Intermittent slow dynamics in jammed soft matter

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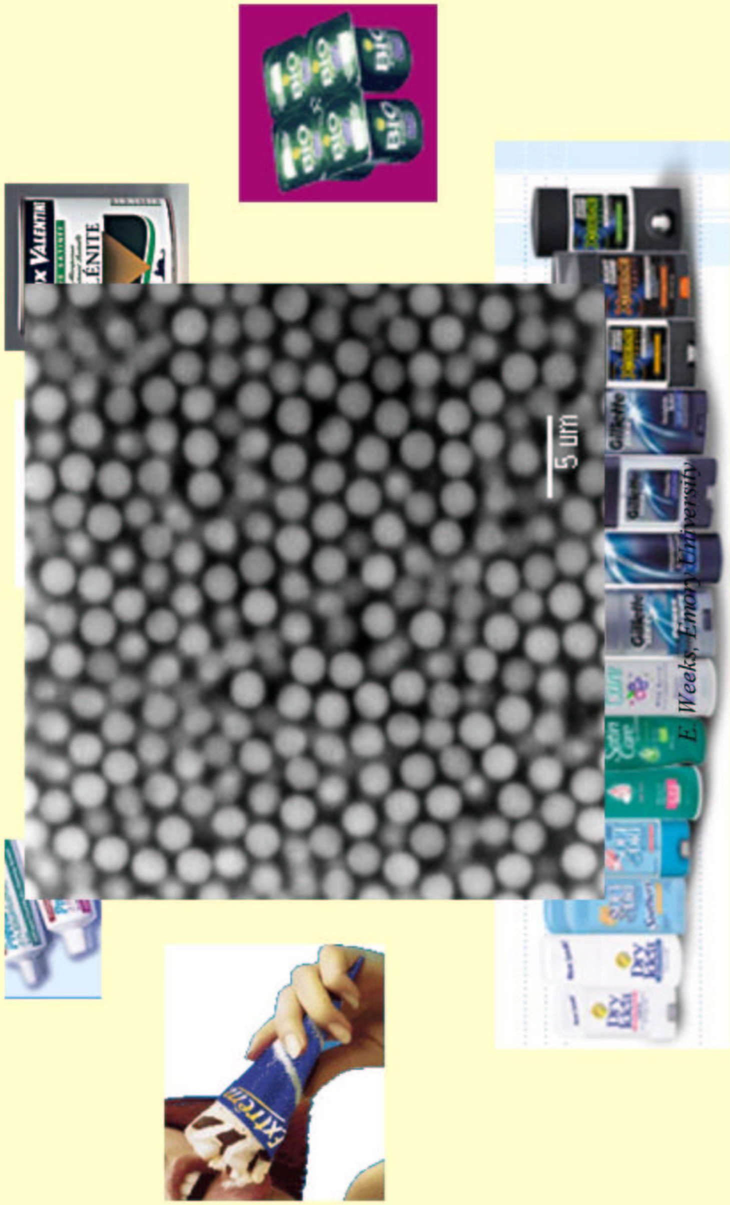
H. Bissig		UM2

Soft Glassy Materials

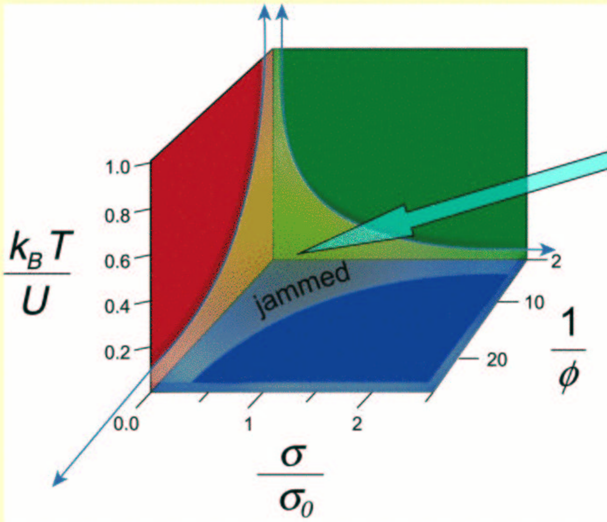


Outline

- The **jamming** phase transition
- Slow dynamics: role of **internal stress** relaxation
- **Intermittent** slow dynamics
- Is temporal heterogeneity **universal**?

Jamming and slow dynamics

Jamming Phase Diagram
Liu and Nagel, Nature 1998 **Repulsive Interactions**
Trappe et al, Nature 2001 **Attractive Colloids**



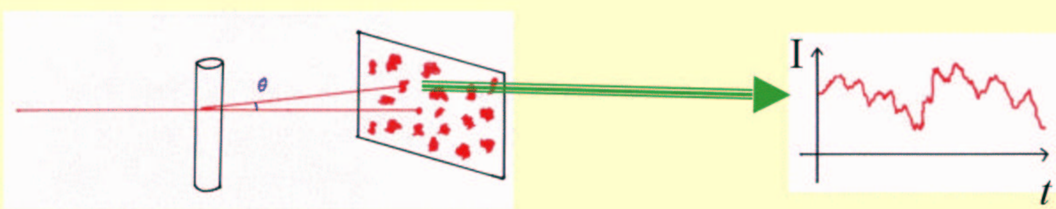
Ultraslow relaxations

Aging

Heterogeneous dynamics

????

Dynamic Light Scattering



$q = 4\pi n/\lambda \sin(\theta/2)$ $\xi \sim 1/q$

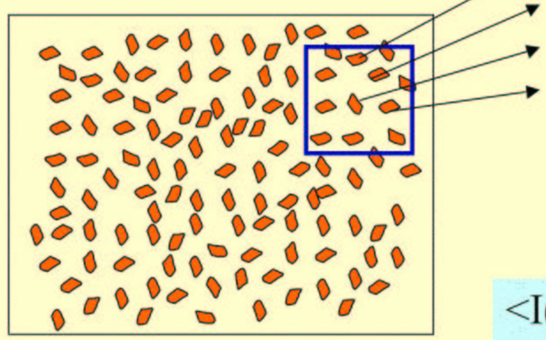
- we *measure* : $< I(t)I(t+\tau) >_t$

Dynamic Structure Factor

$$f(q,\tau) = \left\langle \frac{1}{N} \sum_j \exp\left[i\mathbf{q} \cdot \Delta\mathbf{r}_j(\tau)\right] \right\rangle$$

The Multispeckle technique

Average $g_2(t)$ measured **in parallel** for many "statiscally equivalent" **speckles**



$I_1(q,t)I_1(q,t+\tau)$
$I_2(q,t)I_2(q,t+\tau)$
$I_3(q,t)I_3(q,t+\tau)$
$I_4(q,t)I_4(q,t+\tau)$
...

