Slow relaxations and noise in glassy systems

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(CNRS URA 2464)

Thanks to S. Ciliberto (ENS Lyon)

Seminar at Tokyo University, Hongo Campus, November 2010
with the support of G-COE

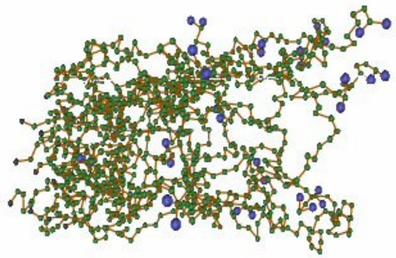
A few examples of "glasses" in physics

Silicates, etc.

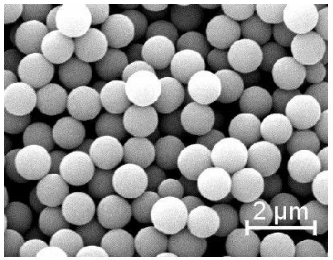
Molecular glasses

(ex: glycerol)

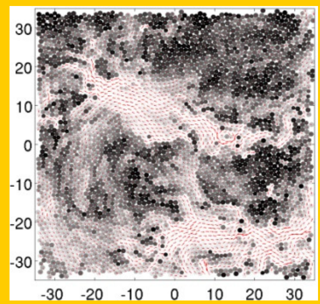
Polymers



Colloids

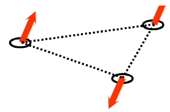


Granular matter



Spin glasses

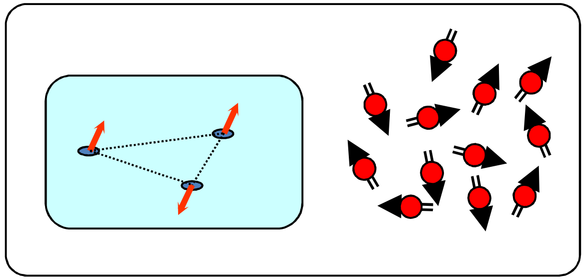
$$H = - \sum J_{ij} S_i S_j$$



What is a spin glass ?

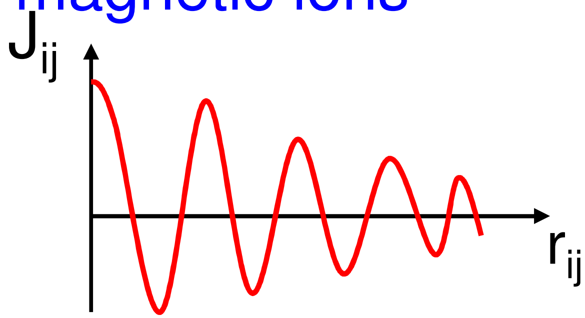
Theory : random bonds $H = -\sum J_{ij}\, \mathbf{S_i}.\mathbf{S_j}$ $\{J_{ij}\}$ gaussian, or $\pm J$

conceptually simple…



a disordered *and* frustrated magnetic system

***"Real"* spin glasses :** random dilution of magnetic ions



example: metallic alloys, Cu:Mn 3%

RKKY interactions

same generic behaviour in all samples

(Tc≠0 in 3d, slow dynamics, aging...)

→ « *model* » *disordered systems*

1. Response: slow relaxations, aging

2. Fluctuations (noise): comparison with response

1. Response: slow relaxations, aging

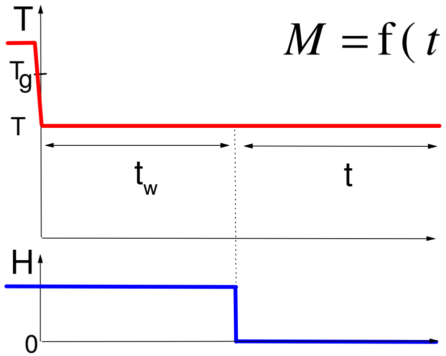
2. Fluctuations (noise): comparison with response

