Probing dynamical heterogeneity by XPCS

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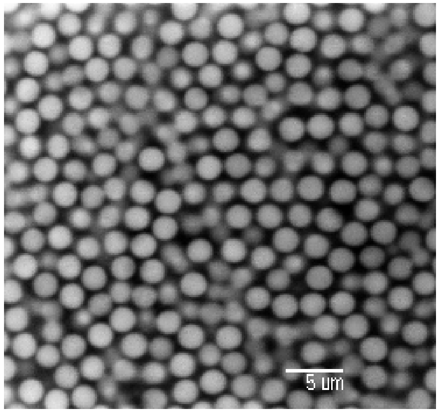
Soft glassy/jammed materials



Overview

- Slow dynamics and dynamical heterogeneities in soft glasses

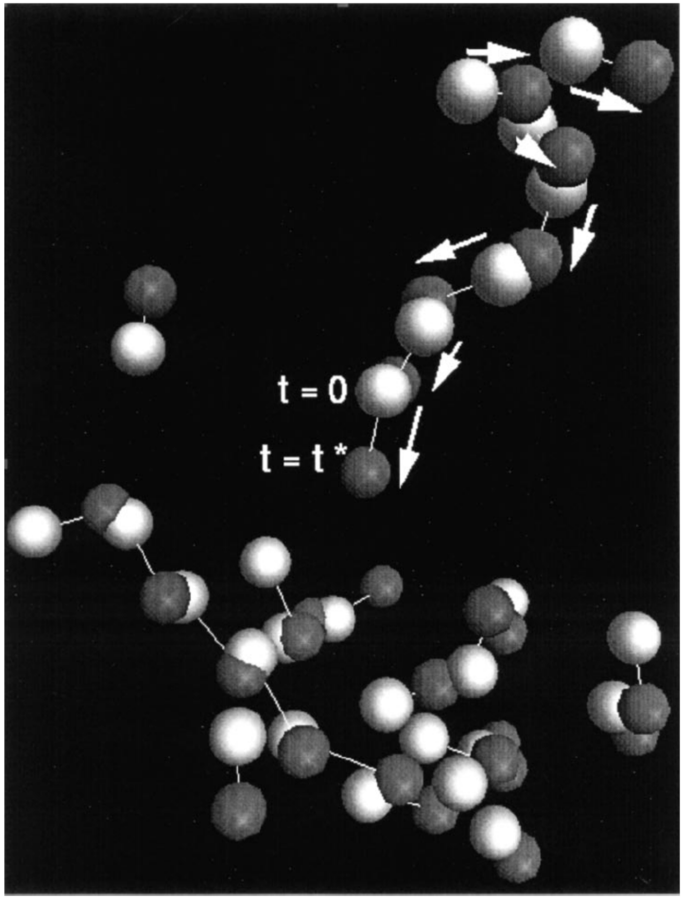
- How to probe DH: Time Resolved Correlation (TRC)