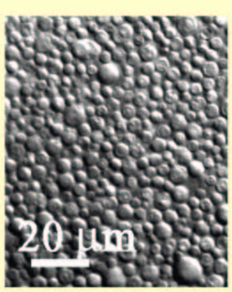
$\langle I(q,t)I(q,t+\tau)\rangle_{\text{Ensemble}}$

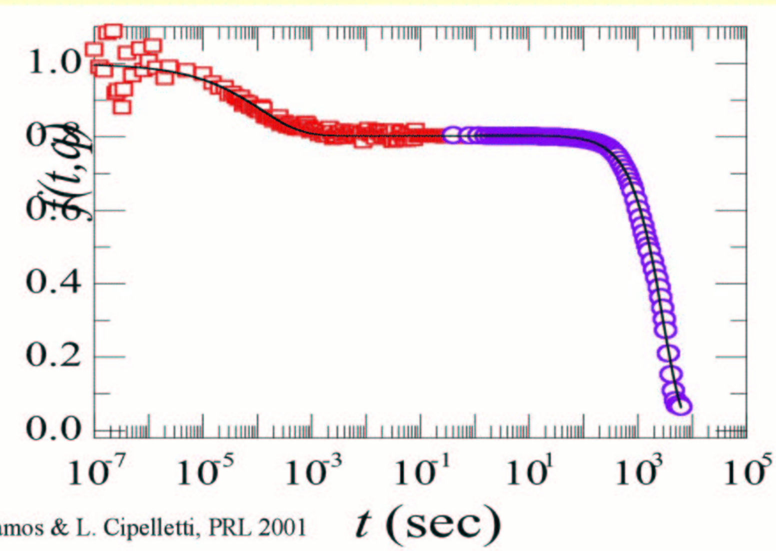
Multi-element detector (CCD),
software correlator

- **slow** relaxations, **non-stationary** dynamics
- **non-ergodic** samples

Slow Dynamics

Onion gel



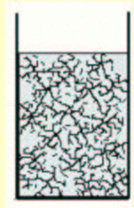


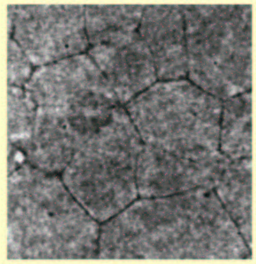
L. Ramos & L. Cipelletti, PRL 2001

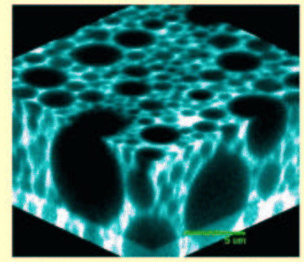
Final relaxation: « **compressed exponential** »
$\exp[-(t/\tau_f)^p]$ with $p\sim1.5$ and $\tau_f \propto q^{-1}$

Other systems…

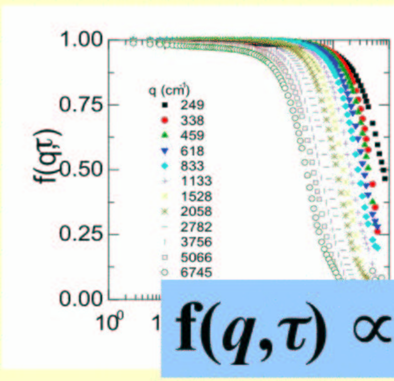
review: Cipelletti *et al.*, Faraday Discuss. **123** (2003)

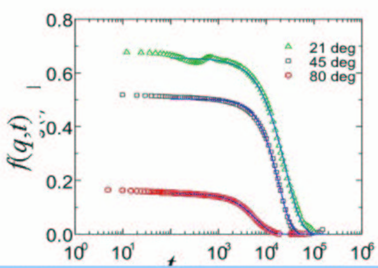


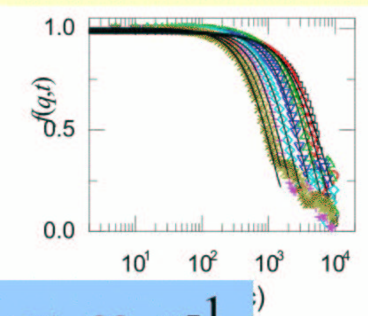




Colloidal particles gel | **Micellar polycrystal** | **Conc. Emulsion**







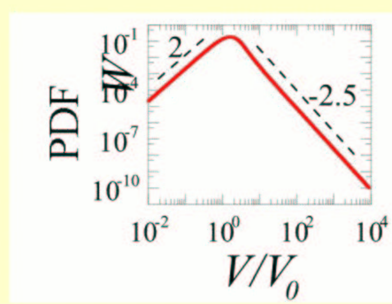
$$f(q,\tau) \propto \exp[-(t/\tau_f)^{3/2}],\ \tau_f \propto q^{-1}$$

Universal, non-diffusive dynamics

- Colloidal gels (highly attractive)
- Aggregated carbon black (weakly attractive) (XPCS)
- Onion gels (repulsive)
- Concentrated emusions (repulsive)
- Micellar polycrystal (repulsive)

Interpretation:

- **« ballistic » motion ($<\Delta r(t)> \propto t$)**
- **wide distribution of velocities**
- **internal stress relaxation**



