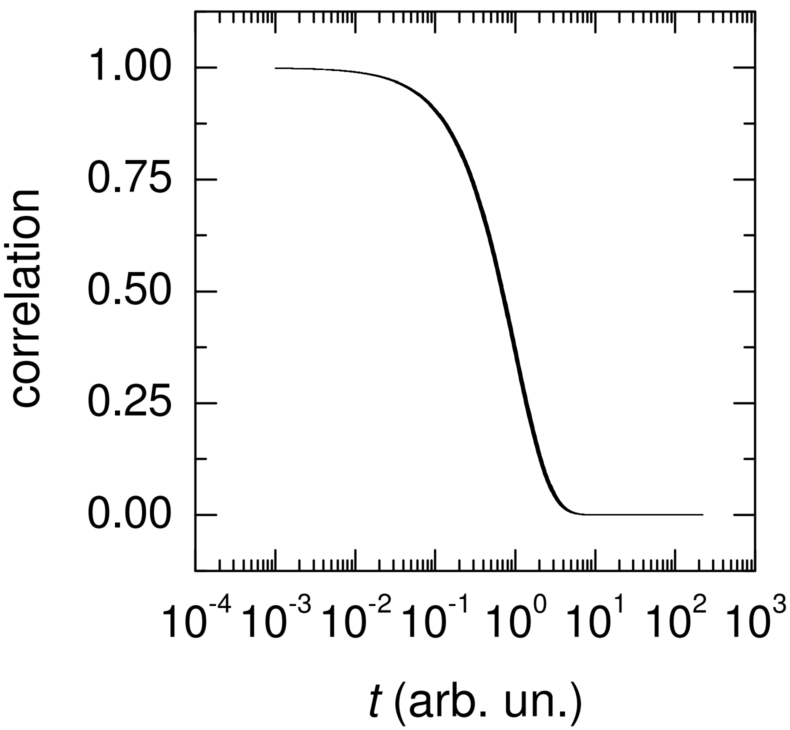
Time-resolved correlation functions $f(t,t+\tau)$ (no space resolution)