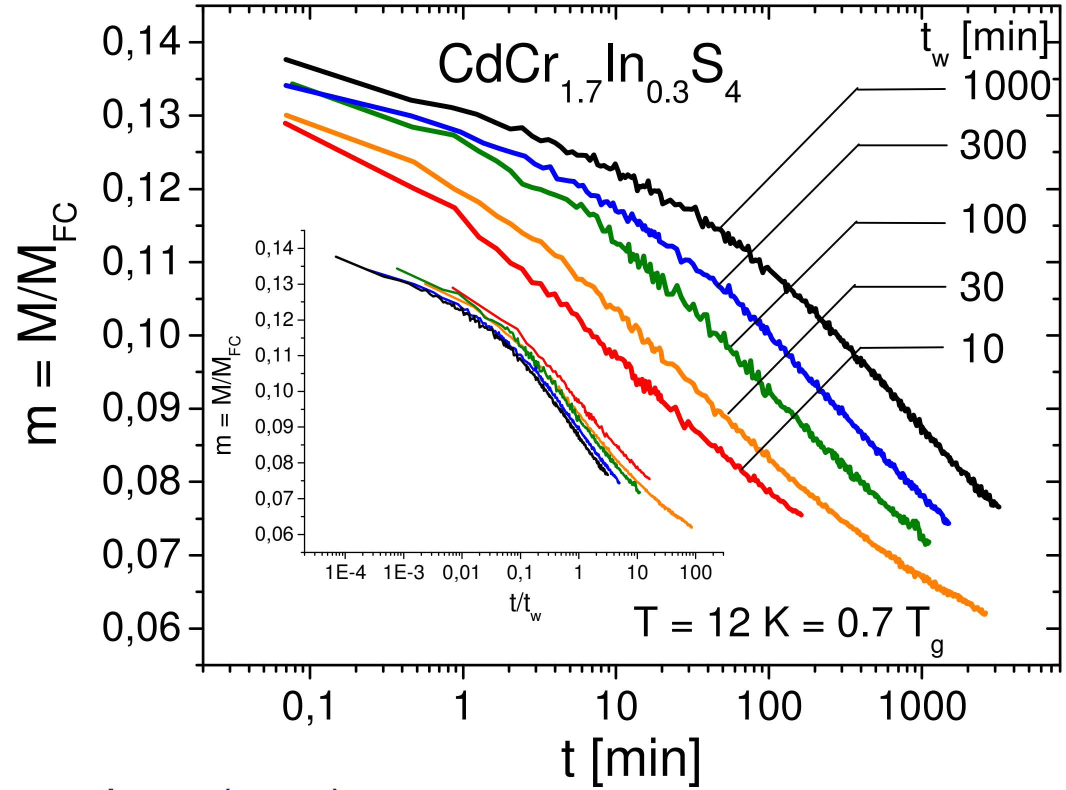
Spin glasses: slow dynamics + aging

80' Uppsala (Lundgren, Nordblad)
Saclay (Hammann, Ocio, Alba, Vincent)

relaxation of theThermo-Remanent Magnetization (TRM)

$$M = f(t, t_w)$$





t_w : waiting time
t : observation time
t_w+t : « age »
H : weak magnetic field

→ Non-stationary dynamics : (t, t_w)
Approximate scaling variable : t / t_w (or t / t_w^{μ} with $\mu<1$)

THE JOURNAL OF CHEMICAL PHYSICS **123**, 174507 (2005)

Shear stress relaxation and physical aging study on simple glass-forming materials

X. Shi, A. Mandanici, G.B. McKenna, Texas Tech University

Shear relaxation response at different aging times at 6K below *Tg*

m-toluidine (fragile glass-former)
T_g = 187K

Master curve as a function of $t/a(t_w)$ (offset by one decade for clarity).

Insert: shift factors $a(t_w)$ used for master curve
log a vs log tw : slope 0.61 (~μ for SG*)*

The leveling off of shift factors at longer aging times indicates that the sample has aged into equilibrium.