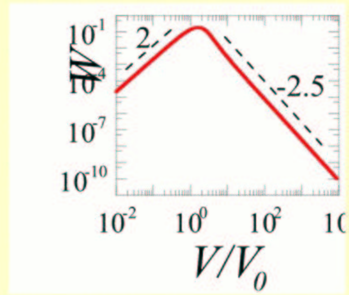
Velocity distribution function W

The dynamic structure factor $f(q,t)$ depends uniquely on $x=qt$

$$W(V) = 2V/\pi \int_0^\infty dx\, x\, f(x) \sin(xV)$$

Levy law

in our case: $W(V) = 2V/V_0 \frac{d}{dV}\left[L_{m,0}(V/V_0)\right]$



for $V >> V_0$

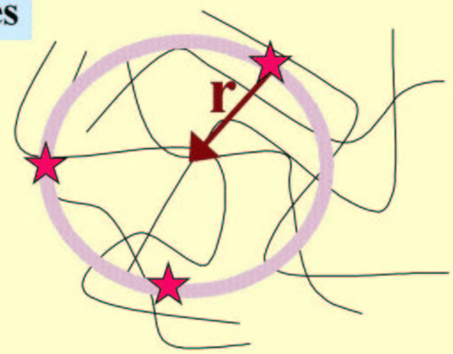
$W(V) \sim V^{-(m+1)} \sim V^{-2.5}$

Simple models : Relaxation of internal stresses

Deformation field: $\Delta R(t) \approx \alpha(t) r^{-2}$

Ballistic motion ⟹ $\alpha(t) \sim t$

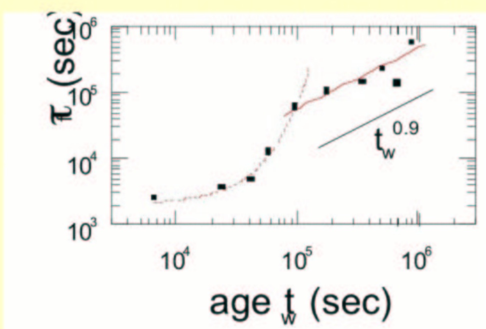
$V = dR/dt \sim r^{-2}$

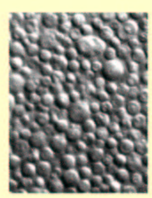


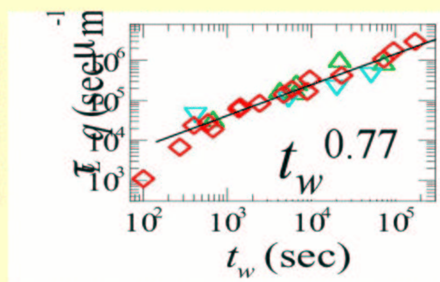
Homogeneous distribution of stress sources:

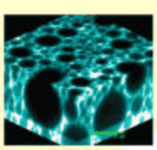
$dN \sim r^2 dr$ and $W(V) = dN/dV \sim r^5 \sim V^{-2.5}$