Temporally heterogeneous dynamics

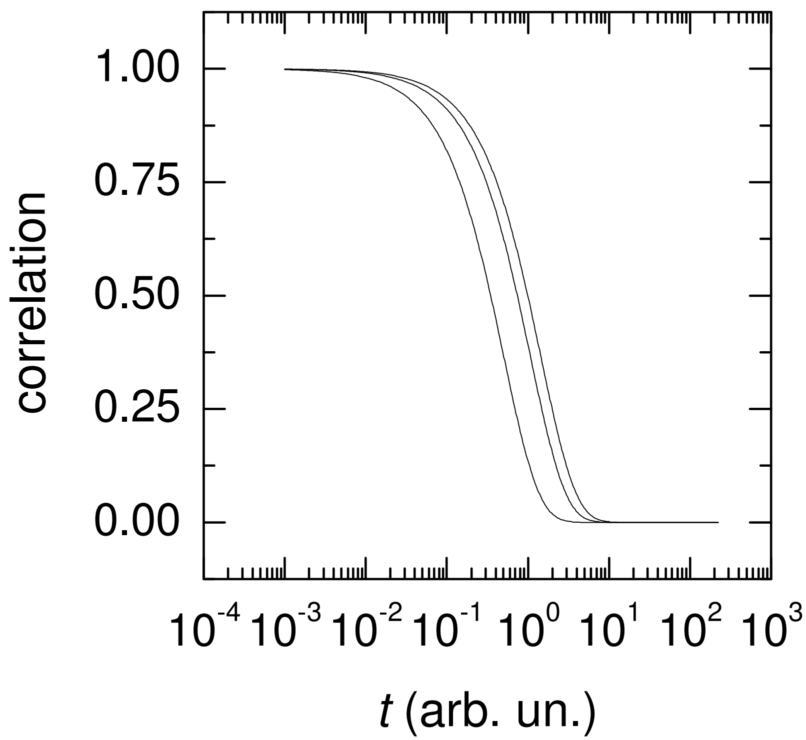


homogeneous

Temporally heterogeneous dynamics

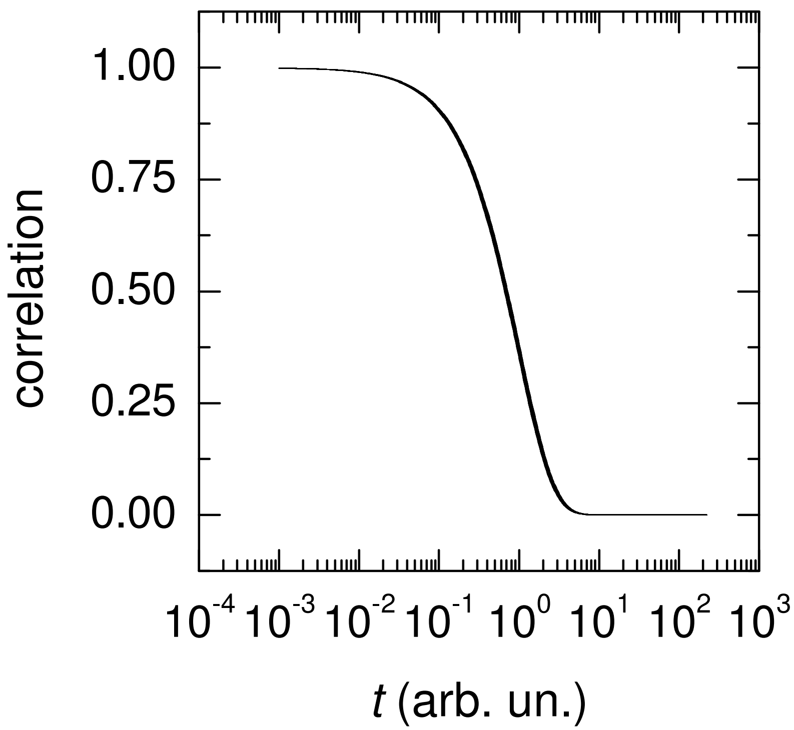


homogeneous

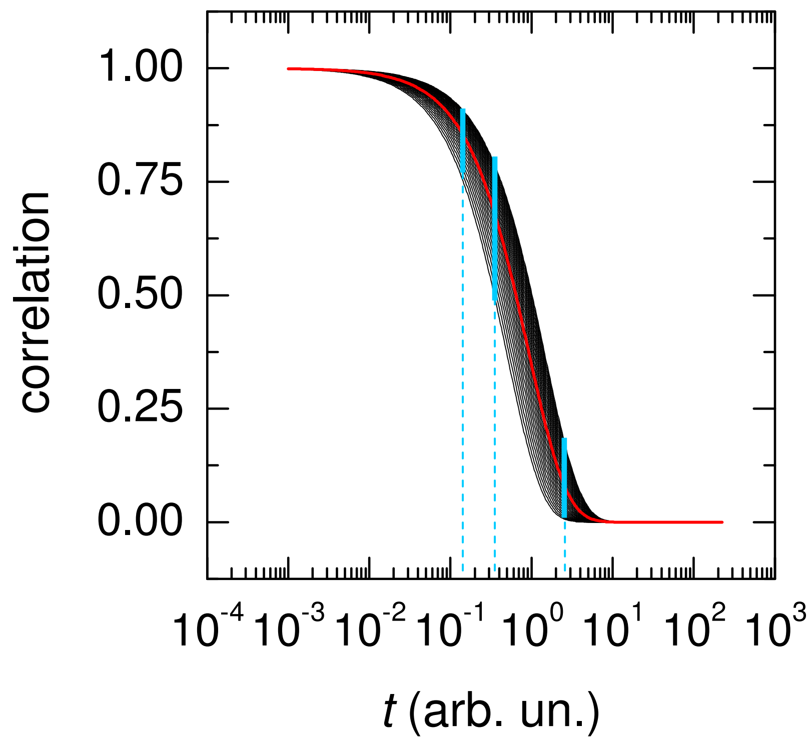


heterogeneous

Temporally heterogeneous dynamics



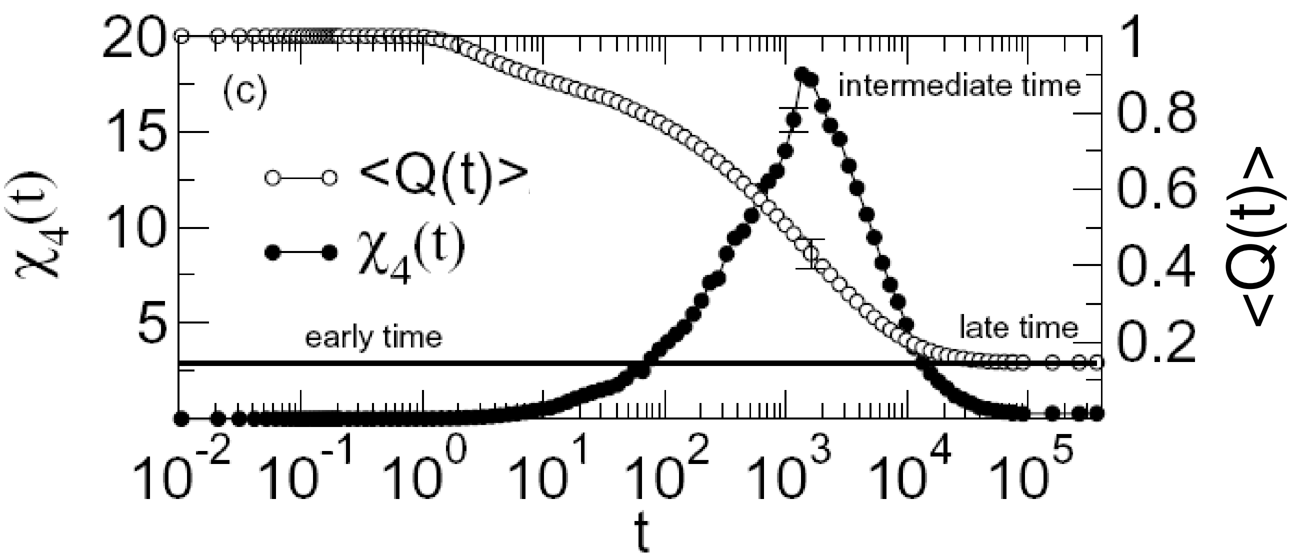
homogeneous



heterogeneous

Dynamical susceptibility in glassy systems

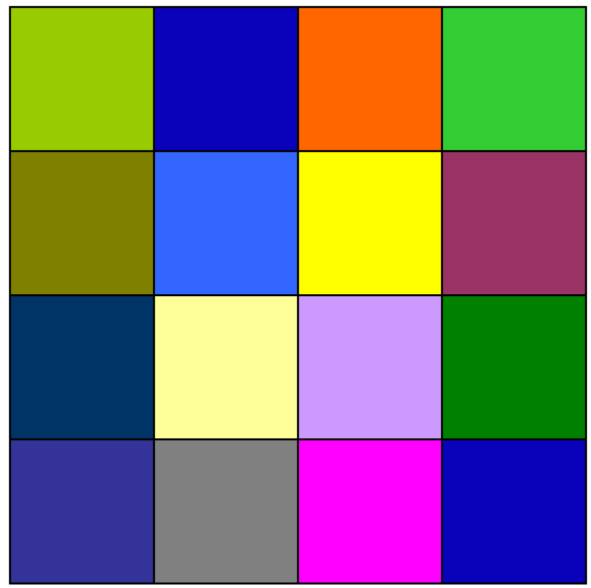
Supercooled liquid (Lennard-Jones)



Lacevic et al., PRE 2002

$$\chi_4 = N \, \mathrm{var}[Q(t)]$$

Dynamical susceptibility in glassy systems



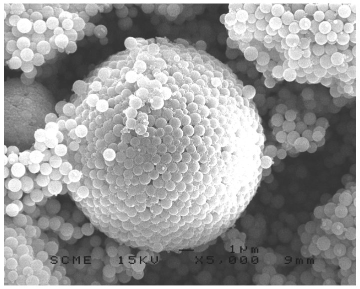
N_{blob} regions

$$\chi_4 = N\,\text{var}[Q(t)] \sim N\,(1/N_{\text{blob}}) = N/N_{\text{blob}}$$

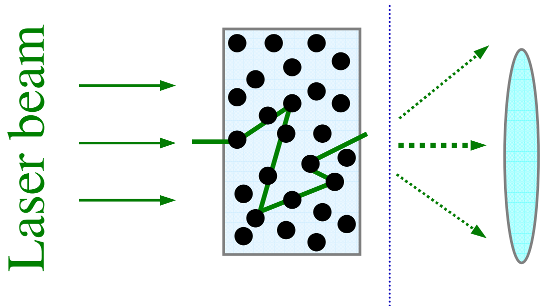
$$\chi_4(\tau) \sim \left\langle \int d^3\mathbf{r}\, f\left(0,t',t'+t\right) f\left(\mathbf{r},t',t'+t\right) \right\rangle_{t'}$$

How can we measure χ_4?

Time-resolved light scattering experiments (TRC)



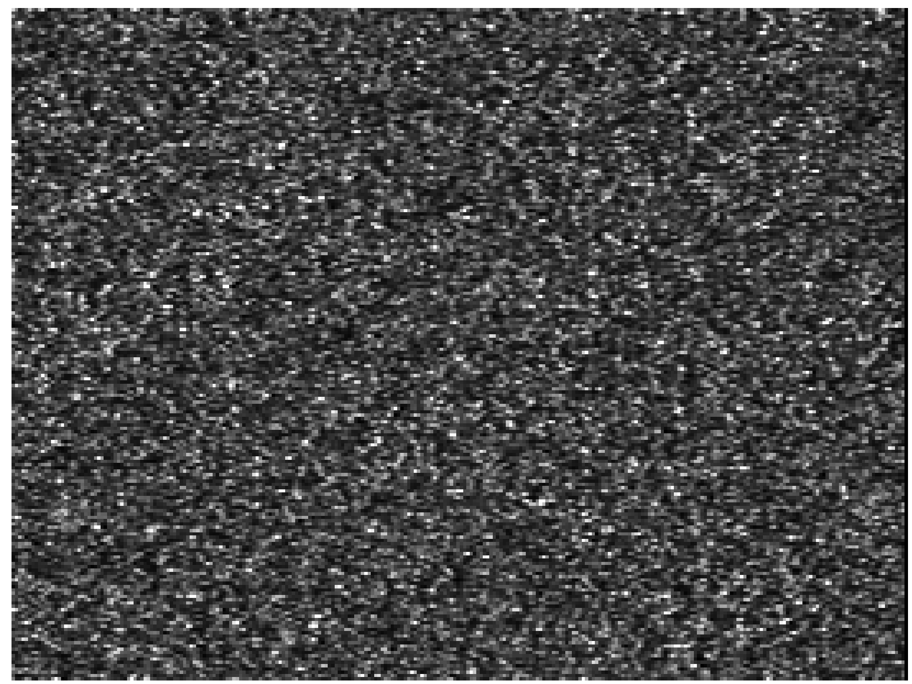
Experimental setup



Random walk w/ step l^*

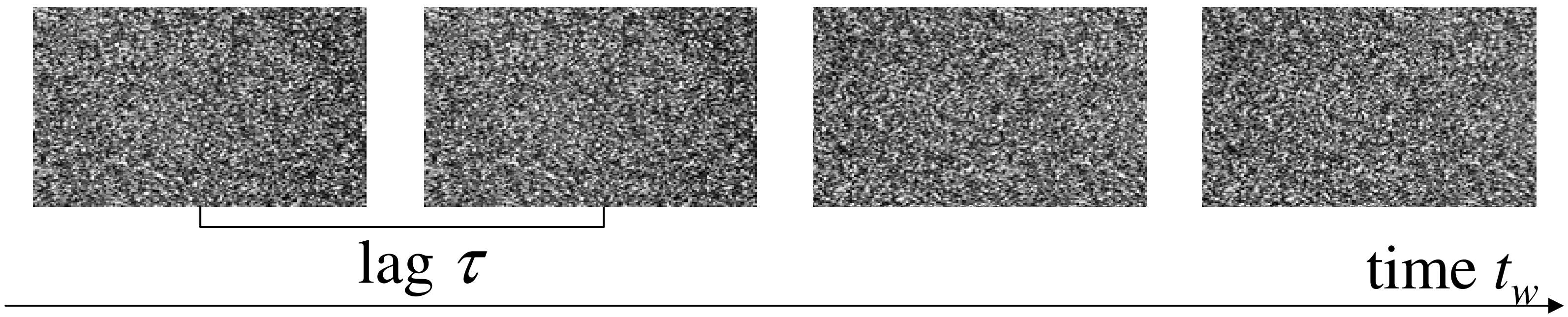
CCD Camera

CCD-based (multispeckle)
Diffusing Wave Spectroscopy



Change in speckle field mirrors change in sample configuration

Time Resolved Correlation

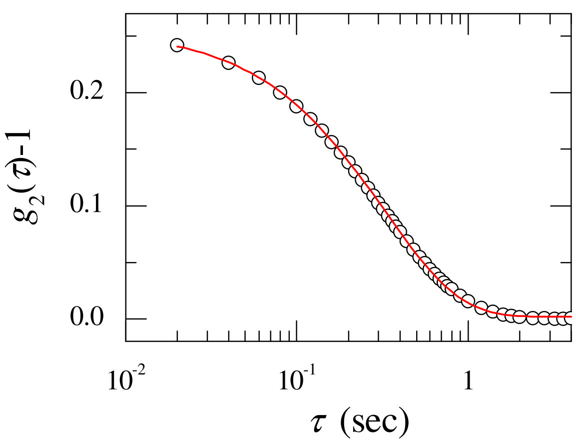


degree of correlation $$c_I(t_w,\tau) = \frac{< I_p(t_w)\, I_p(t_w+\tau) >_p}{< I_p(t_w) >_p < I_p(t_w+\tau) >_p} - 1$$

degree of correlation $c_I(t_w, \tau) = \dfrac{< I_p(t_w)\, I_p(t_w + \tau) >_p}{< I_p(t_w) >_p < I_p(t_w + \tau) >_p} - 1$

Average over t_w ⬇

intensity correlation function **$g_2(\tau) - 1$**



$g_2(\tau) - 1$ ➡ Average dynamics

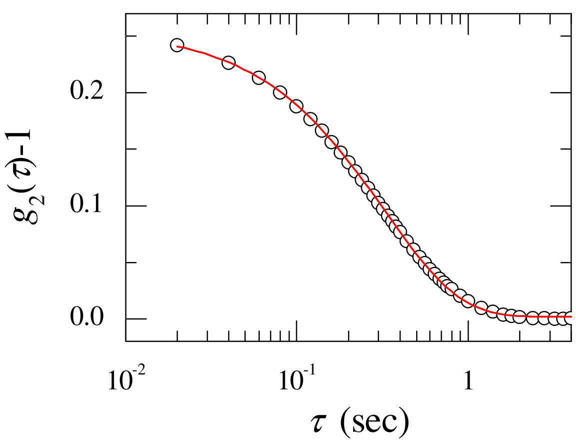
degree of correlation $c_I(t_w,\tau) = \dfrac{<I_p(t_w)\, I_p(t_w+\tau)>_p}{<I_p(t_w)>_p <I_p(t_w+\tau)>_p} - 1$