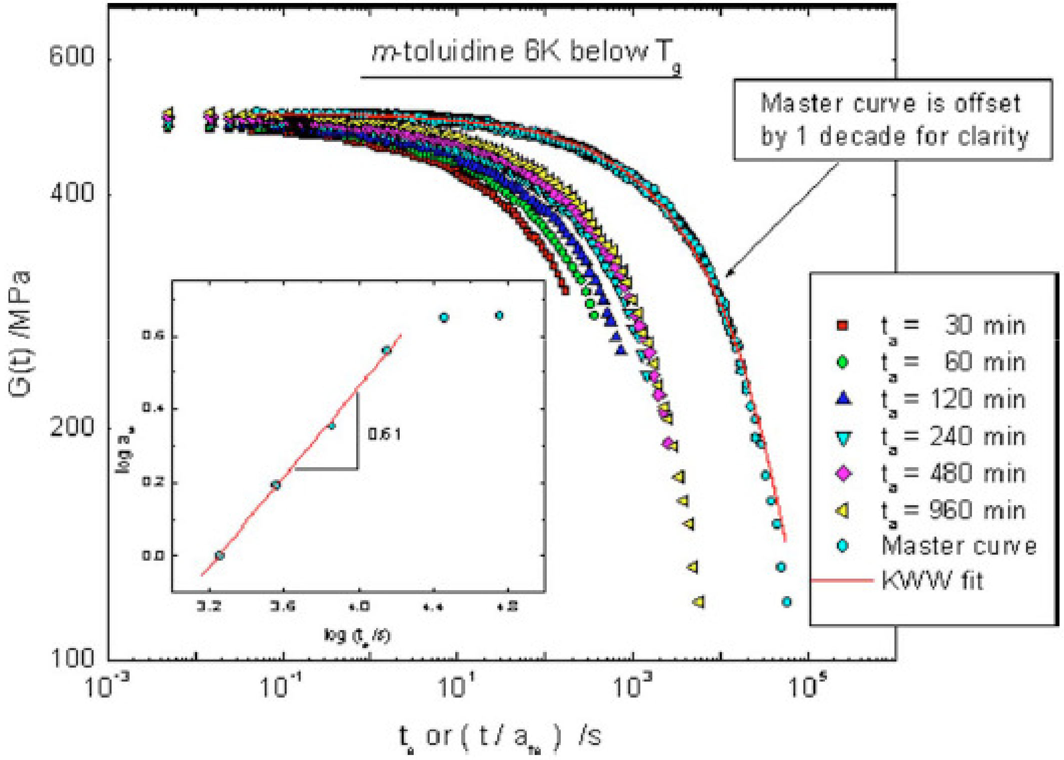


FIG. 7. Shear relaxation response at different aging times for *m*-toluidine at 181 K, which is 6 K below the nominal T_g. The time-aging time master curve was constructed and is offset by one decade for clarity. The curve represents the KWW function fit to the master curve. The insert shows the shift factors used. The leveling off at longer aging times indicates that the sample has aged into equilibrium.

THE JOURNAL OF CHEMICAL PHYSICS 123, 174507 (2005)

Shear stress relaxation and physical aging study on simple glass-forming materials

X. Shi, A. Mandanici, G.B. McKenna, Texas Tech University

Shear relaxation response at different aging times at 6K below *Tg*

Sucrose benzoate (fragile glass-former)
T_g = 337K

Shift factors for master curve :
slope 0.26 (~μ for SG)

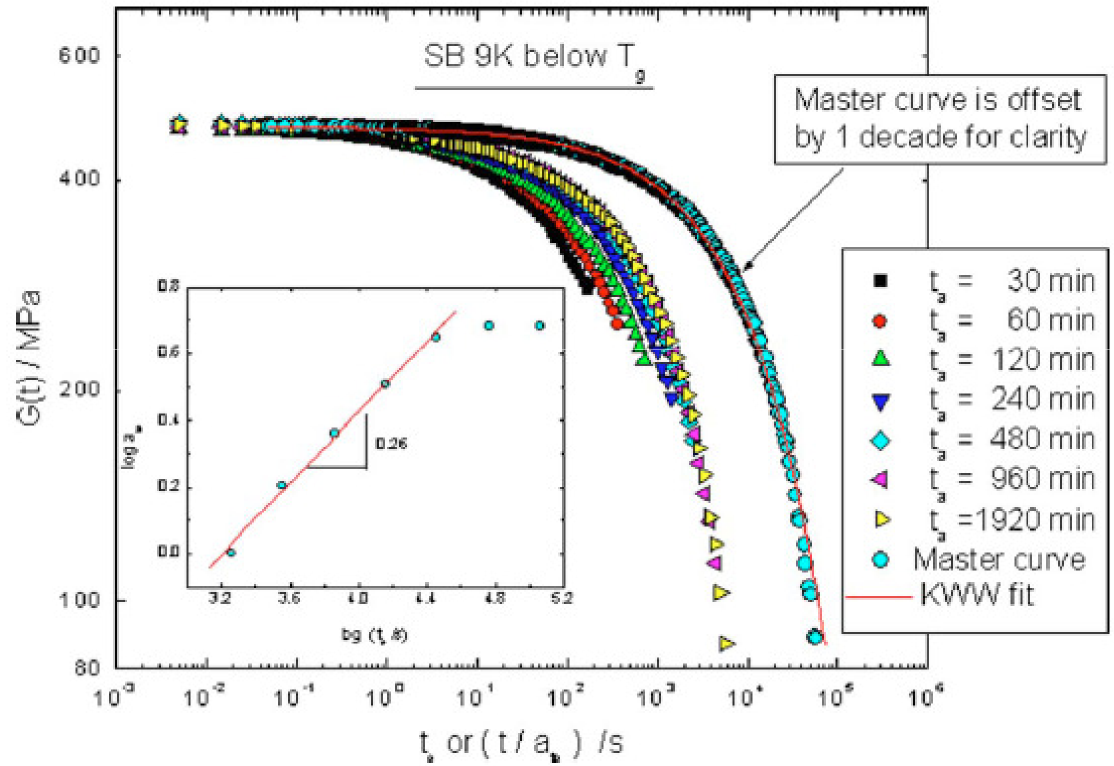
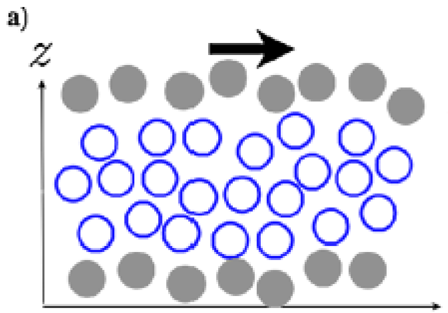


FIG. 10. Shear relaxation response at different aging times for sucrose benzoate at 328 K, which is 9 K below the nominal T_g. The time-aging time master curve was constructed and is offset by one decade for clarity. The curve represents the KWW function fit to the master curve. The insert shows the shift factors used. The leveling off at longer aging times indicates that the sample has aged into equilibrium.