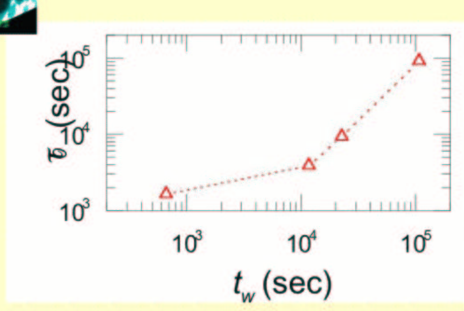
Aging in jammed systems

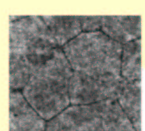


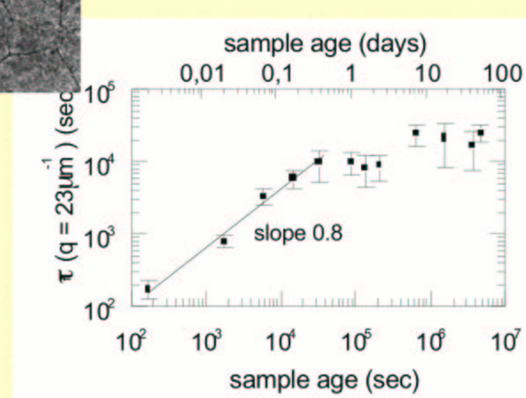


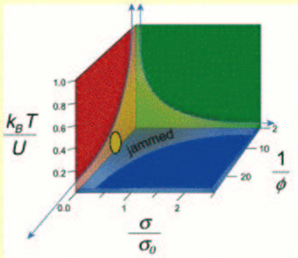










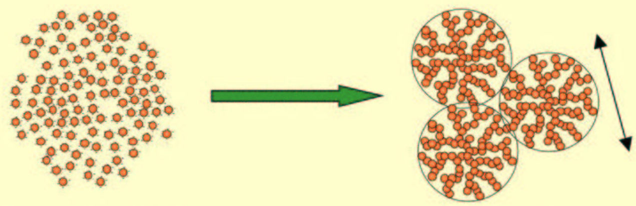


Colloidal gel

With H. Bissig, V. Trappe

Polystyrene Spheres

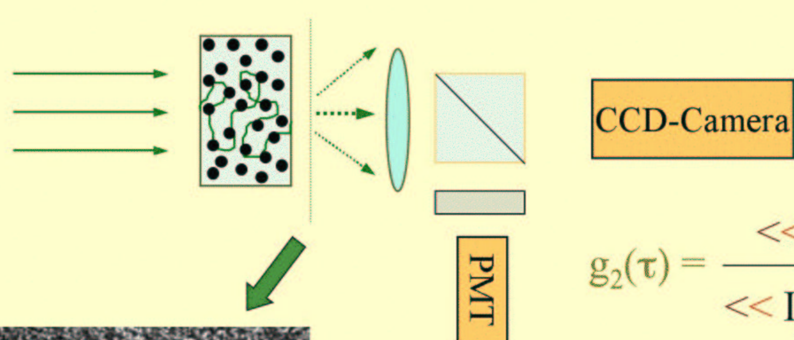
$a = 85$ nm , $\phi = 4.5\%$

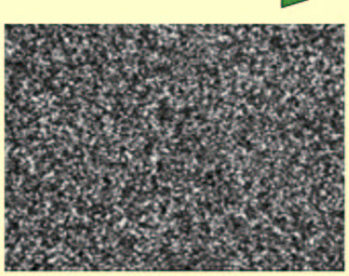


$$(NH_2)_2CO + H_2O \xrightarrow{\text{UREASE}} CO_3^{2-} + 2NH_4^+$$

Experimental setup

Diffusing Wave Spectroscopy

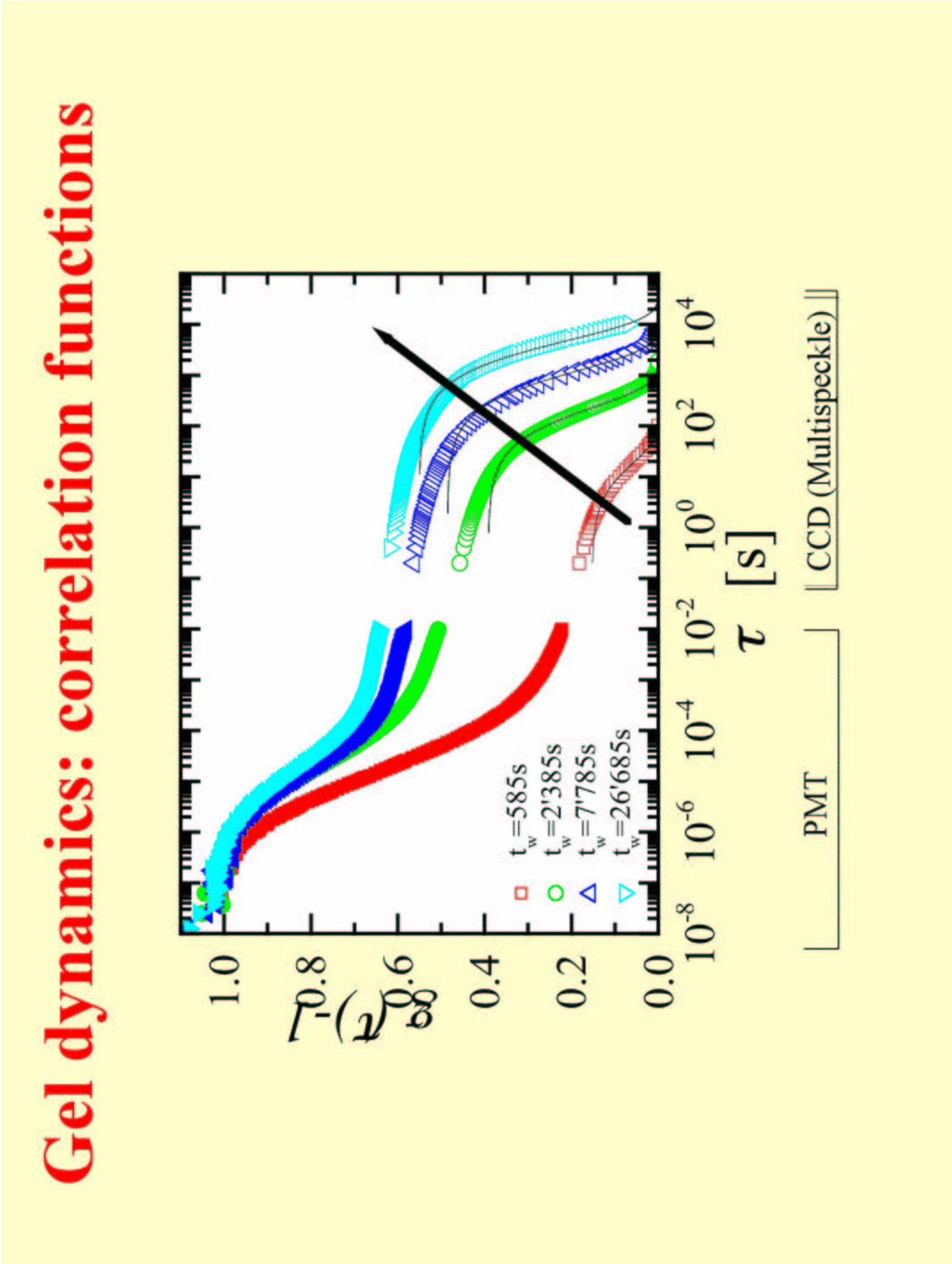


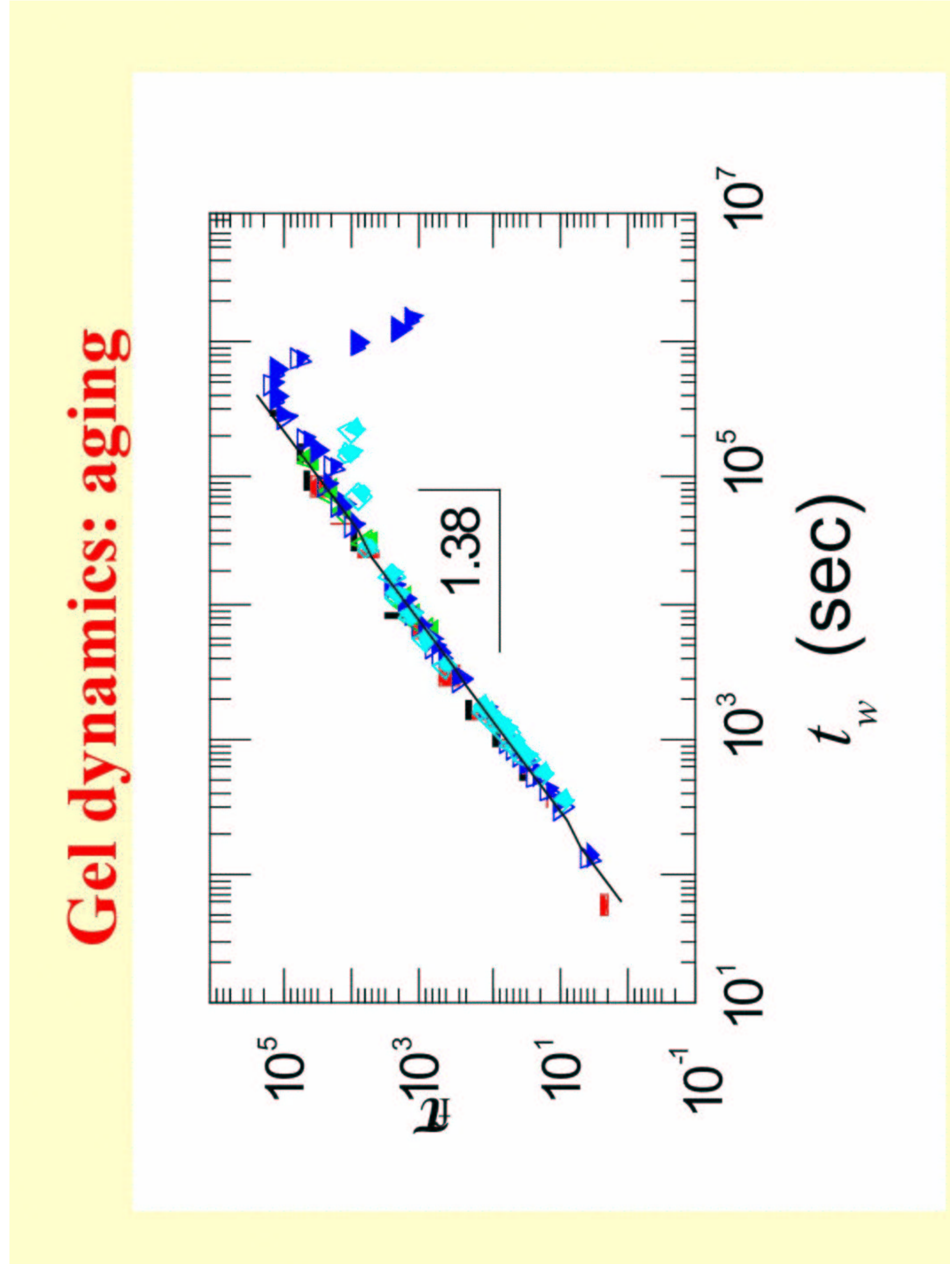


Slow Dynamics:
Multispeckle Technique

$$g_2(\tau) = \frac{\langle\langle I_p(t)\, I_p(t+\tau)\rangle_p\rangle_t}{\langle\langle I_p(t)\rangle_p\rangle_t \, \langle\langle I_p(t+\tau)\rangle_p\rangle_t}$$