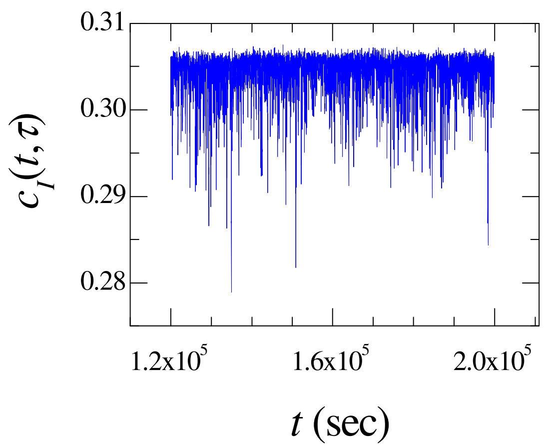
Average over t_w

intensity correlation function $g_2(\tau)-1$

fixed τ, *vs.* t_w

fluctuations of the dynamics

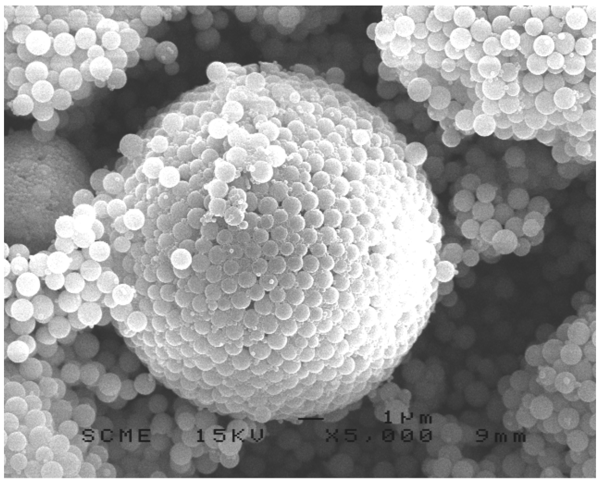




$g_2(\tau)-1$ → Average dynamics

$\mathrm{var}(c_I)(\tau)$ → 'dynamical susceptibility' $\chi_4(\tau)$

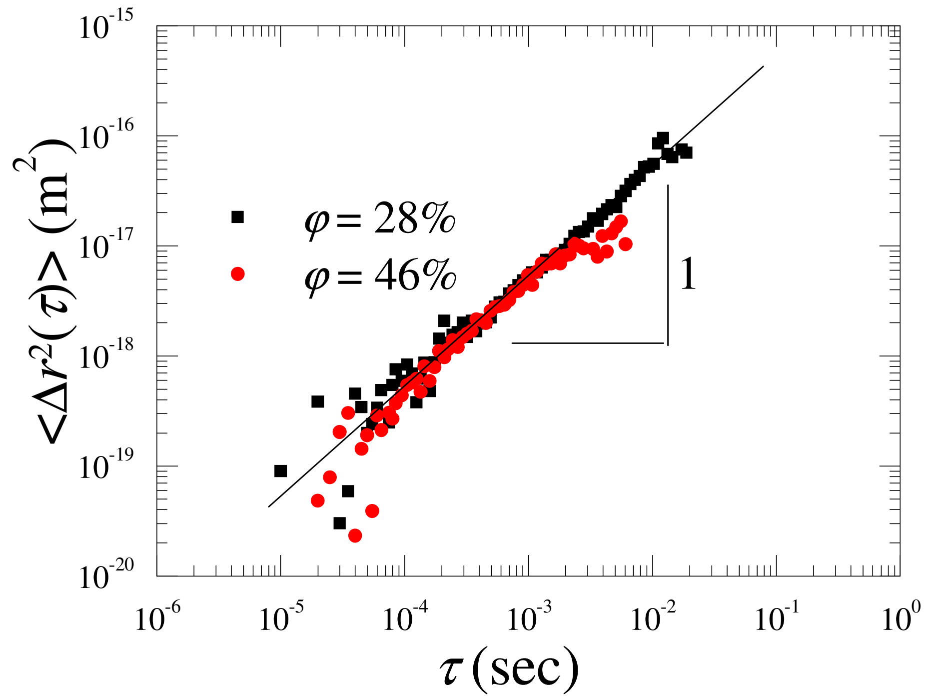
Experimental system



PVC xenospheres in DOP

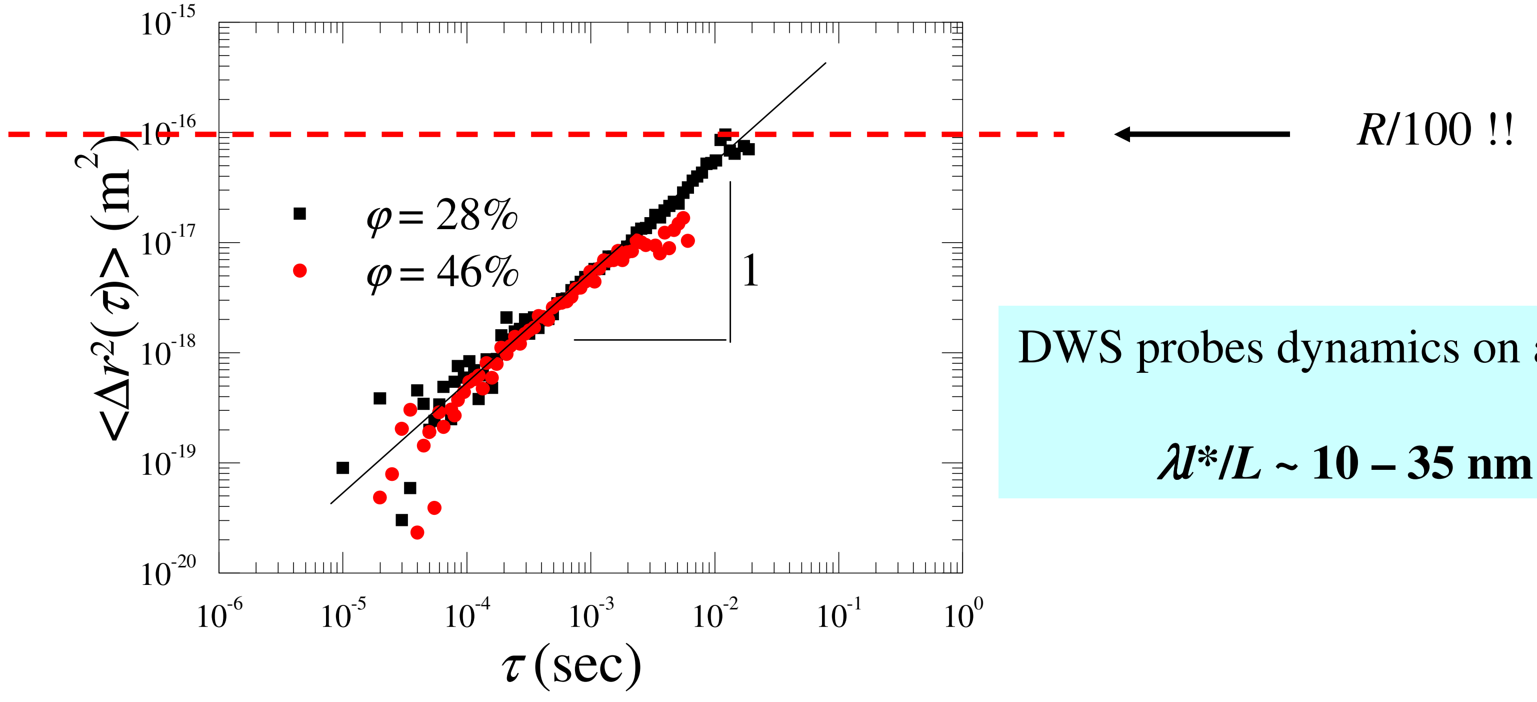
- radius $R \sim 10$ µm
- Polydisperse
- Brownian
- Excluded volume interactions
- $\varphi = 64\% - 75\%$ (Note: $\varphi_g \sim 58\%$)