Stress relaxation: Monte Carlo simulations

H. Yoshino and M. Mézard, paper in progress – see also PRL 105, 015504 (2010)

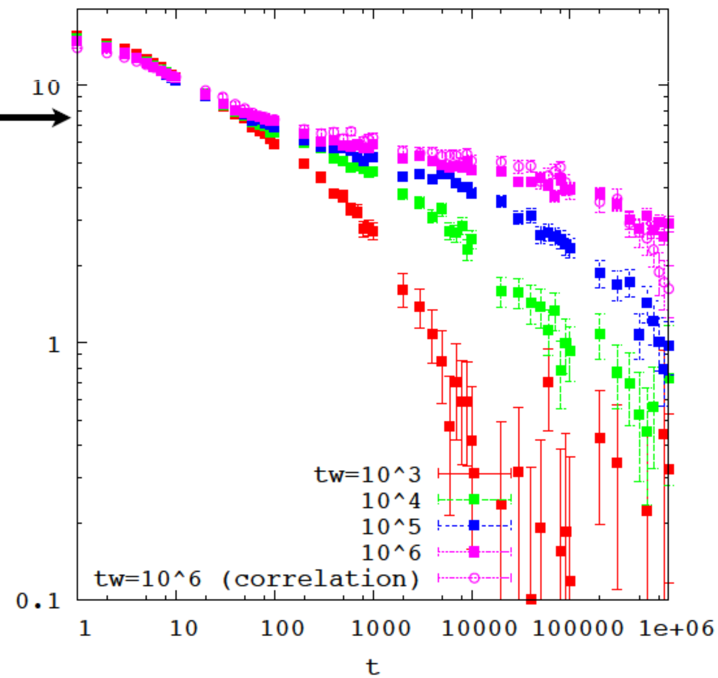
N particles interacting via 2-body potentials