- XPCS measurement of DH in a gel system

- Bringing together Imaging and Scattering

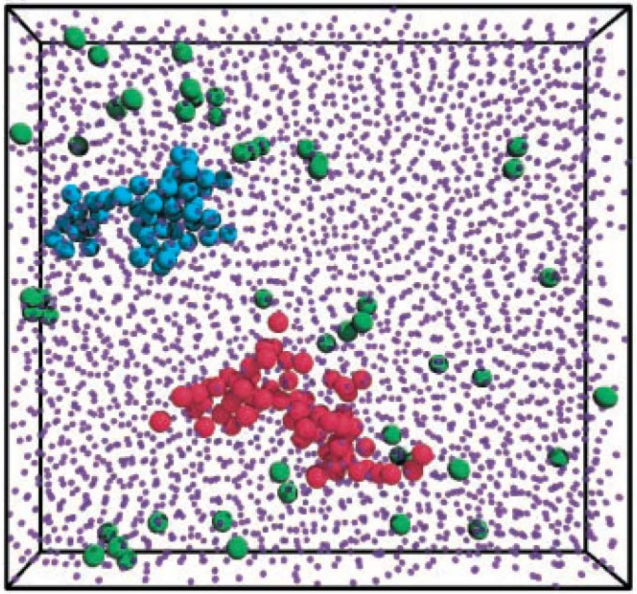
Dynamical Heterogeneity

LJ supercooled fluid



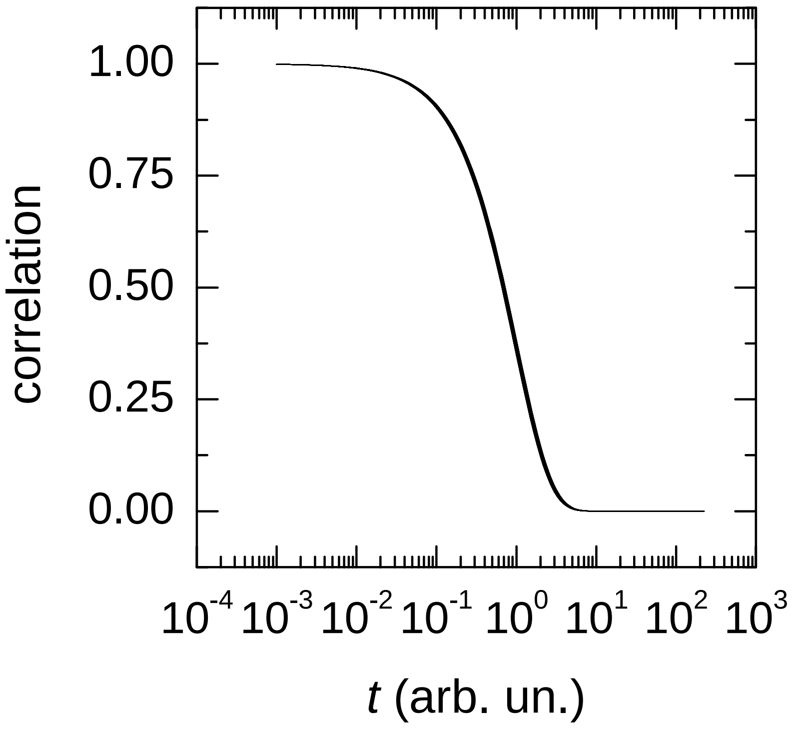
Donati et al. PRL 98

Supercooled colloidal HS



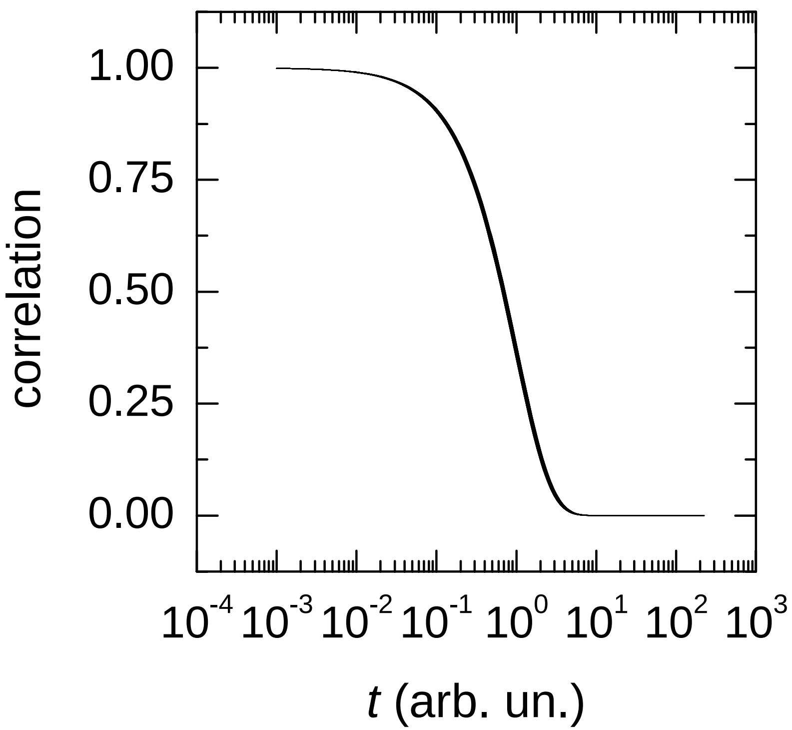
Weeks et al. Science 2000

Temporally heterogeneous dynamics

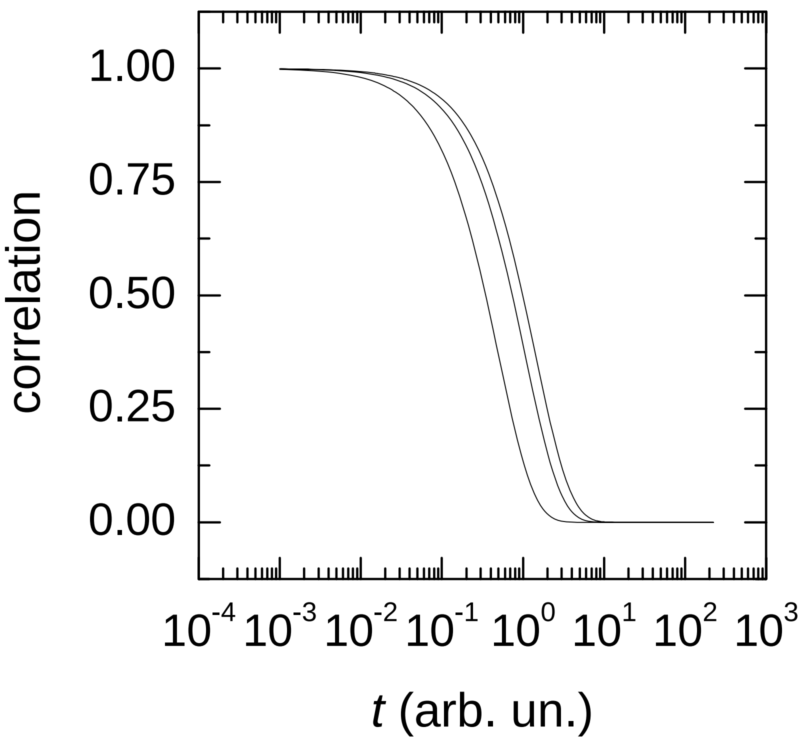


homogeneous

Temporally heterogeneous dynamics

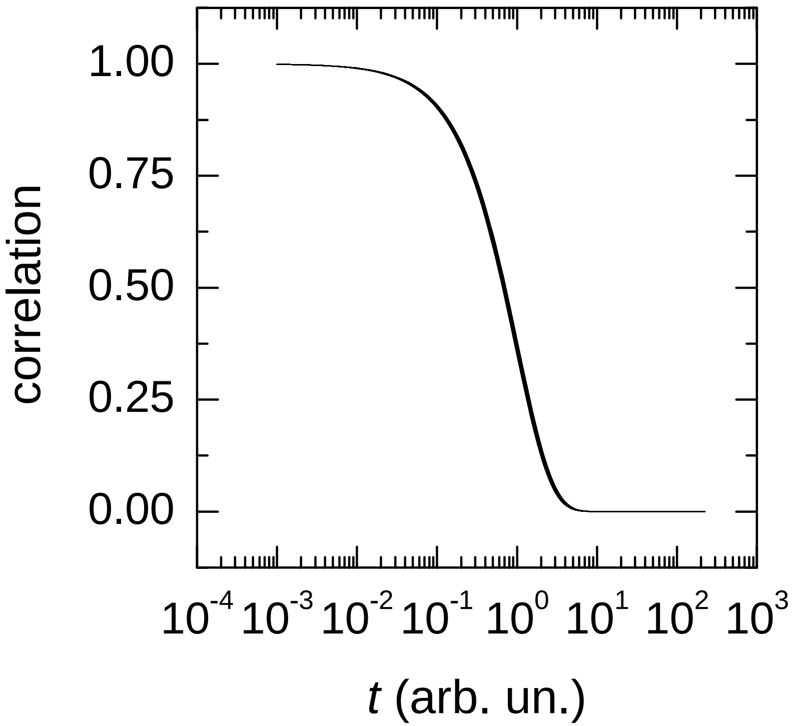


homogeneous

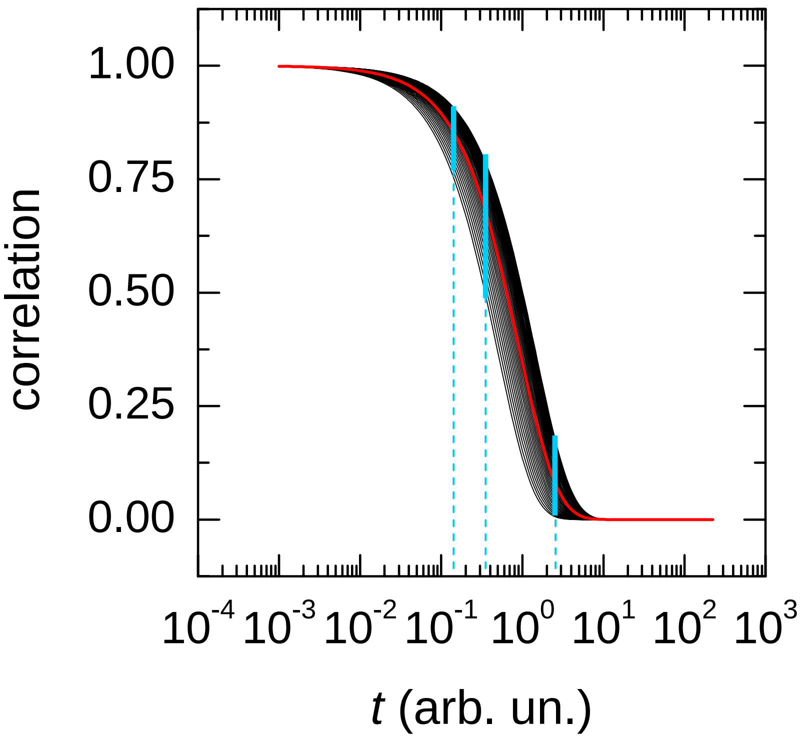


heterogeneous

Temporally heterogeneous dynamics