« Diluted » samples



Brownian behavior

« Diluted » samples

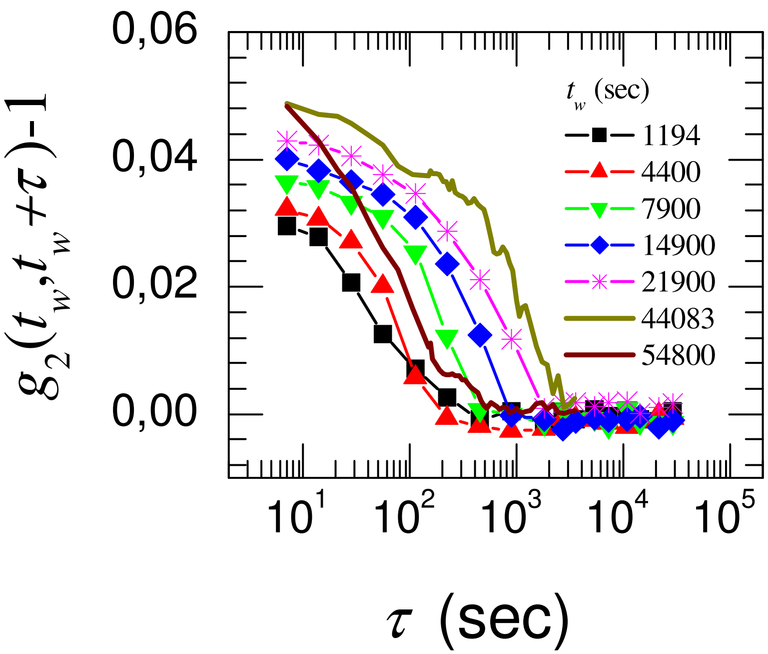


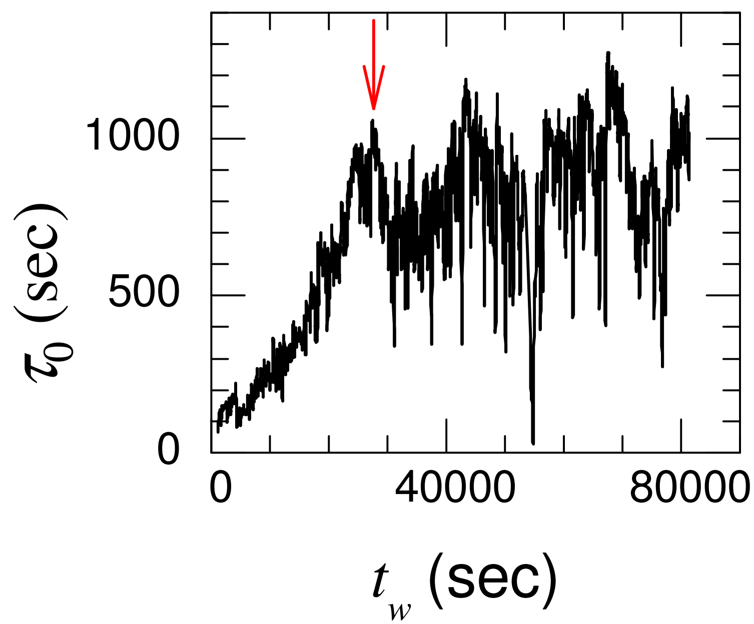
DWS probes dynamics on a length scale

$\lambda l^*/L$ **~ 10 – 35 nm << *R***

2-time intensity correlation function

$\phi = 66.4\%$

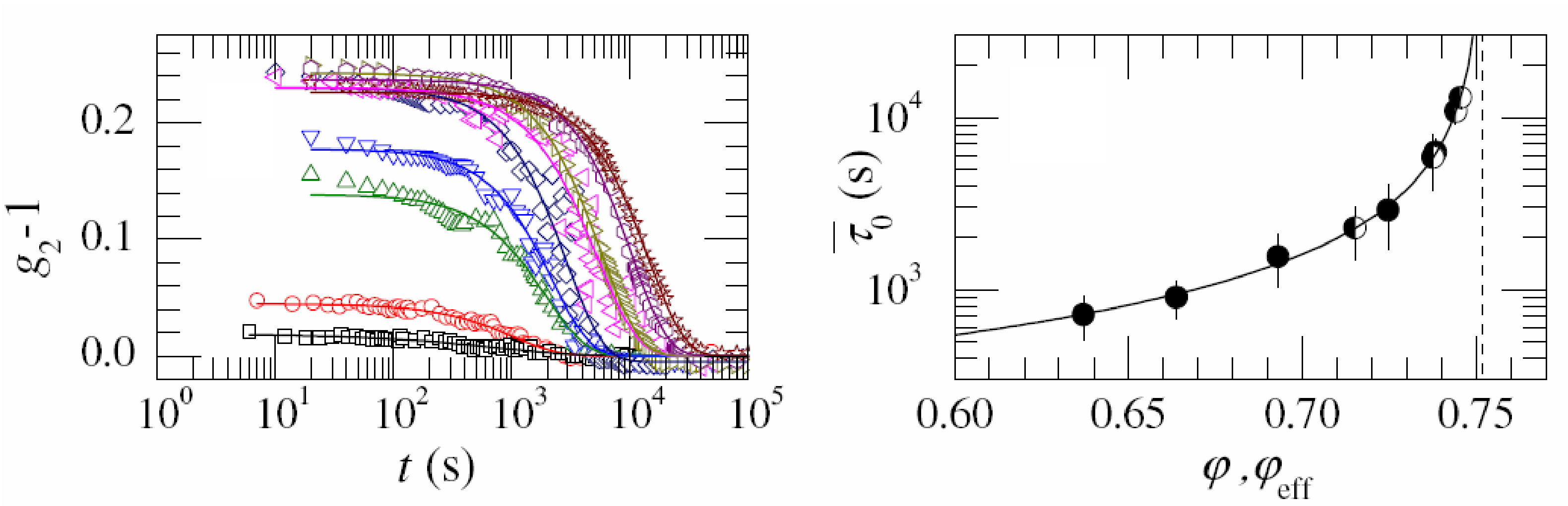




Fit: $g_2(t_w,t_w+\tau) - 1 = a\exp[-(\tau/\tau_0)^\beta]$

- Initial regime: « simple aging » ($\tau_s \sim t_w^{1.1 \pm 0.1}$)
- Crossover to stationary dynamics, **large fluctuations of τ_s**

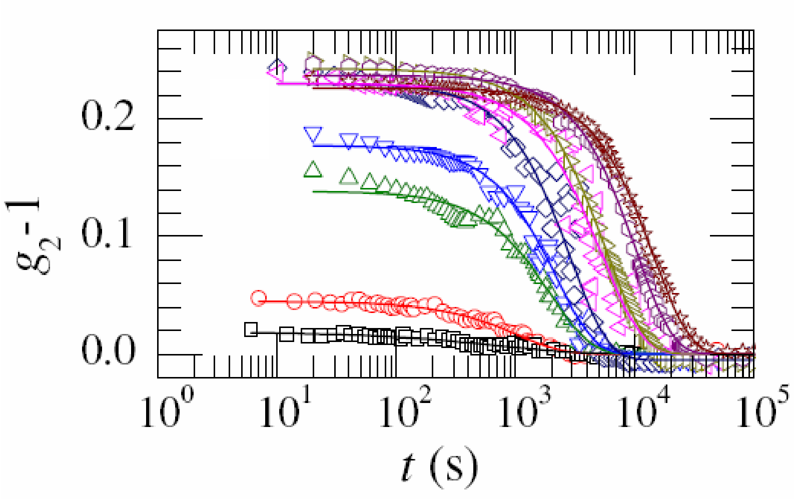
Average dynamics

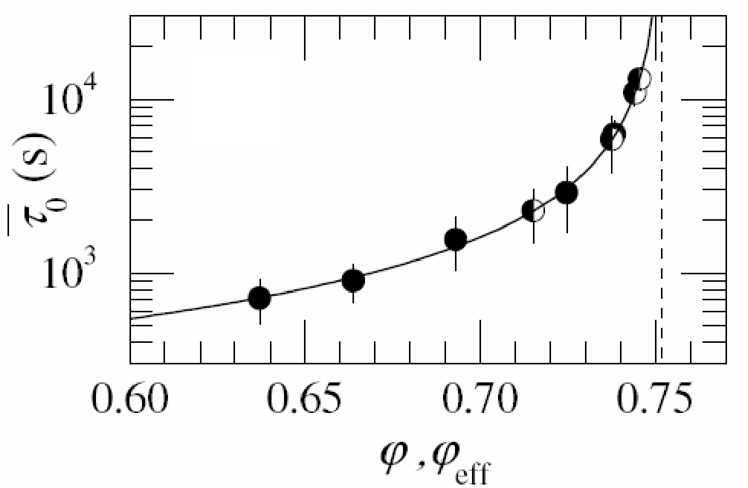


Relaxation time $\overline{\tau}_0 \sim \dfrac{1}{\left|\varphi - \varphi_c\right|^{1.01\pm0.04}}$