Theory

$\hat{\mu} = 7.73$ →

σ / γ



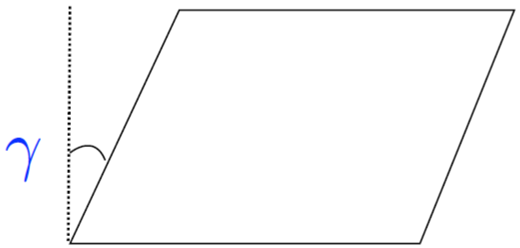
t

$N = 512$

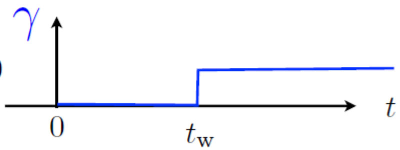
$T = 0.1$

$\gamma = 0.025$

Strain γ



quench at $t = 0$



PRL 103, 138302 (2009) PHYSICAL REVIEW LETTERS week ending 25 SEPTEMBER 2009

Interplay between Shear Loading and Structural Aging in a Physical Gelatin Gel

O. Ronsin, C. Caroli, and T. Baumberger

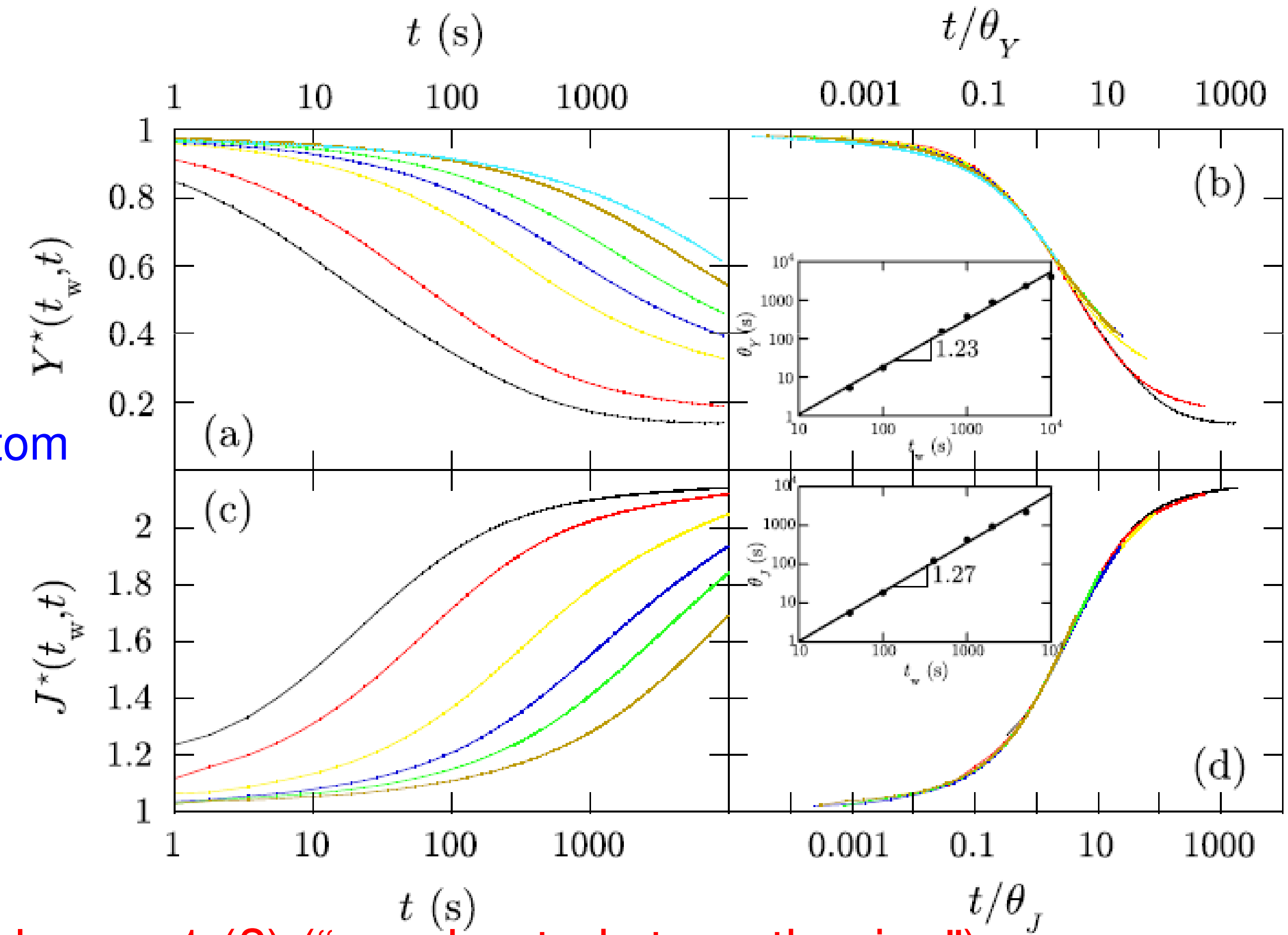
Gelatin :

T_g=29℃, T=20℃

Thermo-reversible gel
Below T_g, renaturation of triple helix structure, stabilized by H-bonds

(a) Normalized stress relaxation modulus *Y* for waiting times, from bottom to top, t_w = 40- 10^4s.
(b) Same data rescaled

(c) Normalized creep compliance *J*
(d) Same data rescaled



Inserts: shift factor slope > 1 (?) (“accelerated strengthening”)

1. Response: slow relaxations, aging

2. Fluctuations (noise): comparison with response

2.a in magnetic systems

2.b in other glassy materials

1. Response: slow relaxations, aging

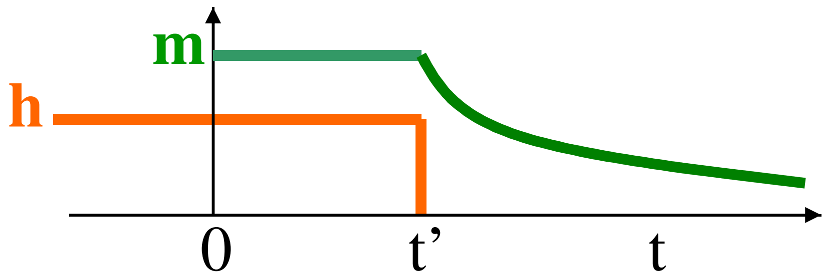
2. Fluctuations (noise): comparison with response

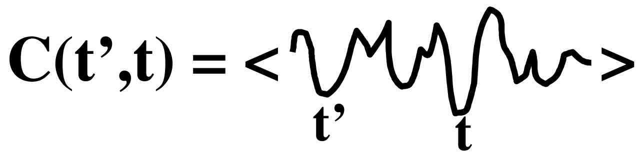
2.a in magnetic systems

2.b in other glassy materials

Spontaneous magnetic fluctuations and response functions

→ Fluctuation-Dissipation relation in an out-of-equilibrium system





σ(t',t) = m/h response at *t* after field cutoff at *t'*

C(t',t) = < m(t').m(t)> autocorrelation of the fluctuations

Fluctuation-Dissipation relation (FDR): ***σ = C/kT***

(ergodic systems at equilibrium)

Extension of FDR to non-equilibrium situations:

σ = C. F(C)/kT *(for large t')*

T / F(C) *≡ effective temperature*

Cugliandolo Kurchan, *J. Phys. A* **27**, 5749 (1994)

spin glass in aging regime = example of out-of-equilibrium system

Measurement of fluctuations and response in the same setup

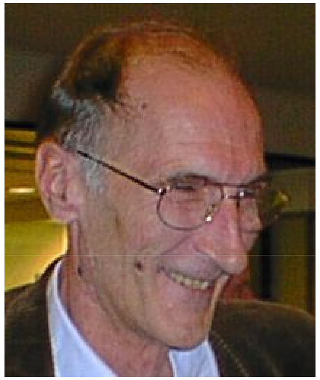
after the pioneering work of Ocio, Bouchiat, Monod, Refregier ~1985

Fluctuations: measurement of spontaneous flux variations (noise)

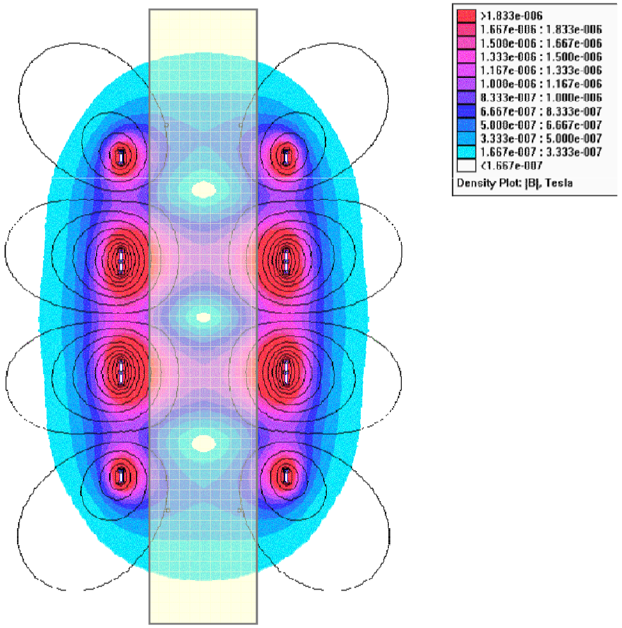
Response: the excitation field is applied via the detection coil

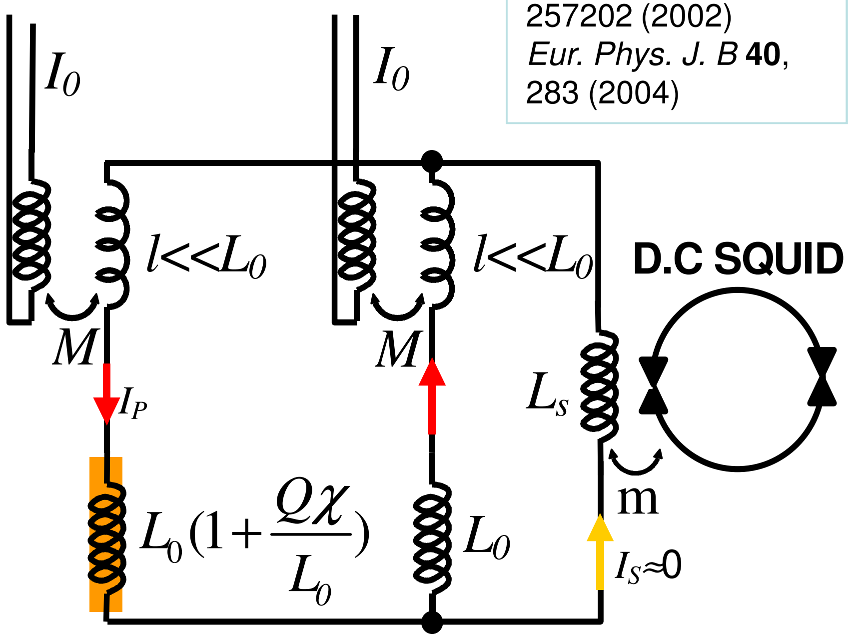
→ same field geometry as in fluctuation measurement