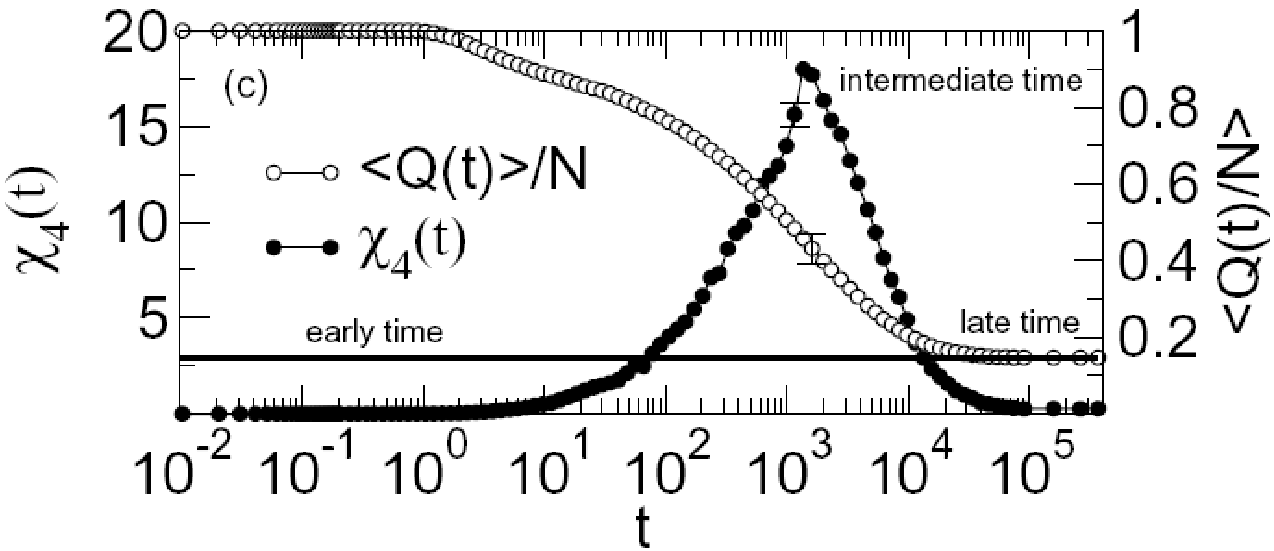
homogeneous



heterogeneous

Dynamical susceptibility in glassy systems

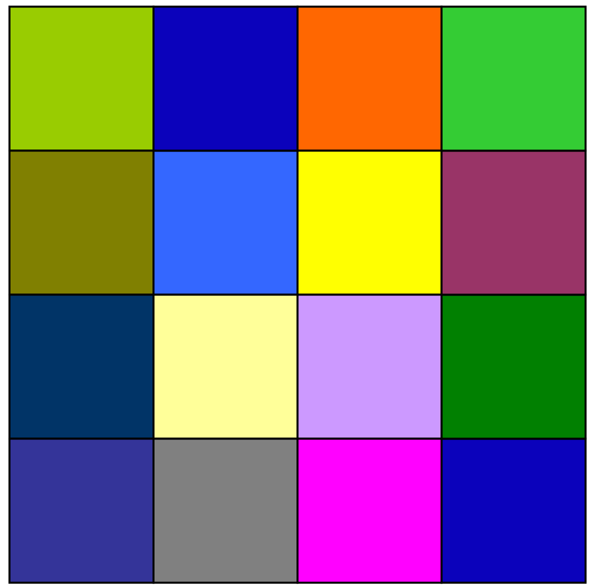
Supercooled liquid (Lennard-Jones)



Lacevic et al., PRE 2002

$\chi_4 \sim \mathrm{var}[Q(t)]$

Dynamical susceptibility in glassy systems

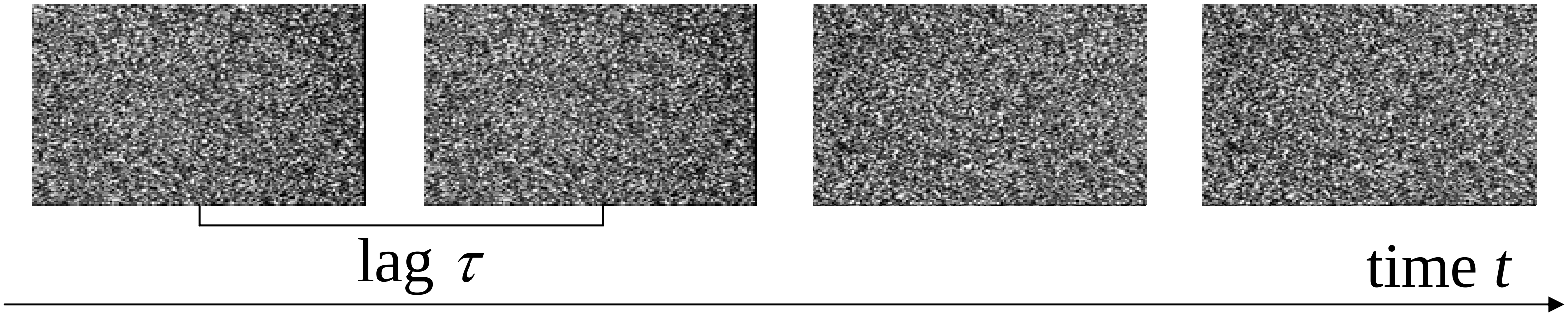


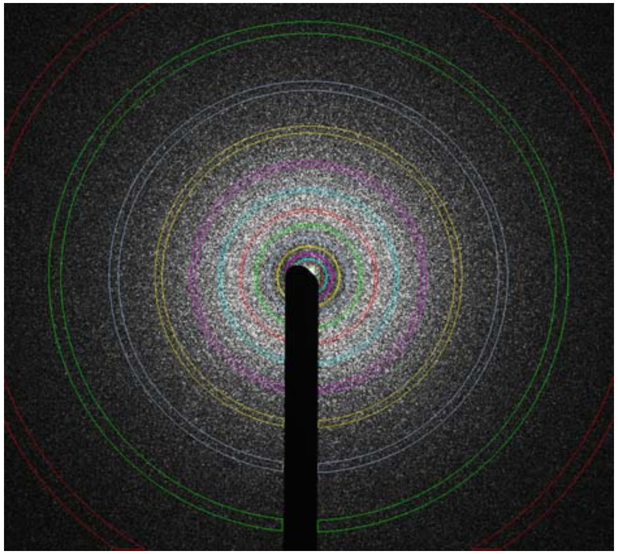
N_{blob} regions

$$\chi_4 \sim \mathrm{var}[Q(t)] \sim 1/N_{\mathrm{blob}}$$

Fluctuations of the dynamics: Time Resolved Correlation

[Cipelletti et al. J. Phys:Condens. Matter 2003],
[Duri et al. PRE 2006]