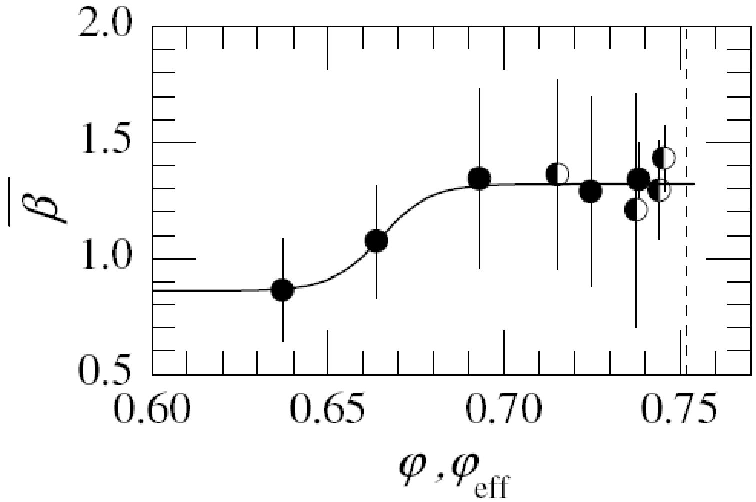
$\varphi_c = 0.752$

Average dynamics



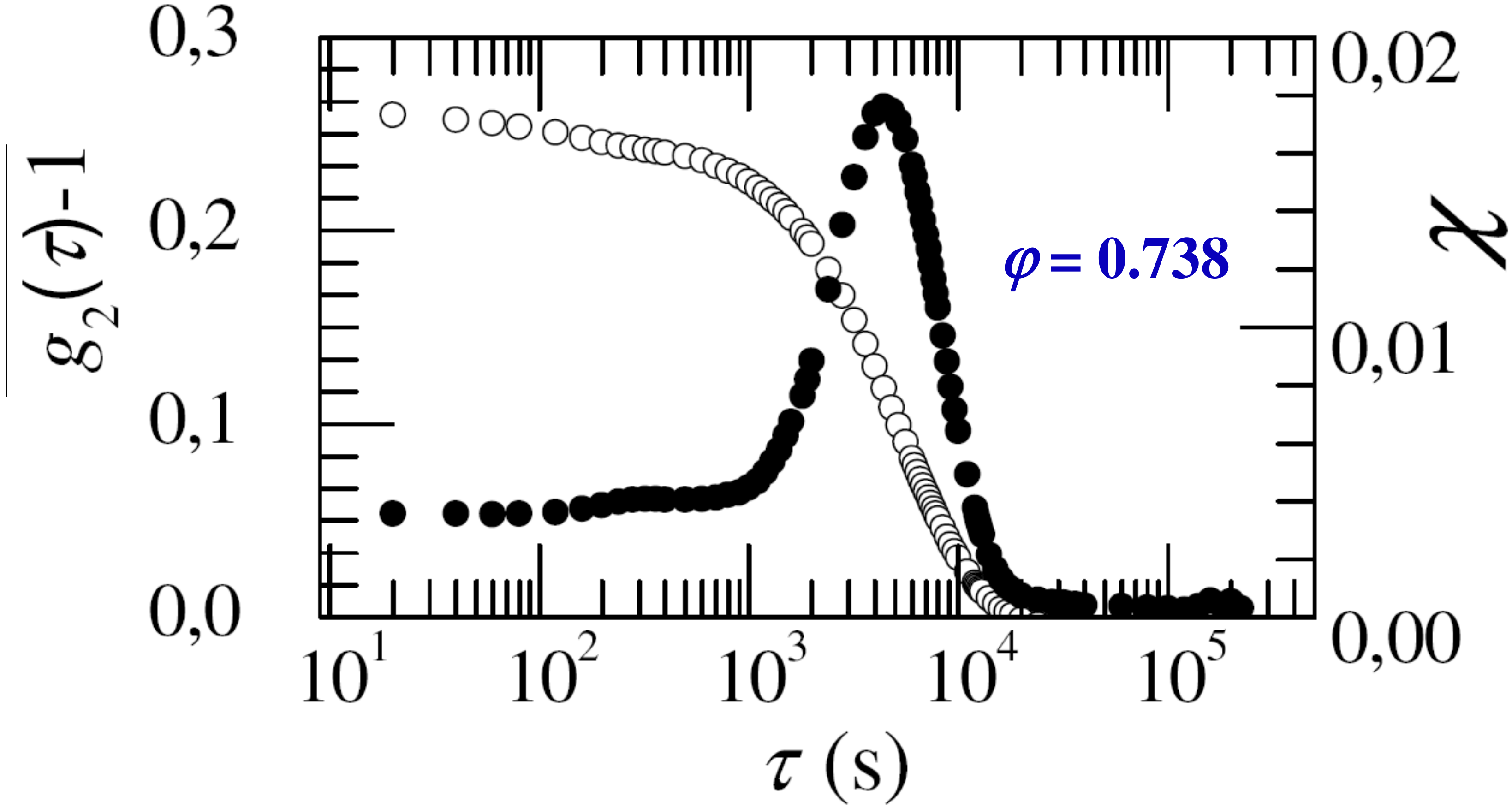


Stretching exponent $\overline{\beta}$

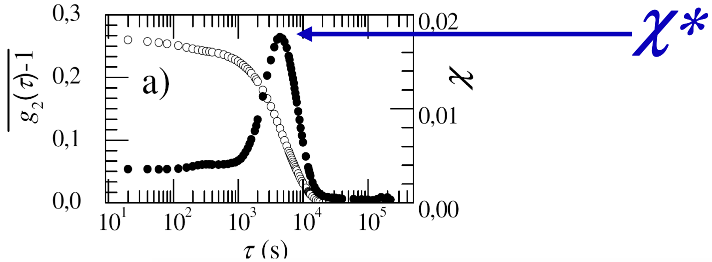


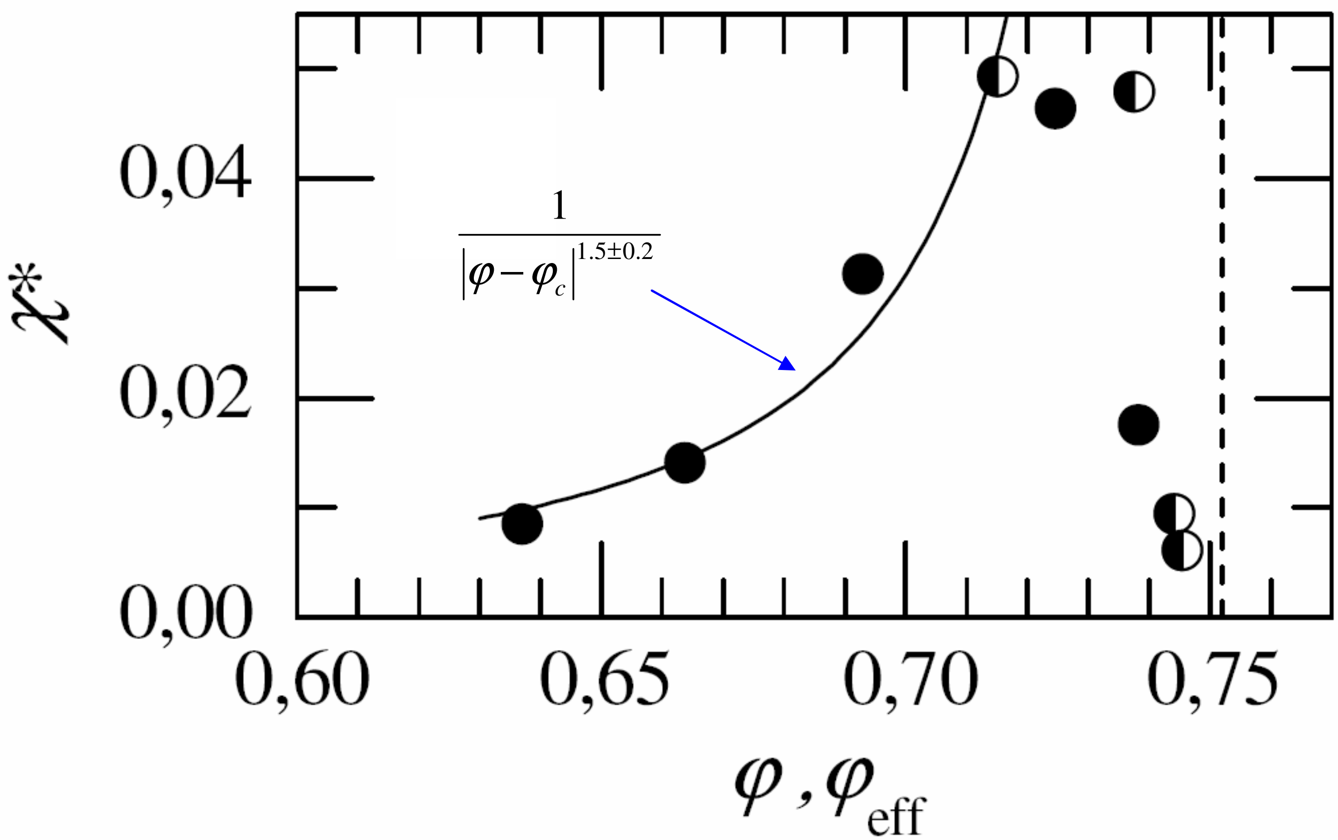
Fluctuations of the dynamics: χ

$$\chi(\tau) \equiv \chi(\tau,\varphi) = \overline{\left(c_I(t,\tau) - \overline{c_I(t,\tau)}\right)^2} / \overline{a}^2$$