D. Hérisson and M. Ocio,
Phys. Rev. Lett. **88**, 257202 (2002)
Eur. Phys. J. B **40**, 283 (2004)



Miguel Ocio
(1943-2003)
D. Hérisson thesis

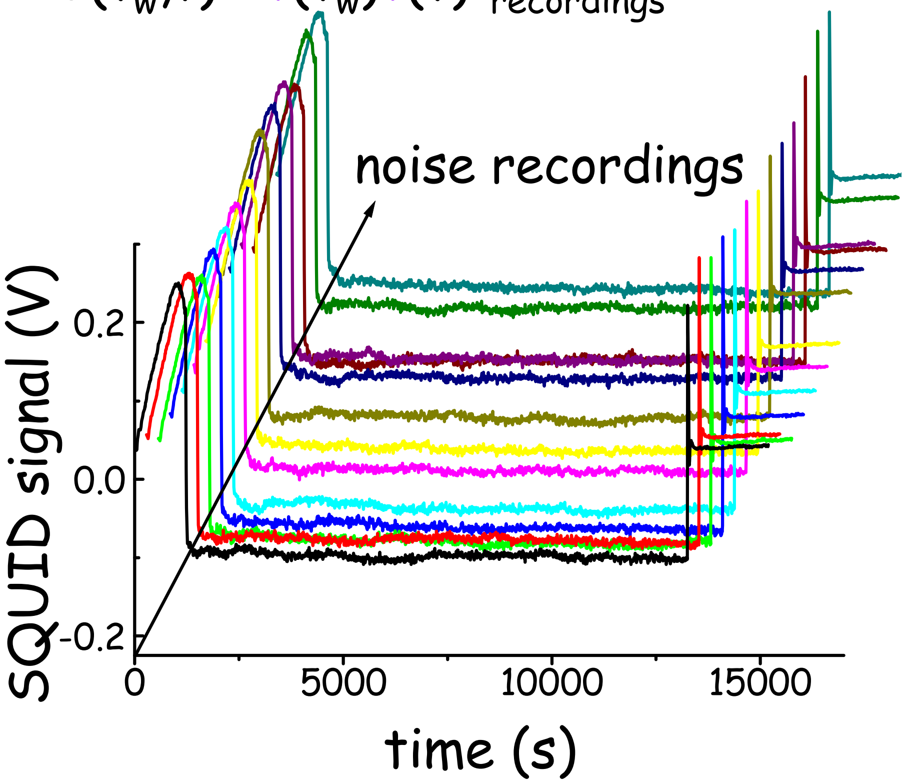




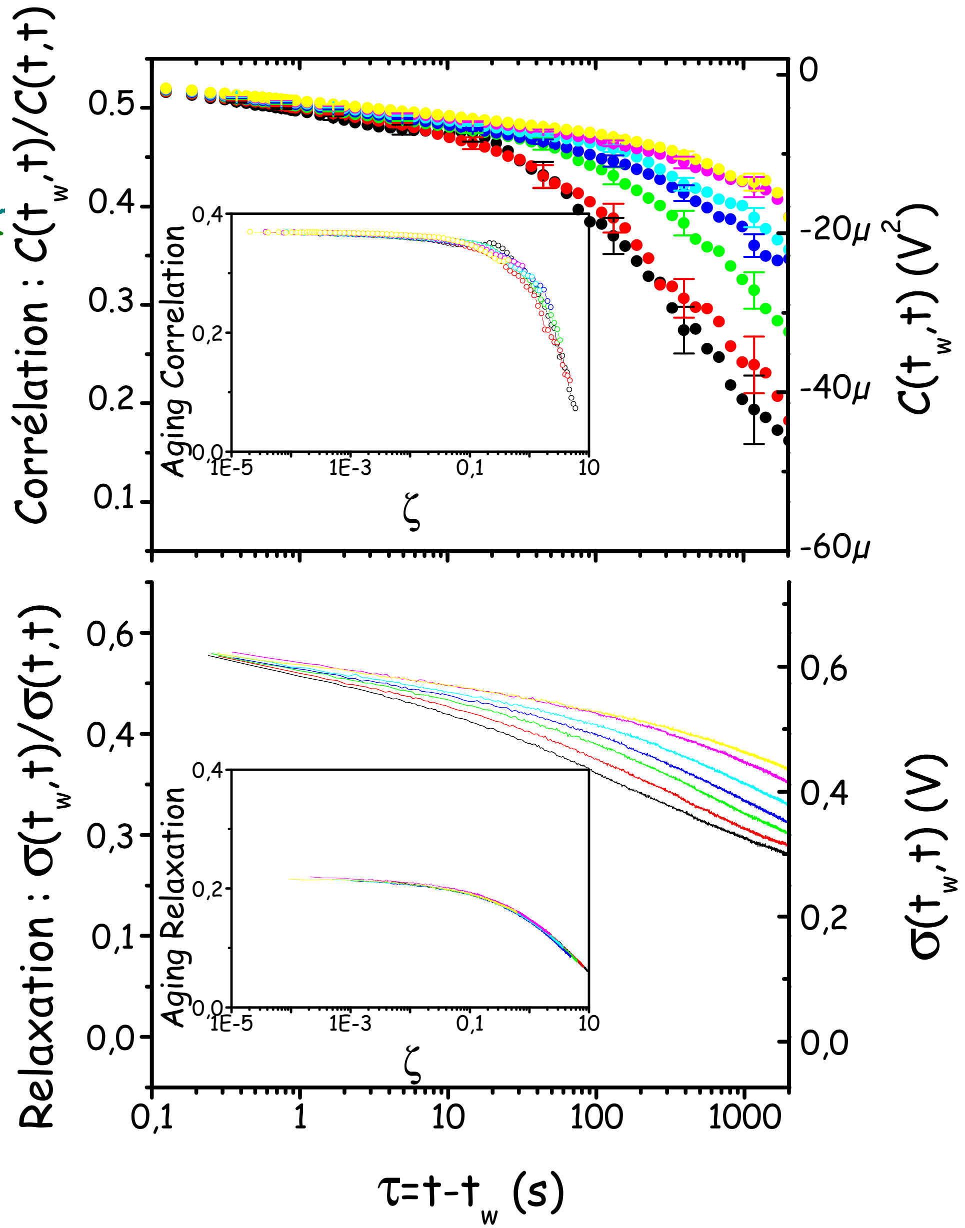
- fluctuations ~ response to 10^{-7} G
- setup calibration by *eddy current measurements* in pure Cu
- long time measurements ⇒ get rid of slow spurious drifts…