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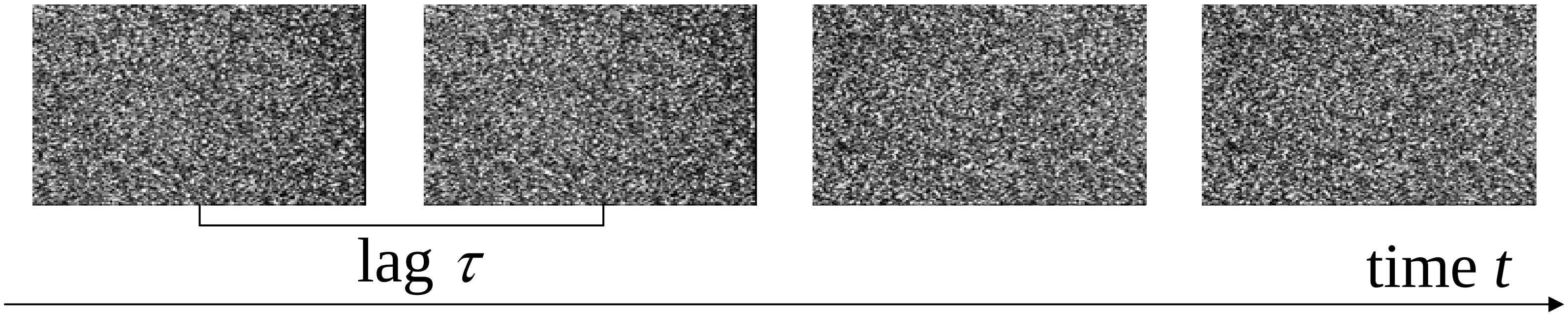

degree of correlation $c_I(t,\tau) = \frac{< I_p(t)\, I_p(t+\tau) >_p}{< I_p(t) >_p < I_p(t+\tau) >_p} - 1$

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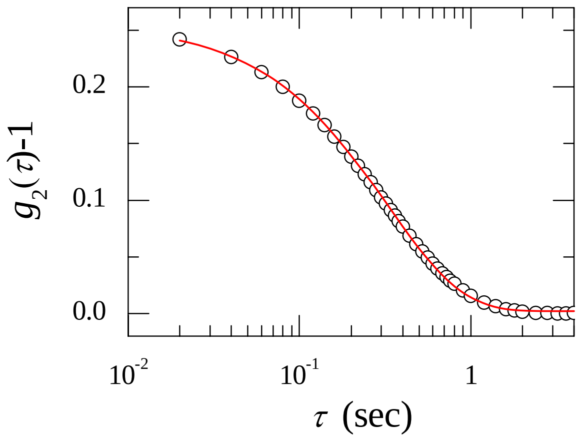
Average over t ⬇

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



$g_2(\tau) - 1$ ➡ Average dynamics

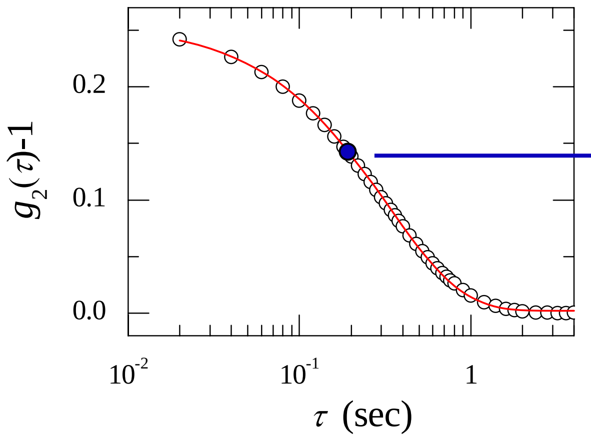
degree of correlation $c_I(t,\tau) = \dfrac{< I_p(t)\, I_p(t+\tau)>_p}{< I_p(t)>_p < I_p(t+\tau)>_p} - 1$

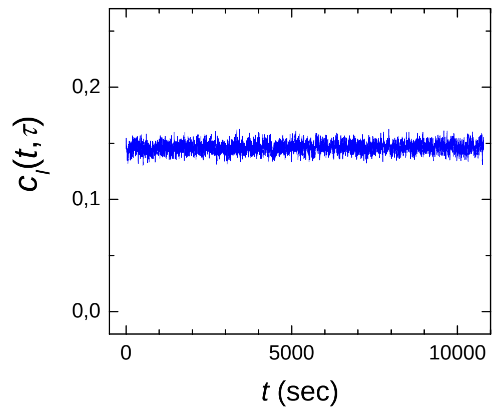
Average over *t*

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

fixed τ, *vs. t*

fluctuations of the dynamics





$g_2(\tau) - 1$ → Average dynamics

Brownian particles

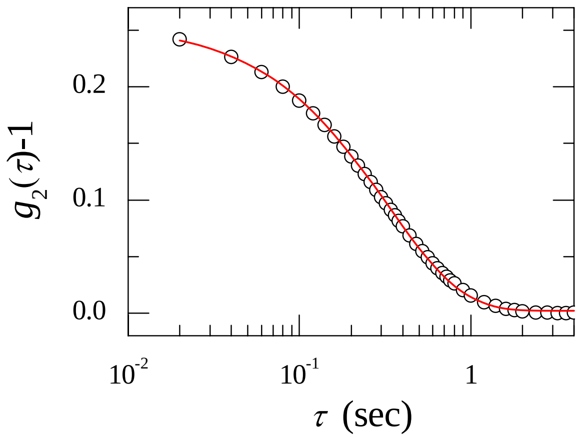
degree of correlation $c_I(t_w,\tau) = \dfrac{< I_p(t_w)\, I_p(t_w+\tau)>_p}{< I_p(t_w)>_p < I_p(t_w+\tau)>_p} - 1$

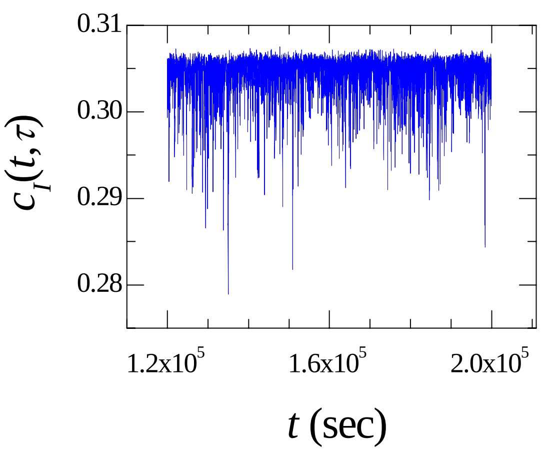
Average over t_w

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

fixed τ, *vs.* t_w

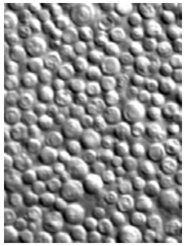
fluctuations of the dynamics





$g_2(\tau) - 1$ → Average dynamics

Onion gel



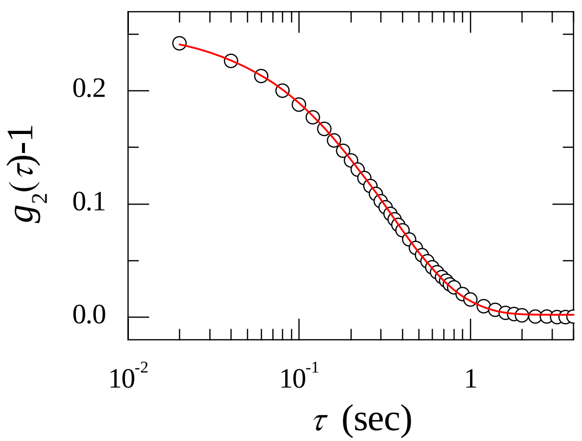
degree of correlation $c_I(t_w,\tau) = \dfrac{< I_p(t_w)\, I_p(t_w+\tau)>_p}{< I_p(t_w)>_p < I_p(t_w+\tau)>_p} - 1$