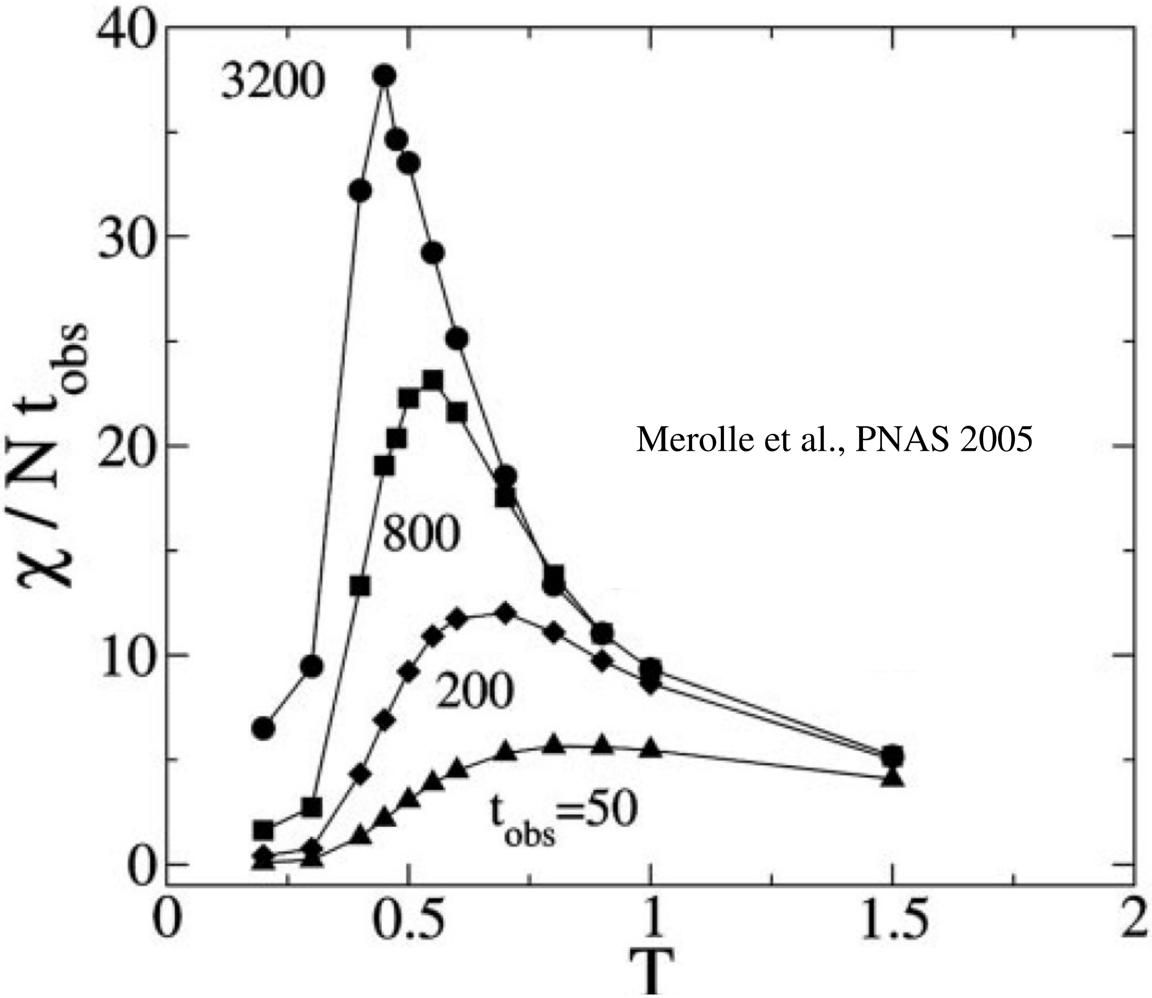


Fluctuations of the dynamics: χ^* *vs* φ

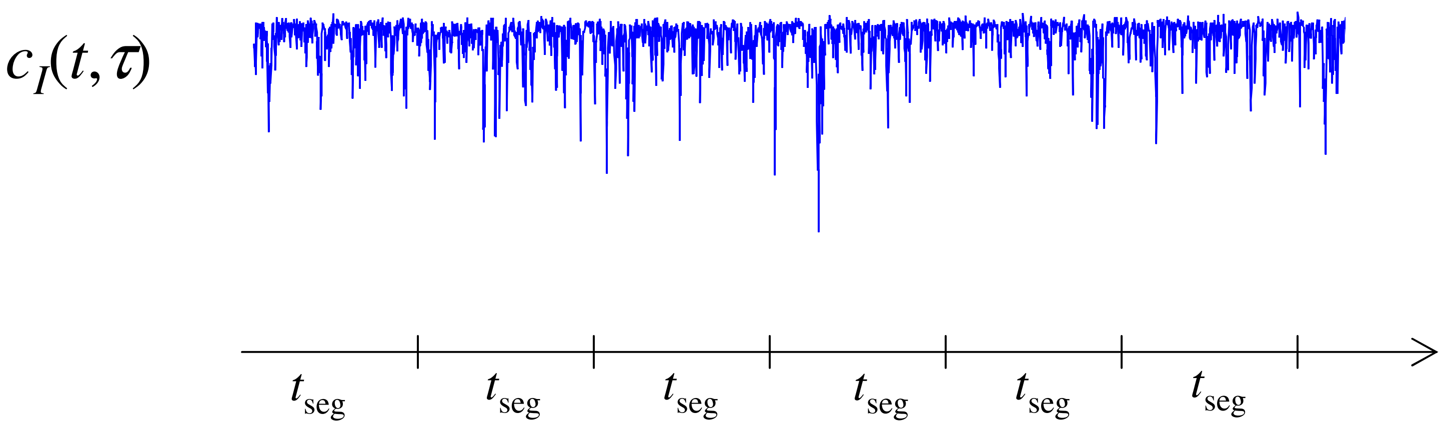




Measurement time issue?

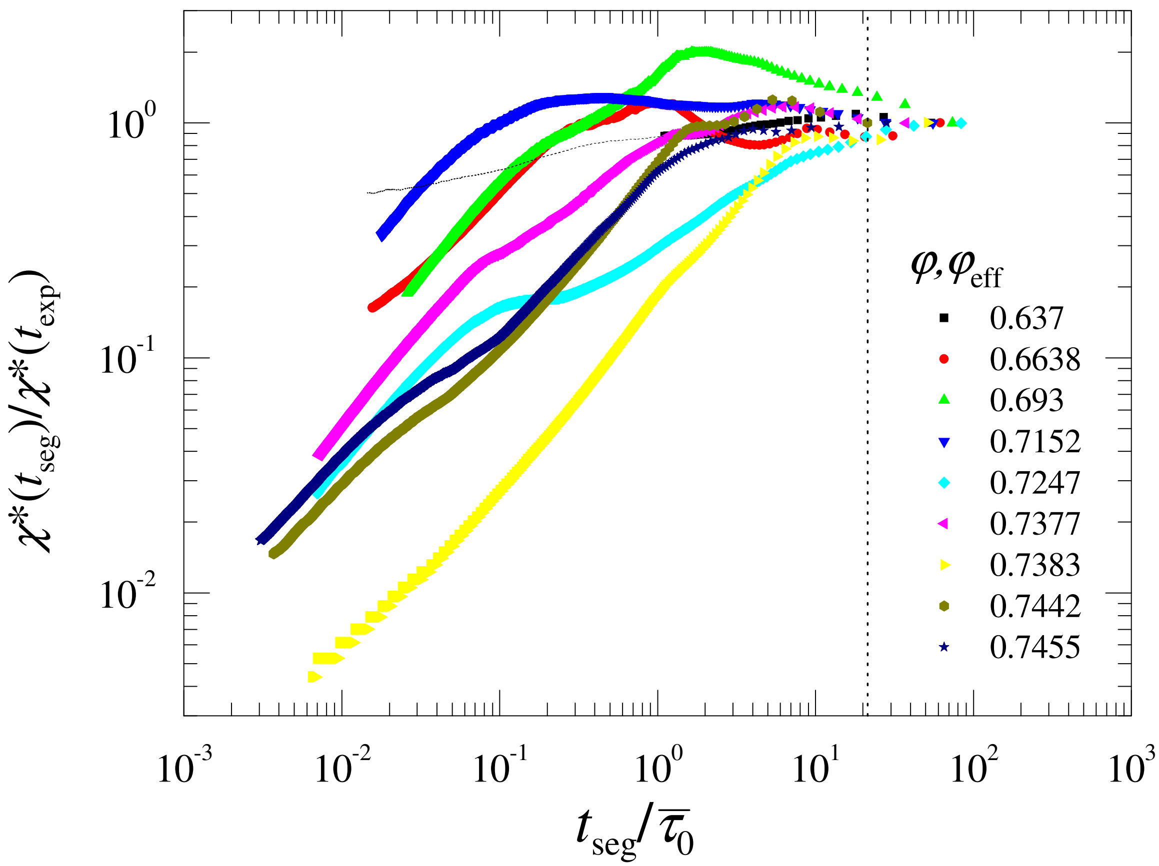


Measurement time issue?



Does $\chi^*(t_{\mathrm{seg}},\varphi)$ depend on t_{seg} ?

Not a measurement time issue !



Proposed physical mechanism

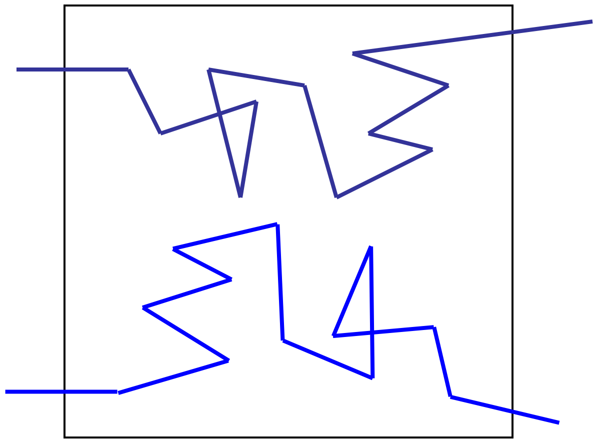
Competition between :

Growth of ξ on approaching φ_g ⟶ N_{blob} ↘ χ^* ↗

Smaller displacement associated with each rearrangement event (tigther packing) ⟶ More events required to relax system χ^* ↘

DWS and intermittent dynamics

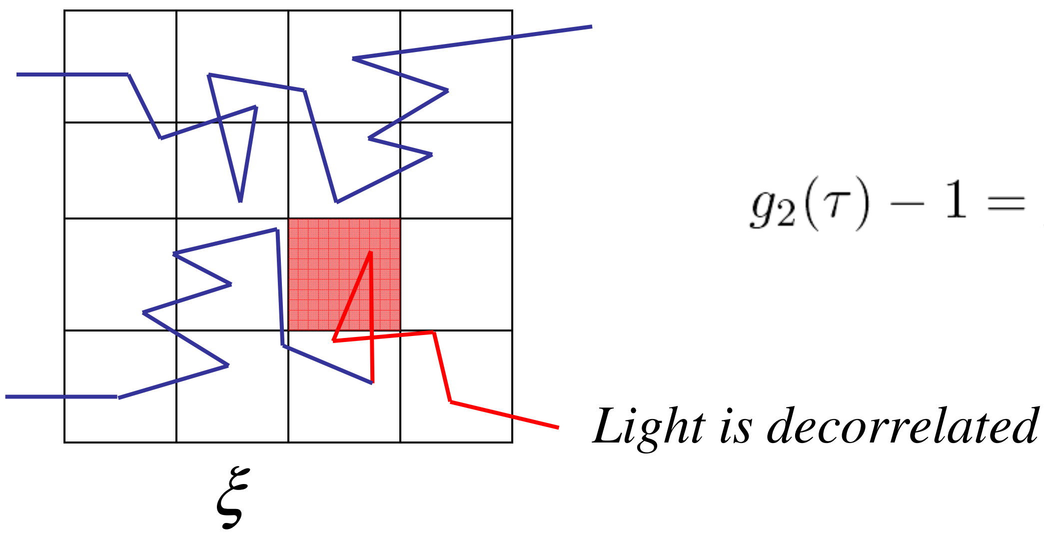
Inspired by Durian, Weitz & Pine (Science, 1991)



$$g_2(\tau) - 1 = g_1^2(\tau) = \left[\sum_s g_1^{(s)}(\tau)\right]^2$$

DWS and intermittent dynamics

Inspired by Durian, Weitz & Pine (Science, 1991)