Fast Dynamics:
Double Cell Technique


Gel dynamics: correlation functions
g2(τ)-1
1.0
0.8
0.6
0.4
0.2
0.0
10-8 10-6 10-4 10-2 100 102 104
τ [s]
t_w=585s
t_w=2'385s
t_w=7'785s
t_w=26'685s
PMT
CCD (Multispeckle)


Gel dynamics: aging
1.38
10-1
101
103
105
107
t_w (sec)

Physical origin ???

g_2-1 **exponential** → Standard DWS formalism → $<\Delta r^2(t)> \sim t$

($\Delta r \sim 10$ nm)

???

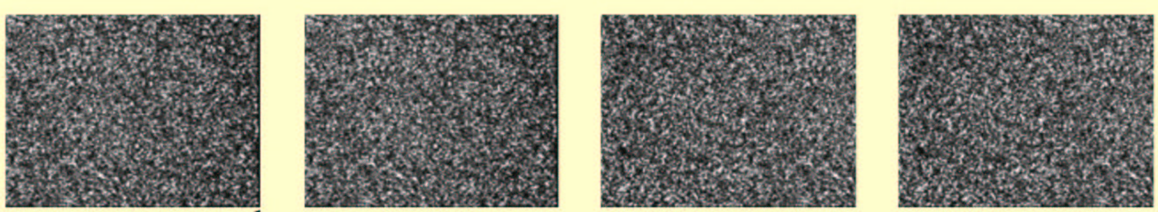
Foams: (Durian *et. al.* Science 1991)

Intermittent dynamics (DWS and microscopy)

g_2-1 **exponential** → **rearrangement events localized in space and time**

Time Resolved Correlation (TRC)

Cipelletti *et al.*, J Phys Condens. Matt. **15** (2003)