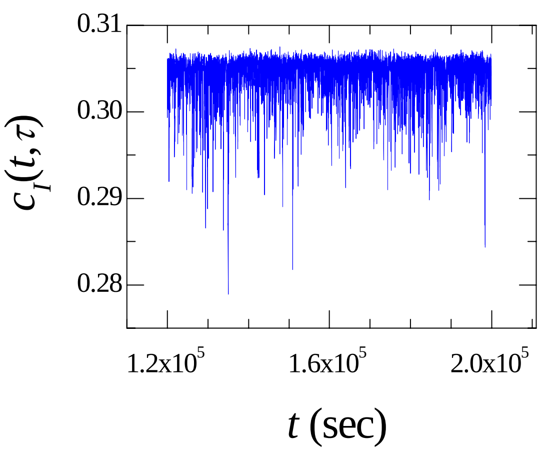
Average over t_w

fixed τ, *vs.* t_w

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

fluctuations of the dynamics





$g_2(\tau) - 1$ → Average dynamics

var(c_I) -> χ_4

XPCS measurements of the dynamics of a Carbon Black gel

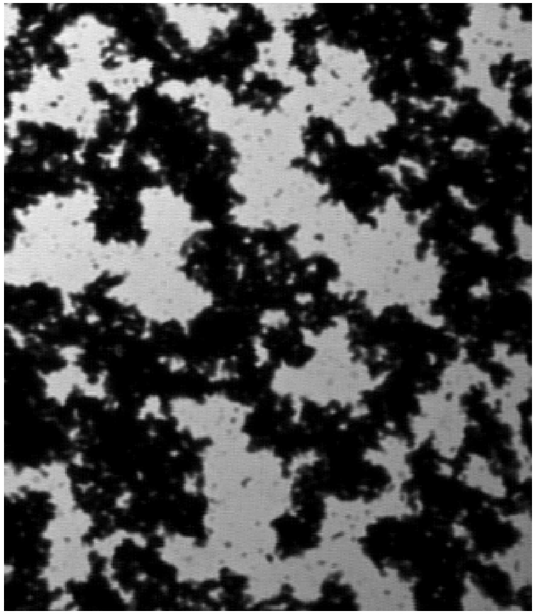
[Trappe et al., PRE 2007]

• Particle size R= 180 nm

• Suspended in mineral oil at $\varphi = 6\%$

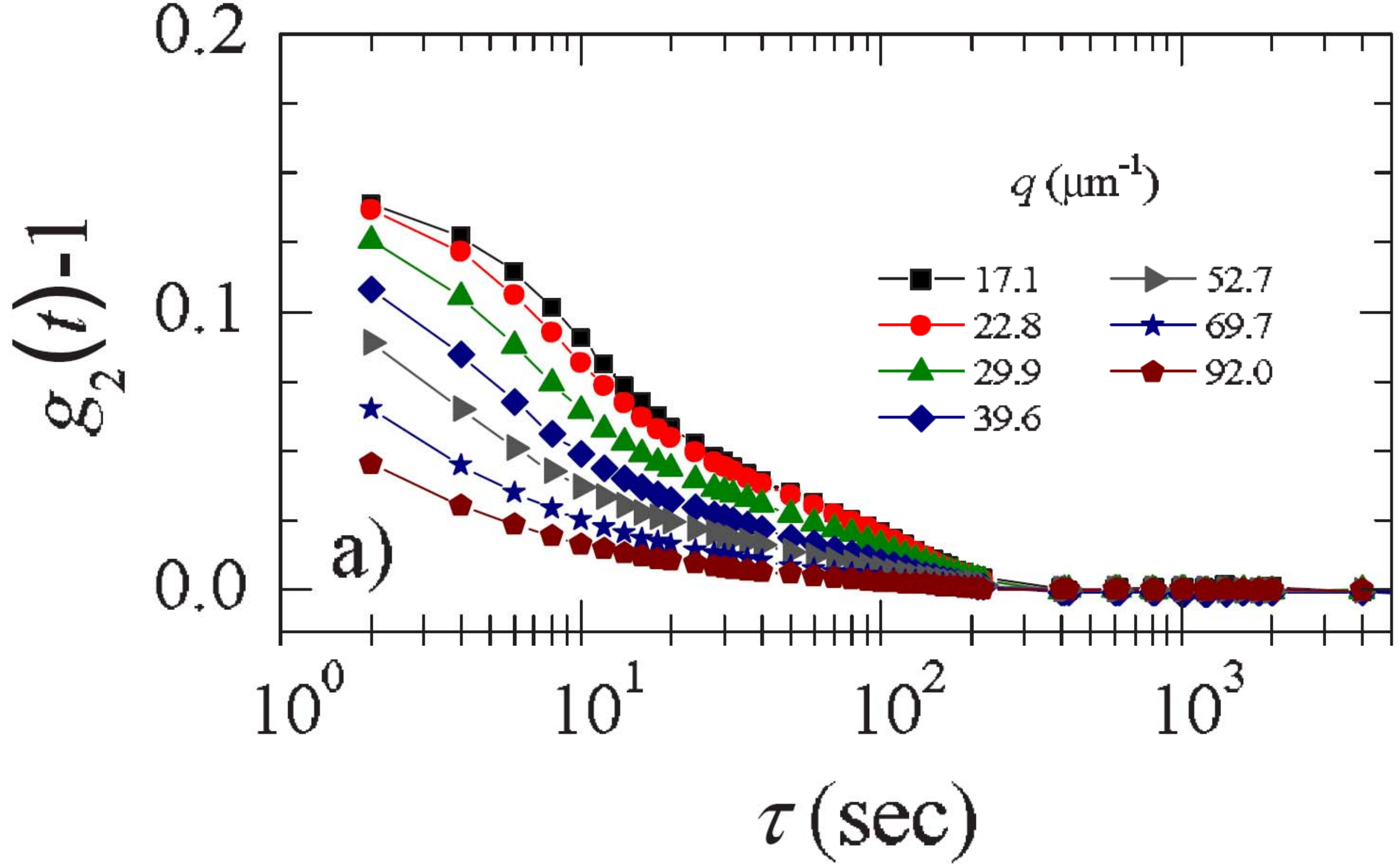
• Attractive interactions controlled by adding a dispersant:
$U \sim 12\ k_B T$ and $U \sim 30\ k_B T$

• XPCS @ ID10 Troika beamline (ESRF)