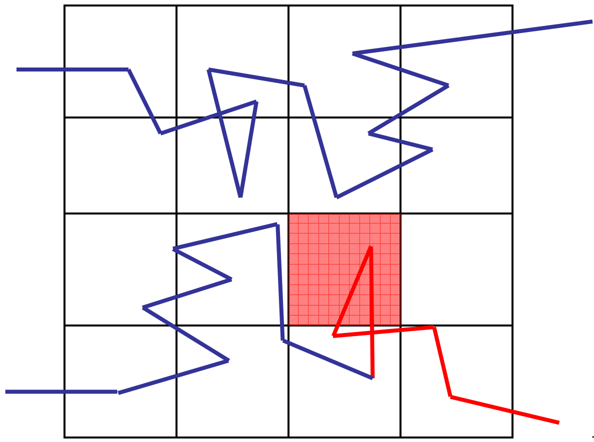


$$g_2(\tau) - 1 = g_1^2(\tau) = \left[\sum_s g_1^{(s)}(\tau)\right]^2$$

$$g_1^{(s)}(t,\tau) = \exp[-n_s(t,\tau)^p \sigma_\phi^2]$$

DWS and intermittent dynamics

Inspired by Durian, Weitz & Pine (Science, 1991)



ξ

$$g_2(\tau) - 1 = g_1^2(\tau) = \left[\sum_s g_1^{(s)}(\tau)\right]^2$$

Light is decorrelated

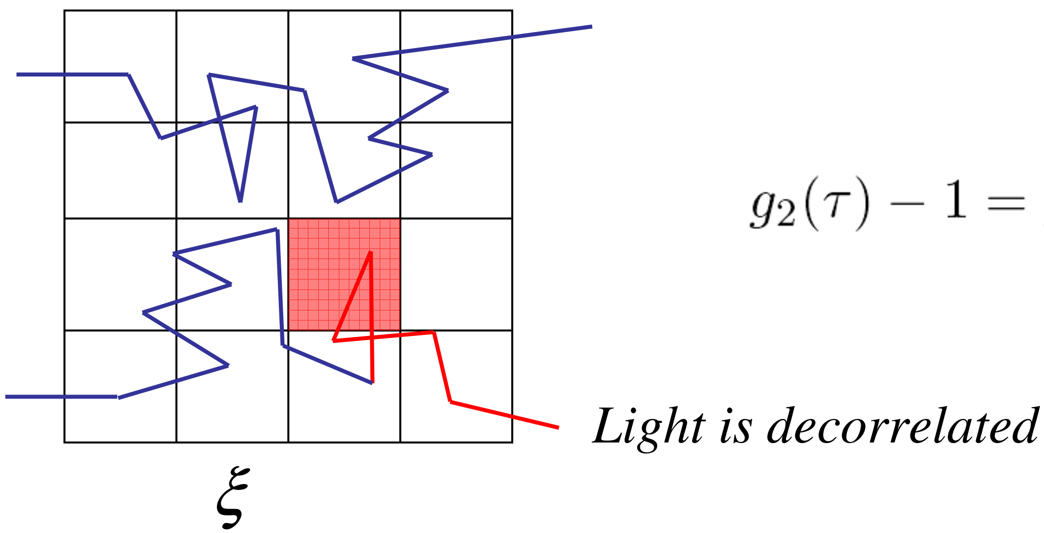
$$g_1^{(s)}(t,\tau) = \exp[-n_s(t,\tau)^p \sigma_\phi^2]$$

Number of events between
t and $t+\tau$

Mean squared change of phase for
1 event ~ Δr^2

DWS and intermittent dynamics

Inspired by Durian, Weitz & Pine (Science, 1991)



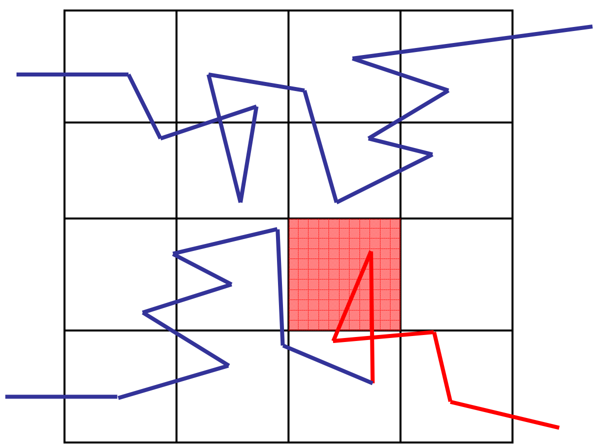
$$g_2(\tau) - 1 = g_1^2(\tau) = \left[\sum_s g_1^{(s)}(\tau)\right]^2$$

$$g_1^{(s)}(t,\tau) = \exp[-n_s(t,\tau)^p \sigma_\phi^2]$$

$p = 1$ « **brownian** » rearrangements

$p = 2$ « **ballistic** » rearrangements

Simulations



- Photon paths as random walks on a 3D cubic lattice
- Lattice parameter = l^*, match cell dimensions
- Random rearrangement events of size ξ^3

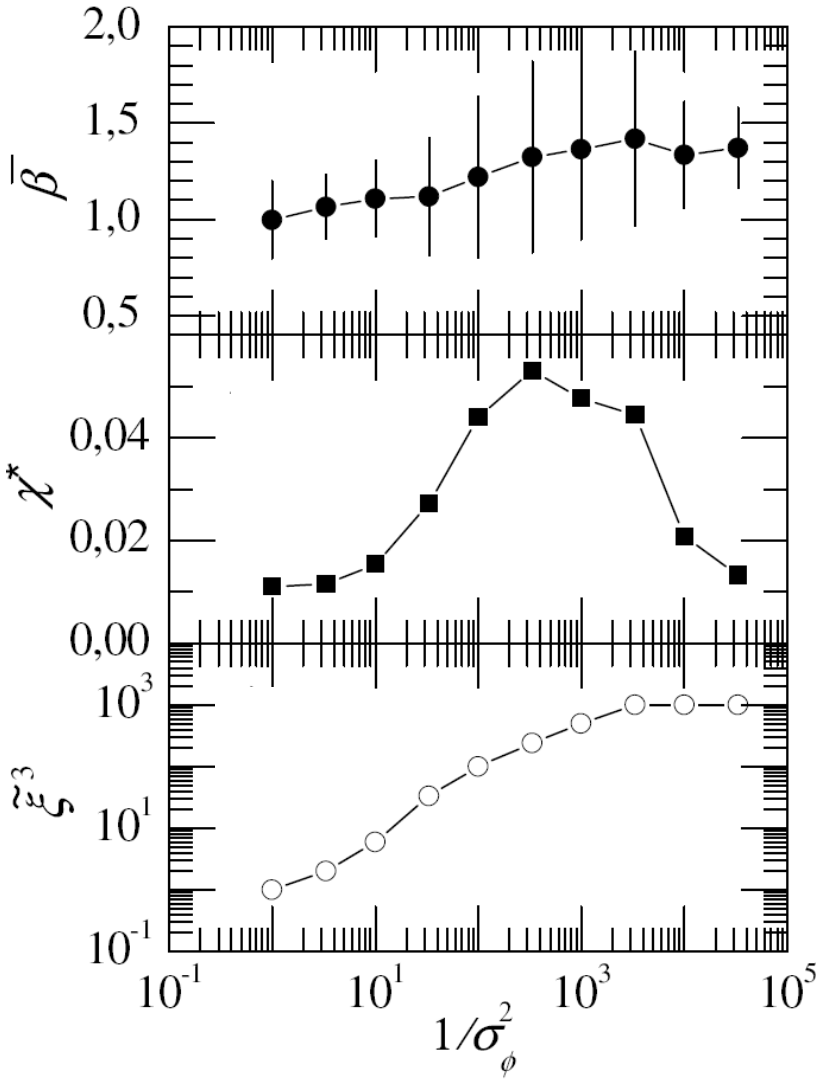
- Calculate $c_I(t,\tau) = \left[\sum_s g_1^{(s)}(t,\tau)\right]^2$ with