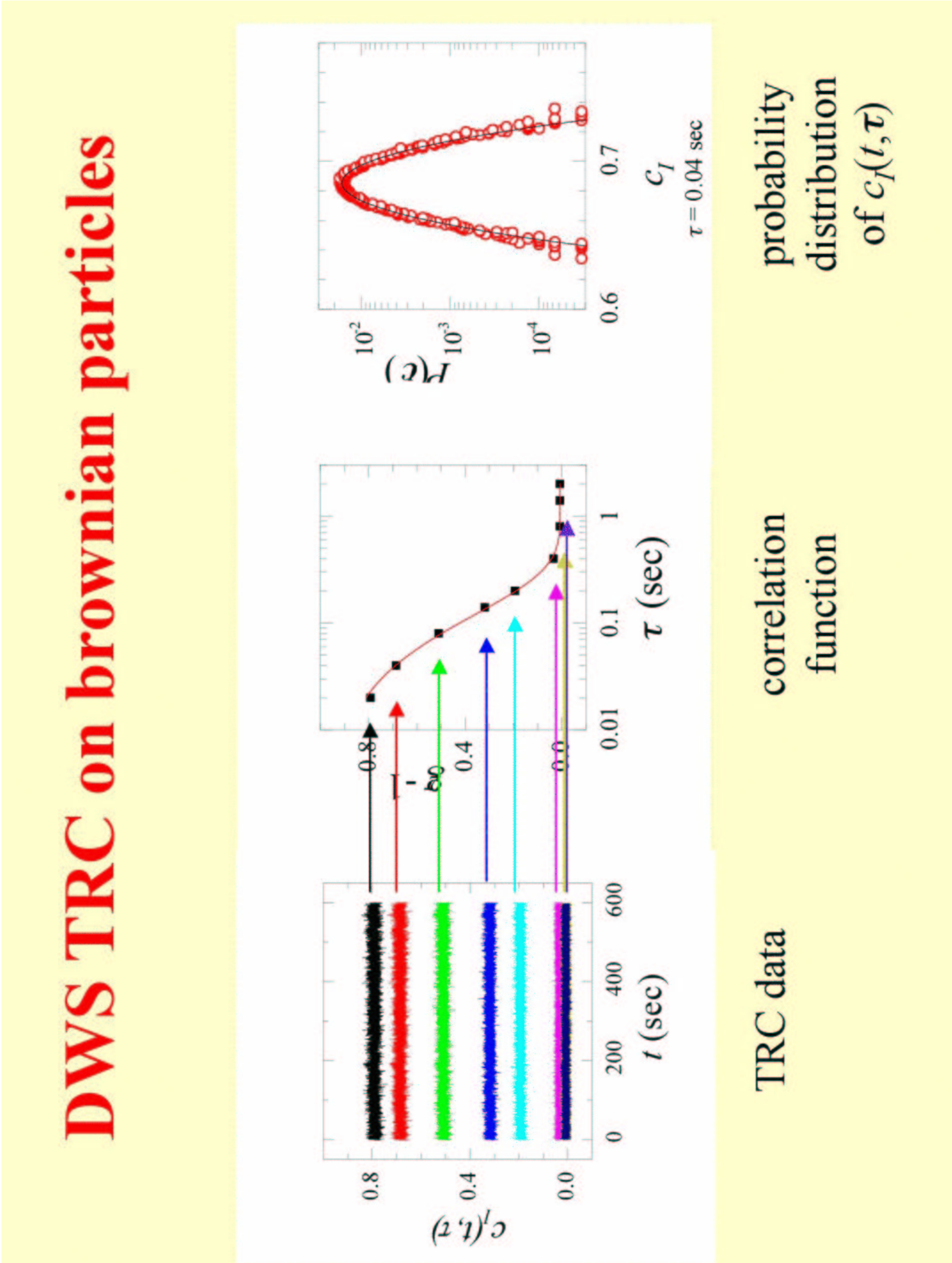
lag τ

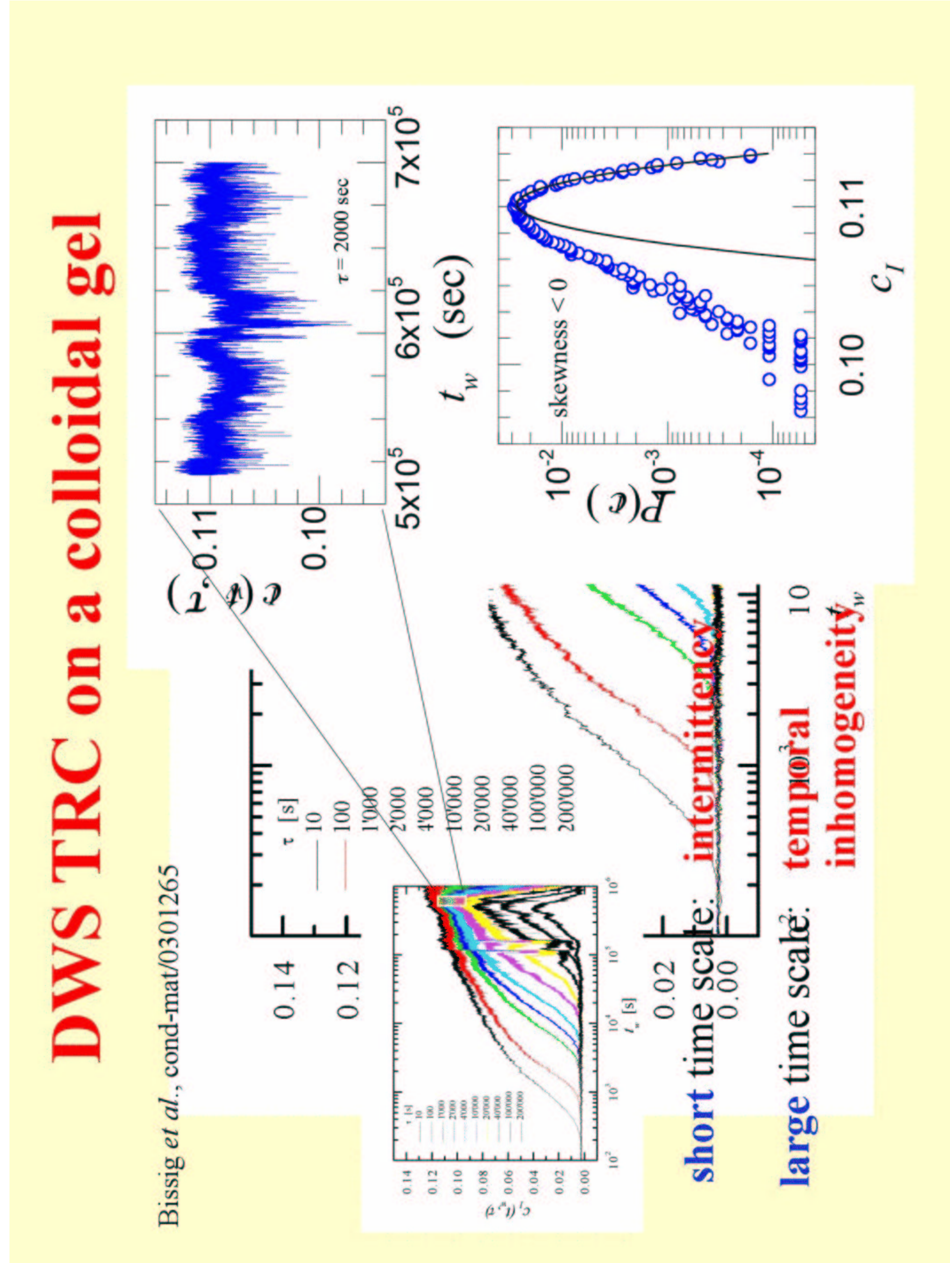
time t

continuous dynamics → **smooth change** of speckle pattern

intermittent dynamics → **sudden change** of speckle pattern

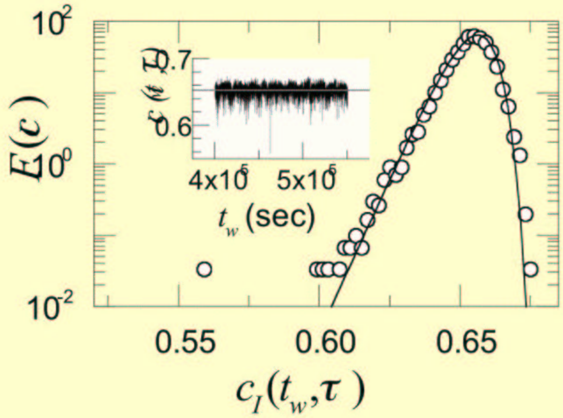
degree of correlation $c_I(t,\tau) = \dfrac{< I_p(t)\, I_p(t+\tau)>_p}{< I_p(t)>_p < I_p(t+\tau)>_p} - 1$


DWS TRC on brownian particles
TRC data
correlation function
probability distribution of $c_I(t,\tau)$
$c_I(t,\tau)$
t (sec)
$g_2 - 1$
τ (sec)
$P(c)$
c_I
$\tau = 0.04$ sec


DWS TRC on a colloidal gel
Bissig *et al.*, cond-mat/0301265
short time scale: intermittency
large time scale: temporal inhomogeneity
$c_I(t_w,\tau)$
t_w (sec)
$\tau = 2000$ sec
skewness < 0
$P(c)$
c_I