Average dynamics

$U \sim 12\ k_B T$

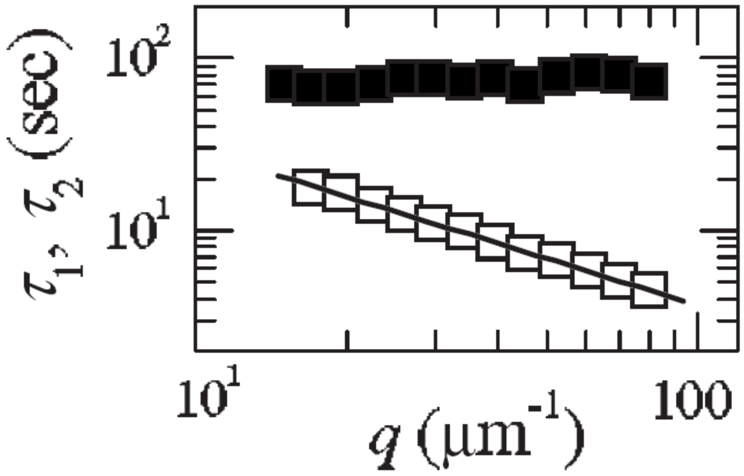


Average dynamics

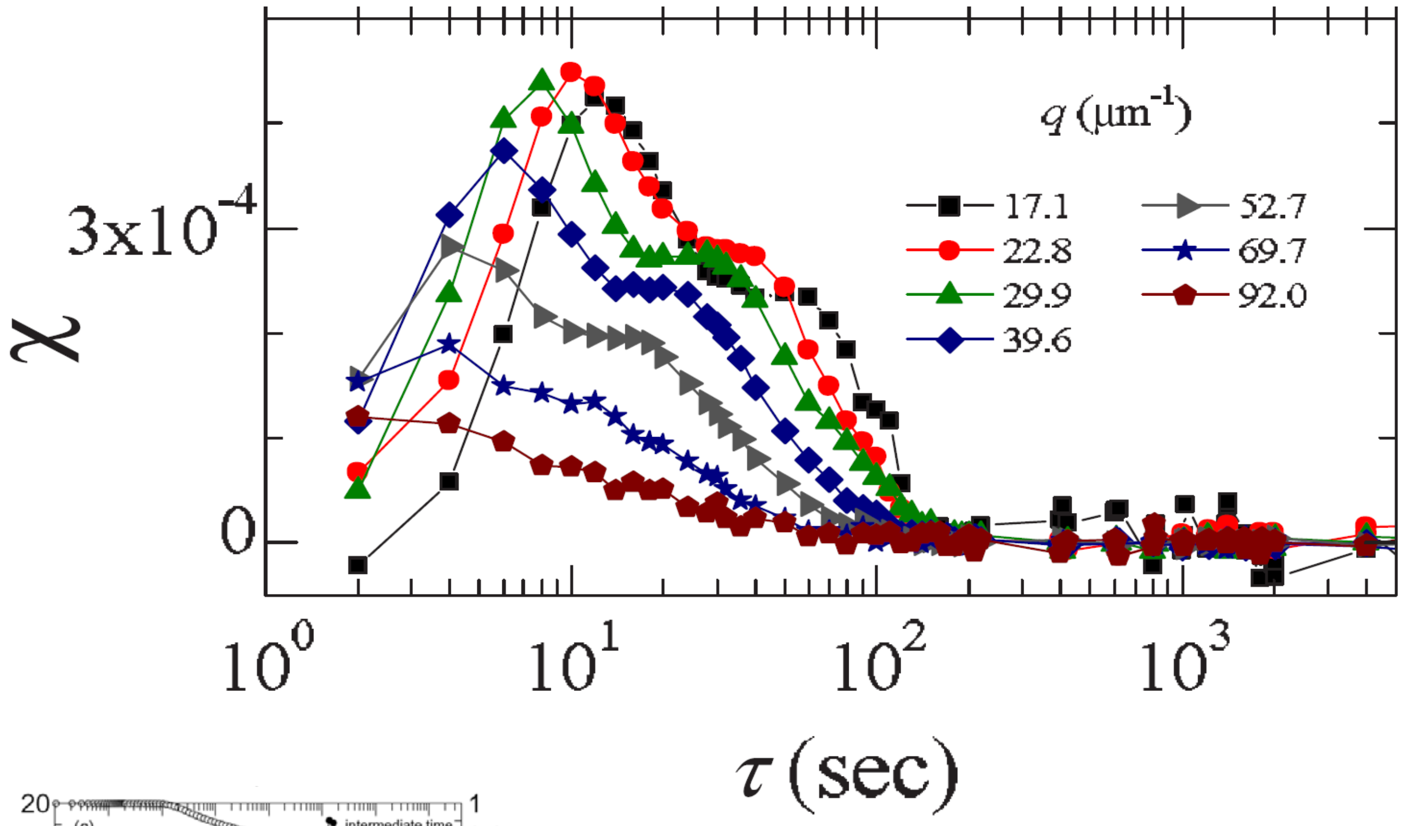
Fit : $A\exp[-(\tau/\tau_i)^p]$

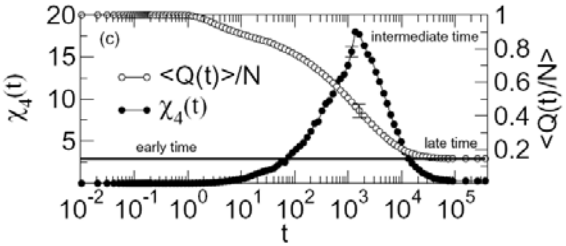
Initial decay: $\tau_1 \sim q^{-0.91}$, $p = 1.2$
Stress-relaxation driven dynamics

Final decay: $\tau_2 \sim q^0$, $p = 1$
Bond breaking?
(no final decay for the gel with $U \sim 30\ k_B T$)