$$g_1^{(s)}(t,\tau) = \exp[-n_s(t,\tau)^p \sigma_\phi^2]$$

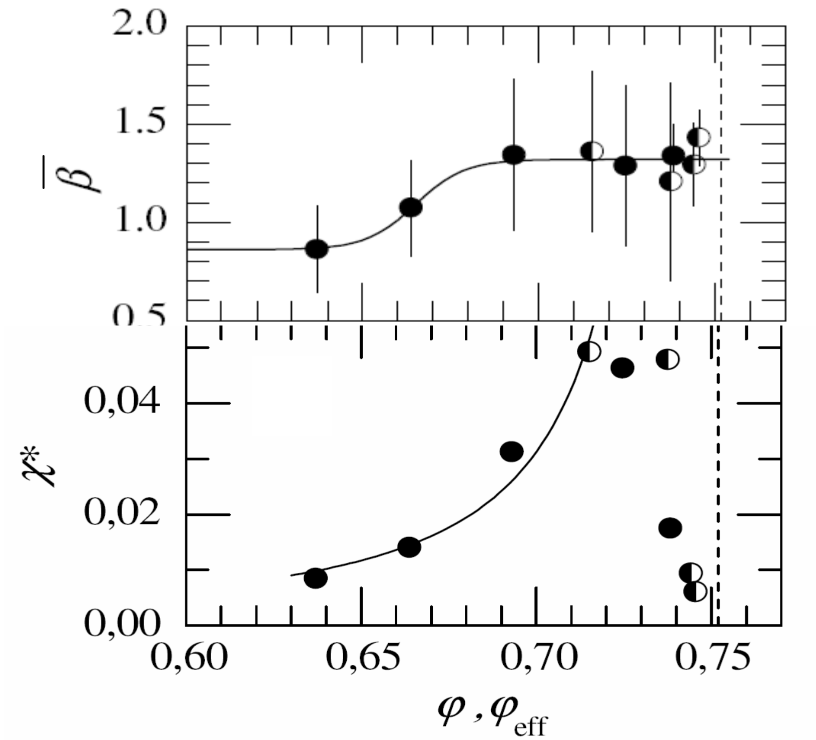
Parameters :

- $\boldsymbol{p}$ (use one single p for all φ)
- ξ^3
- σ^2_ϕ (we expect $\sigma^2_\phi \searrow$ as $\varphi \longrightarrow \varphi_c$)

Simulations *vs.* experiments

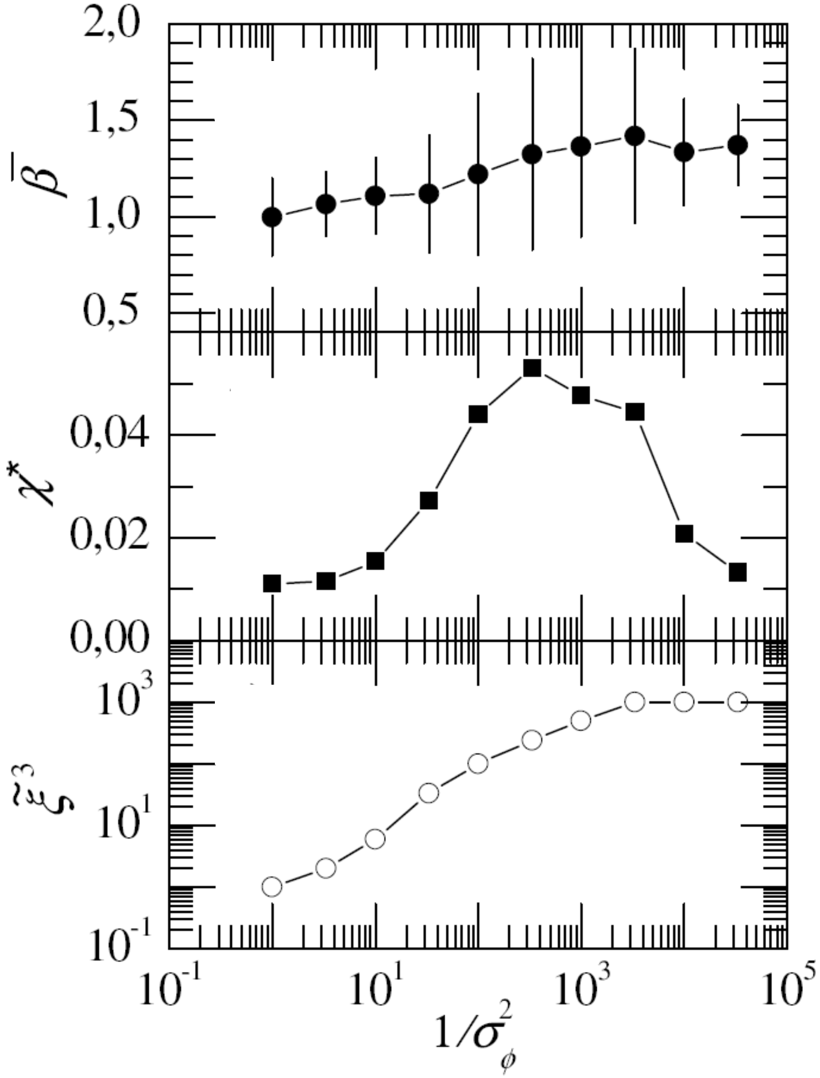


simulations



experiments

Simulation parameters



$p = 1.65$ **supradiffusive motion**

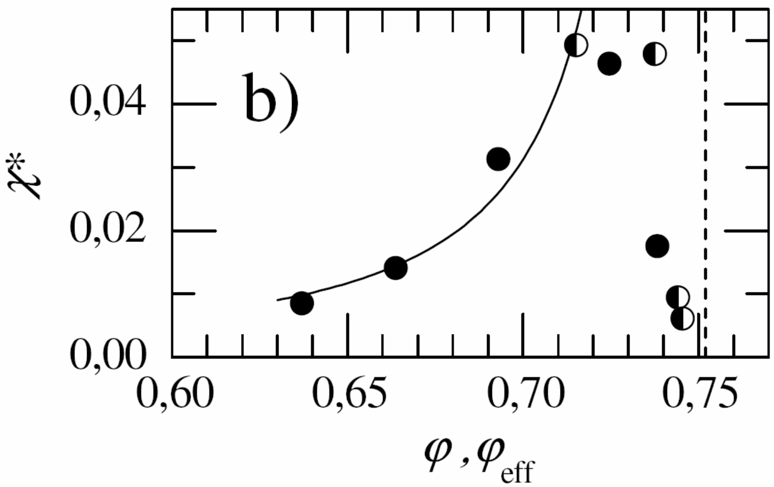
ξ^3 **- grows continuously with φ**

- very large!!

Conclusions

Dynamics **heterogeneous**

Non-monotonic behavior of χ^*

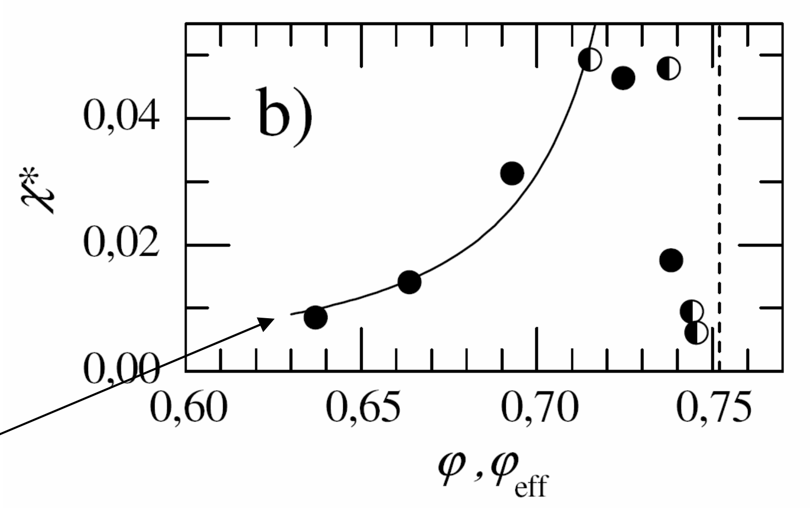


Conclusions

Dynamics **heterogeneous**

Non-monotonic behavior of χ^*

Competition between
- **increasing size** of dynamically
  correlated regions



Conclusions

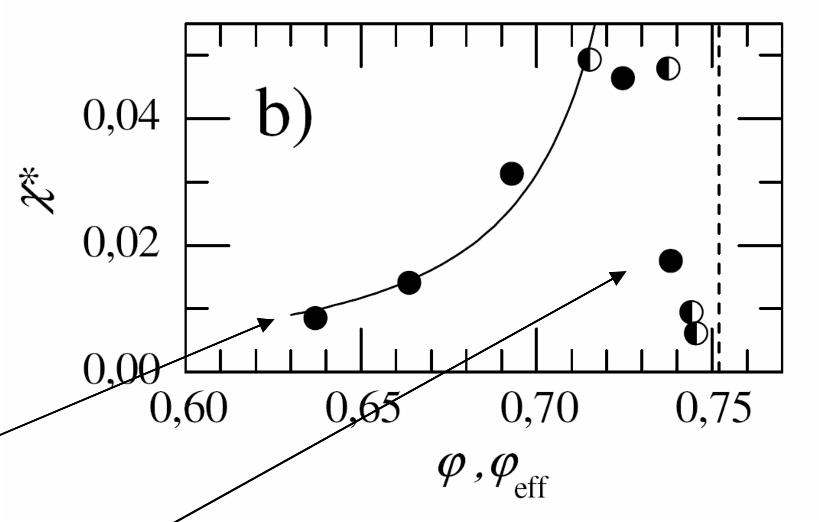
Dynamics **heterogeneous**

Non-monotonic behavior of χ^*

Competition between

- **increasing size** of dynamically correlated regions

- **decreasing effectiveness** of rearrangements



Dynamical heterogeneity dictated by the **number of rearrangements** needed to relax the system

Thanks to…

V. Trappe	CNES
D. Weitz	Softcomp
L. Berthier	ACI
G. Biroli	IUF
M. Cloître	