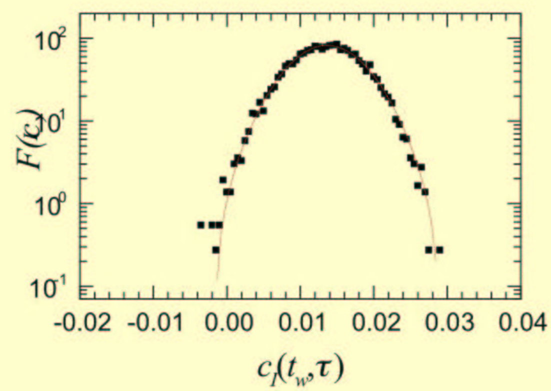
τ dependence of the PDFs

$\tau/\tau_f = 0.01$



PDF: Gumbel, see Bramwell *et al.* PRL **84** 3744 (2000)

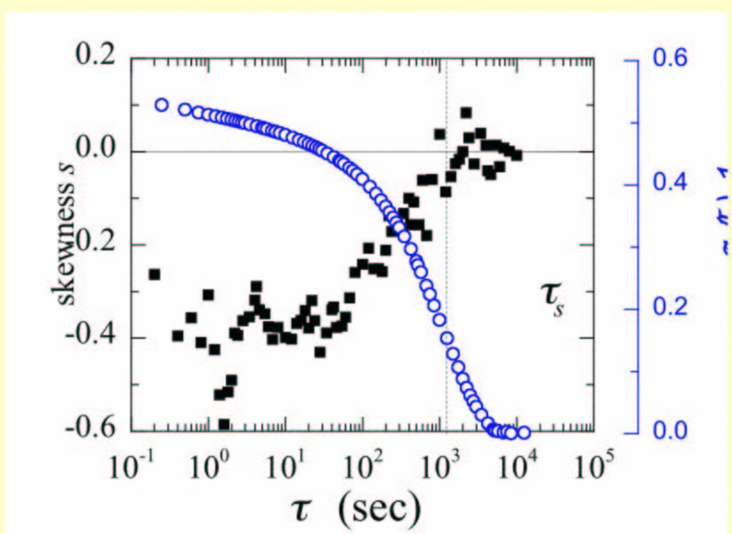
$\tau/\tau_f = 10$



PDF: Gaussian

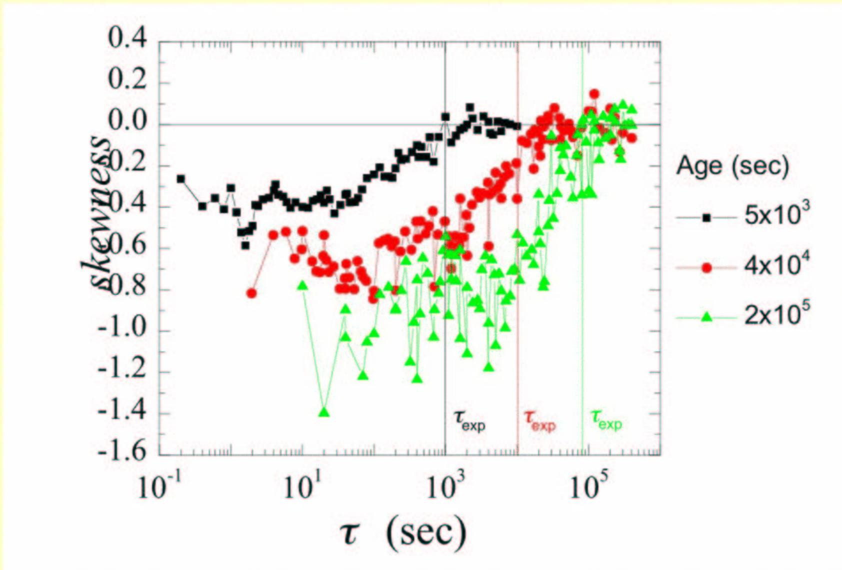
DWS TRC on a colloidal gel

Is intermittent dynamics main mechanism for slow relaxation ?



τ dependence of skewness

Evolution of skewness during aging



Physical picture

Discrete **rearrangements** due to **bond formation** or **bond breaking**

Transient deformation (buckling/unbuckling?)

Permanent (small) change in gel configuration

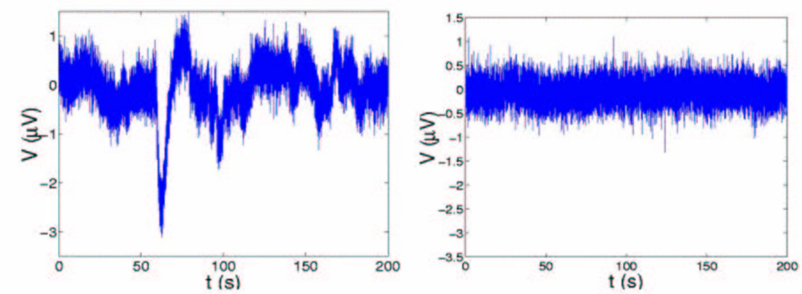
What about other glassy materials?

Dielectric measurements on colloidal and polymer glasses

(Buisson, Bellon, Ciliberto, Lyon)

Signals in Laponite electric measurements

Signal as function of time



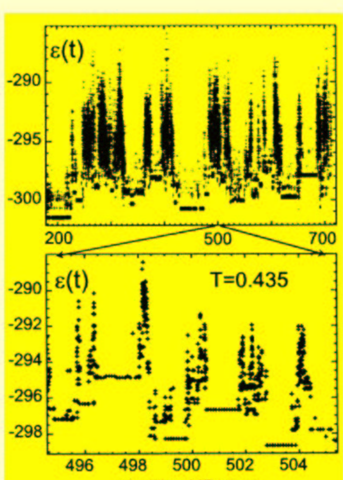
1 hour after preparation

16 hour after preparation.