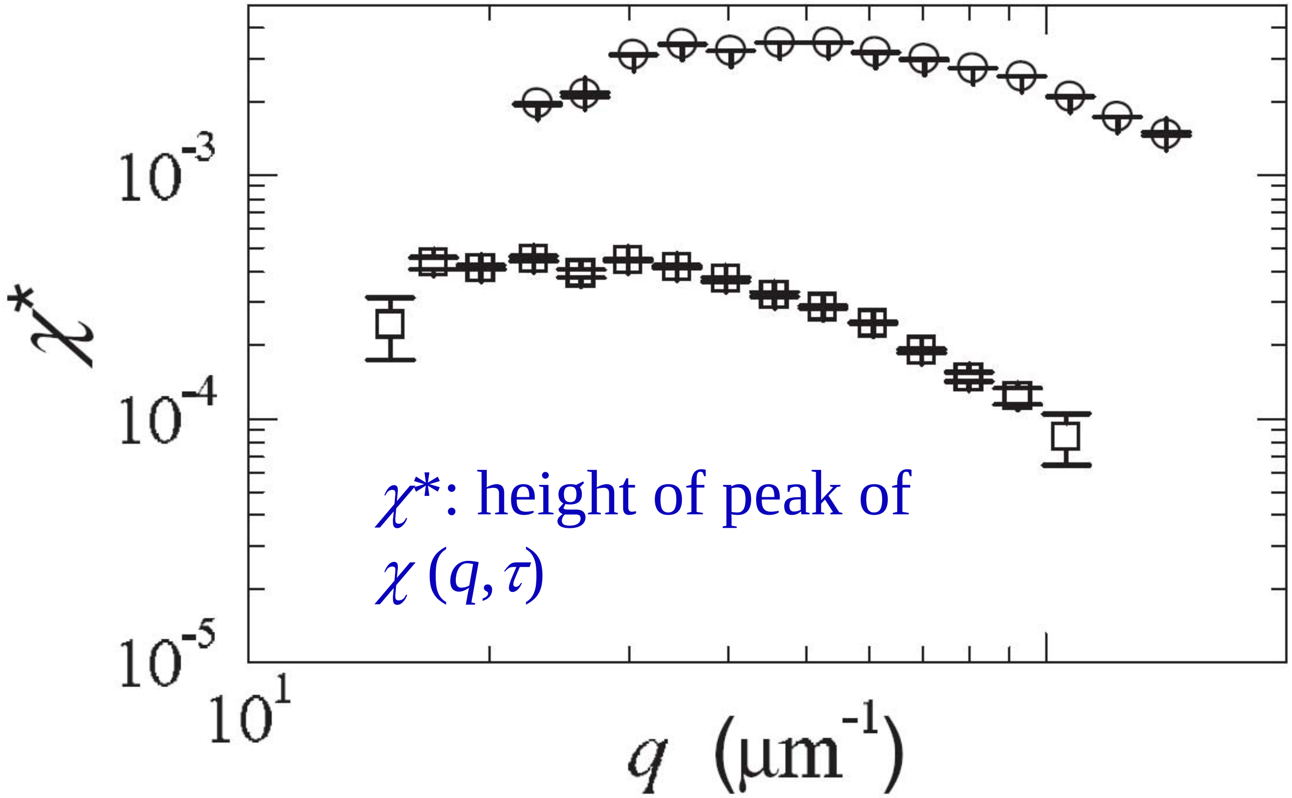


Fluctuations of the dynamics: dynamical susceptibility $\chi = \mathrm{var}[c_I]$





q-dependence of the amplitude of χ

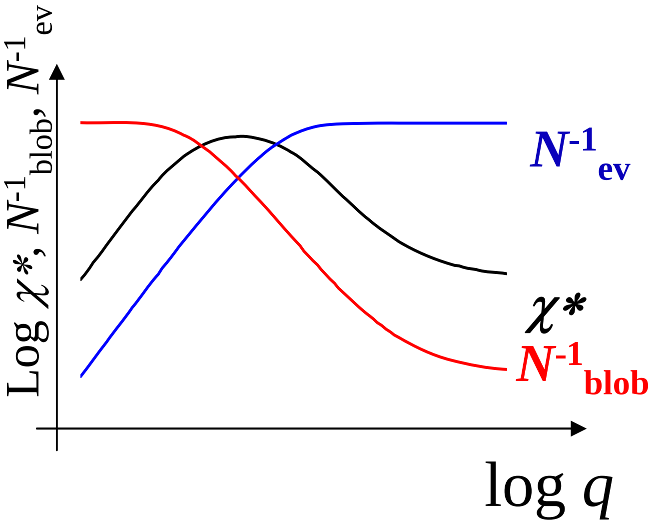


Non-monotonic behavior (see also Charbonneau & Reichman, PRL 2007)

q-dependence of χ*: a simple argument

χ* ~ (# rearrangements in the scattering volume needed to decorrelate the scattered light)$^{-1}$

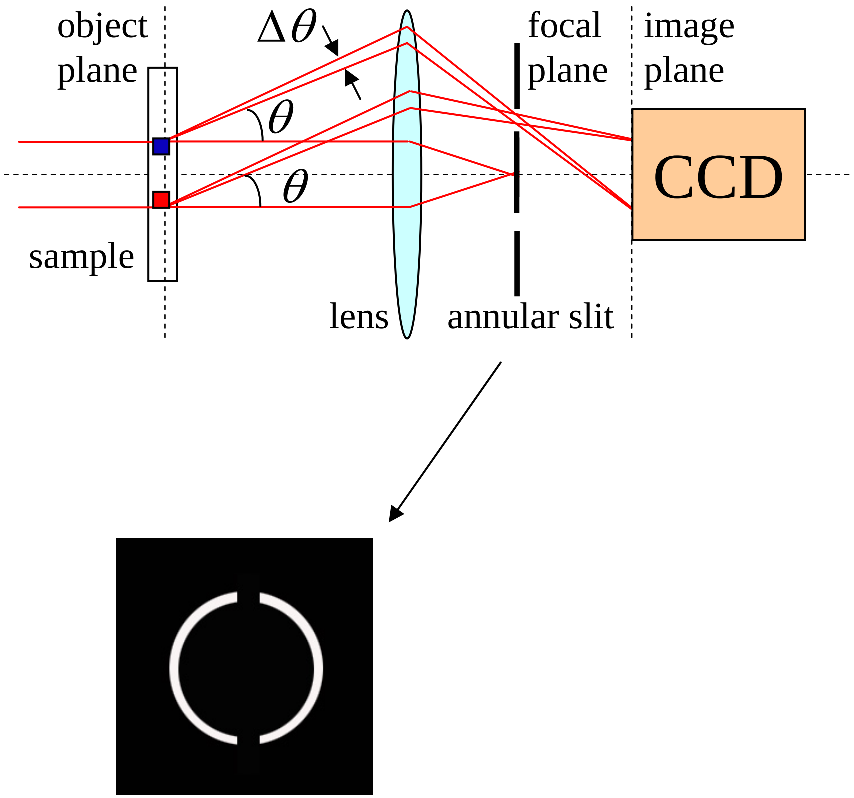
$$\chi^* \sim (N_{\mathrm{blob}} N_{\mathrm{ev}})^{-1}$$



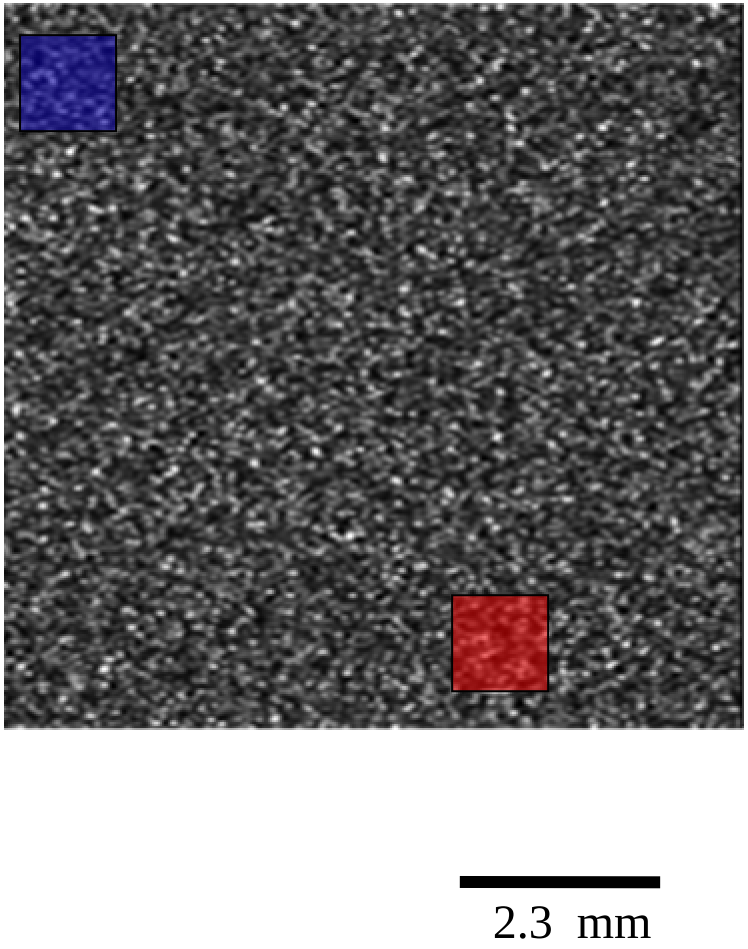
N_{blob} , N_{ev} depend on q, t_w, φ, …