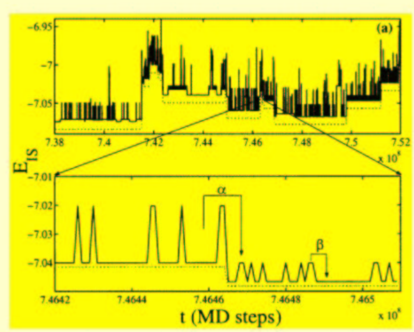
Simulations of supercooled liquids

Energy of « inherent structures » (local minima)

(Doliwa and Heuer, cond-mat/0205283)



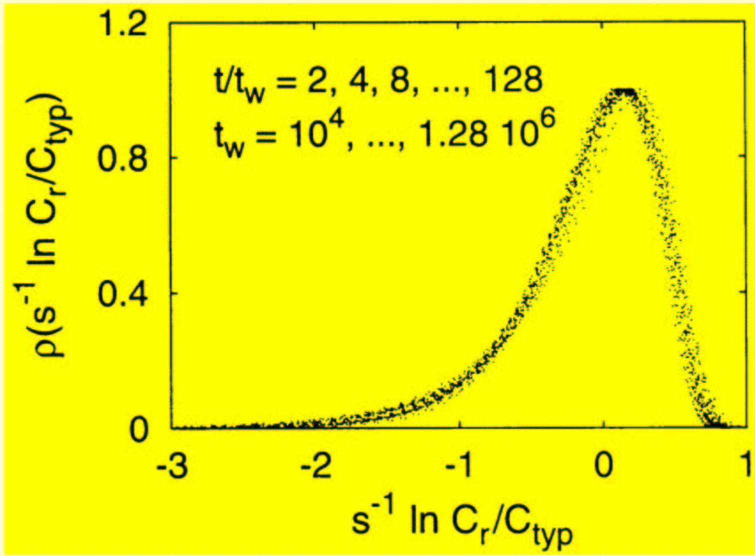
(Denny et al. Cond-mat/0209020)



Simulations on spin glasses

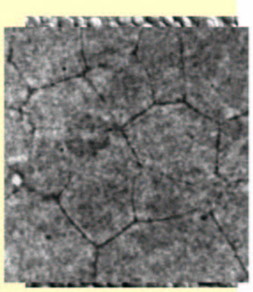
(Castillo, Chamon, Cugliandolo, Kennett, PRL 2002)

Normalized PDF of local correlation in aging spin glasses



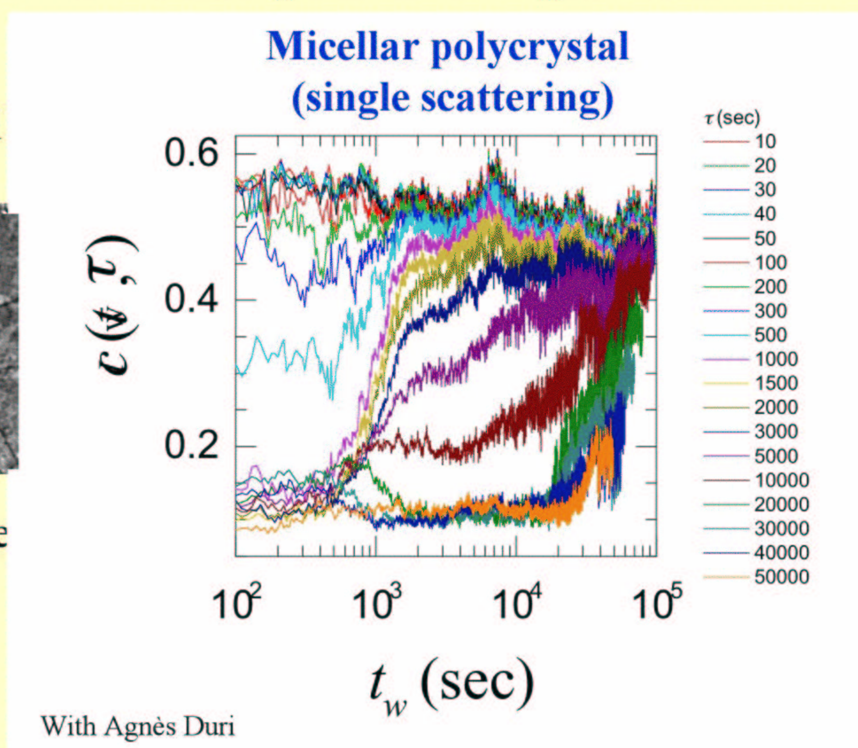
Temporal heterogeneity and intermittence in other jammed systems

• Colloidal



DWS)

• Onion Ge



Conclusions

« soft glasses »	heterogeneous slow dynamics, aging.
New techniques	confocal microscopy, multispeckle DLS, TRC…
General features	**« ballistic » relaxation** (internal stress), **intermittence** and **heterogeneity** of the dynamics

Analogies with structural, polymer, and spin glasses…

Statistical physics of out-of-equilibrium systems

Our works on Jamming & slow dynamics

1. H. Bissig, V. Trappe, S. Romer, L. Cipelletti, *Intermittency and non-Gaussian fluctuations in the dynamics of aging colloidal gels*, submitted (preprint no. cond-mat/0301265).
2. Luca Cipelletti, H. Bissig, V. Trappe, P. Ballesta, S. Mazoyer, *Time Resolved Correlation : a new tool for studying temporally heteroheneous dynamics*, J. Phys. : Condens. Matter **15**, S257 (2003).
3. Luca Cipelletti, Laurence Ramos, S. Manley, E. Pitard, D. A. Weitz, Eugene E. Pashkovski, Marie Johansson, *Universal non-diffusive slow dynamics in aging soft matter*, Farad. Discuss. **123**, 237 (2003).
4. V. Prasad, V. Trappe, A. D. Dinsmore, P. N. Segre, L. Cipelletti and D.A. Weitz, *Universal Features of the Fluid to Solid Transition for Attractive Colloidal Particles*, Farad. Discuss. **123**, 1 (2003).
5. Luca Cipelletti and Laurence Ramos, *Slow dynamics in glasses, gels, and foams*, Current Opinions in Colloidal and Interface Science 7, 228 (2002) .
6. L. Ramos and Luca Cipelletti, *Ultraslow dynamics and stress relaxation in the aging of a soft glassy system*, Phys. Rev. Lett. **87**, 245503 (2001).
7. V. Trappe, V. Prasad, Luca Cipelletti, P. N. Segre, and D. A. Weitz, *Jamming phase diagram for attractive particles*, Nature **411**, 722 (2001).
8. L. Cipelletti, S. Manley, R. C. Ball, and D. A. Weitz, *Universal Aging Features in the Restructuring of Fractal Colloidal Gels*, Phys. Rev. Lett. **84**, 2675 (2000).
9. L. Cipelletti and D. A. Weitz, *Ultralow-angle dynamic light scattering with a charge coupled device camera based multispeckle, multitau correlator*, Rev. Sci. Instrum. **70**, 3214 (1999).

Ref. 5 contains a list of many other references…

See also www.gdpc.univ-montp2.fr7082/~lucacip/index.html