Photon Correlation Imaging (PCIm)

[Duri et al., PRL 2009]

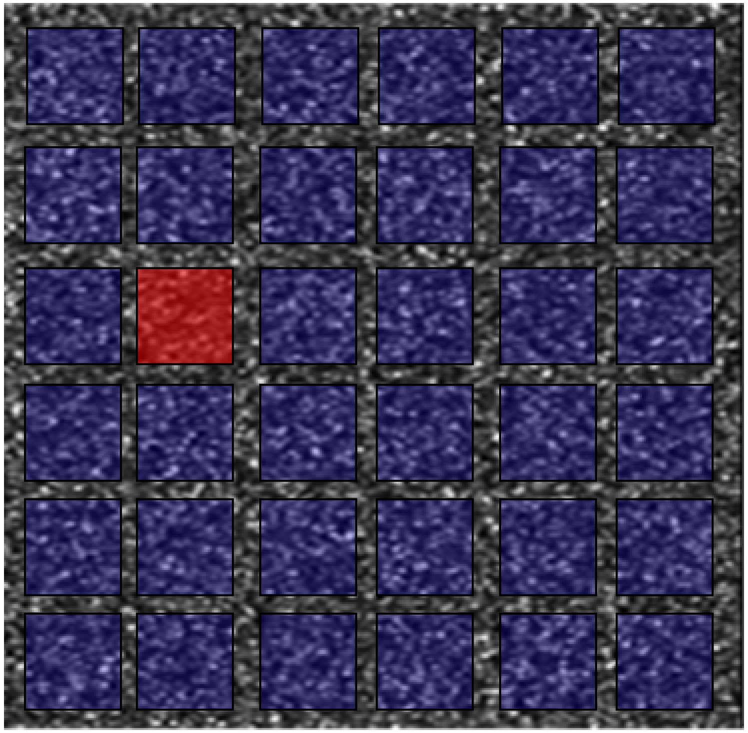


$\theta = 6.4°$ $\longrightarrow$ $q = 1\ \mu m^{-1}$.



Local, instantaneous dynamics: $c_I(t, \tau, \mathrm{r})$

$$c_I(t, \tau, \mathbf{r}) = \frac{< I_p(t)\, I_p(t+\tau)>_{p(\mathbf{r})}}{< I_p(t)>_p < I_p(t+\tau)>_{p(\mathbf{r})}} - 1$$

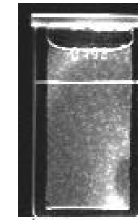


2.3 mm

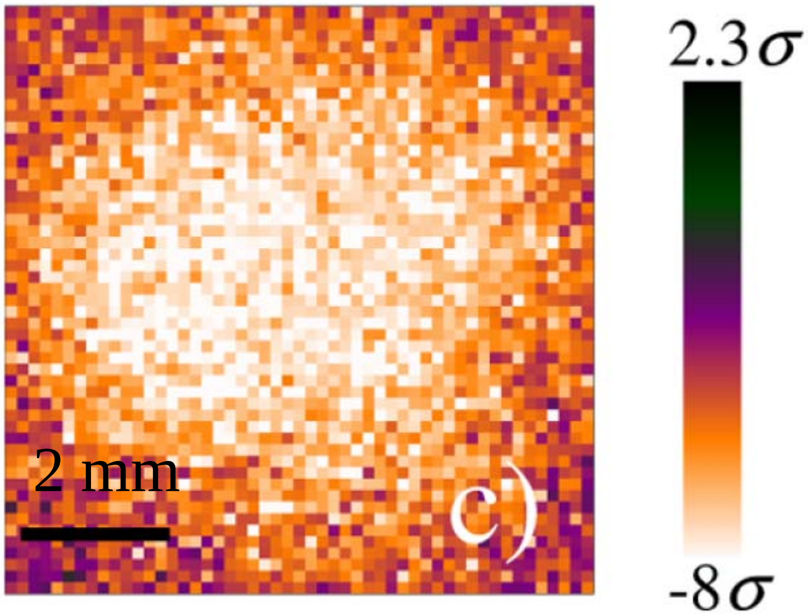
Note: $<< c_I(t, \tau, \mathbf{r}) >_t>_{\mathbf{r}} = g_2(\tau)-1$

$[g_2(\tau)-1]^{1/2}$ ~ dynamic structure factor

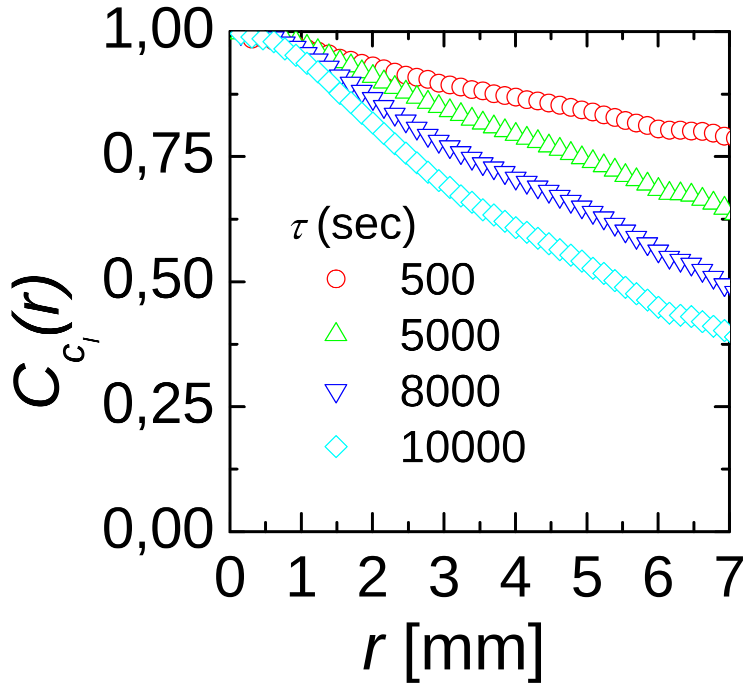
Strongly attractive colloidal gel (PS)



Dynamical activity map



τ = 500 sec
(~ τ_r/10)



Ultra long-ranged correlations!
(particle size: 20 nm
cluster size: ~20 μm)

Conclusions

- **Scattering techniques can measure instantaneous/spatially resolved dynamics**

- **Slow dynamics of systems jammed/close to jamming are heterogeneous**

- **Spatial correlations can be very long ranged!**

- **A PCI setup for XPCS?**

Thanks to…

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EU (SoftComp, Arrested Matter)
IUF

Size of a rearrangment: space-resolved TRC