Noise measurements in a spin glass : autocorrelation function

$C(t_w,t) = \langle v(t_w)\,v(t) \rangle_{recordings}$

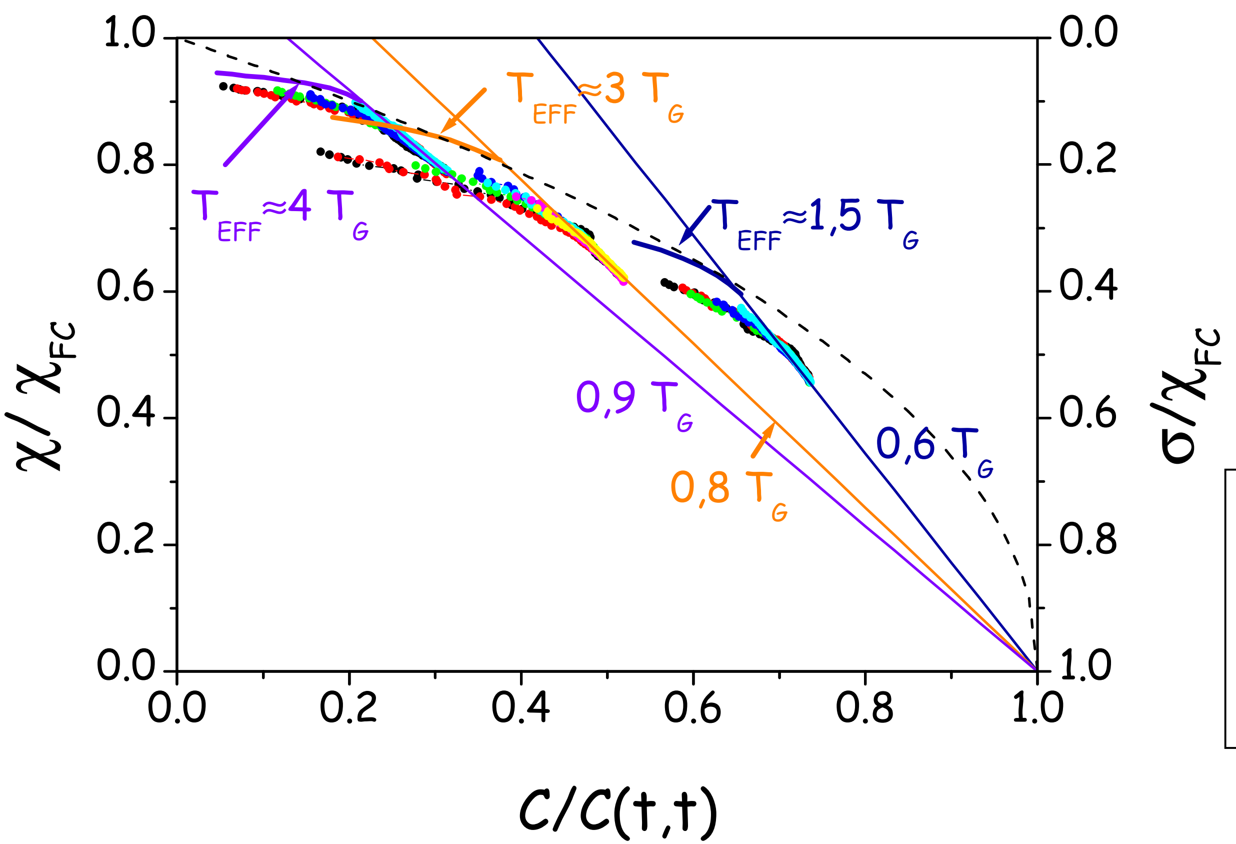


average of $C(t_w,t)$ over 300 measurements of 10000s



→ Comparison of autocorrelation and response, fluctuation-dissipation relations *in the aging regime*

Fluctuation-Dissipation Relation graph



$CdCr_{1.7}In_{0.3}S_4$ spin glass

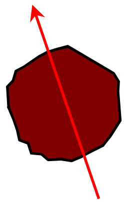
D. Hérisson and M. Ocio,
Phys. Rev. Lett. **88**, 257202 (2002)
Eur. Phys. J. B **40**, 283 (2004)

- clear 1/T regime, and crossover to aging regime $1/T_{eff}$
- vanishing t_w-dependence in the 'extrapolation' → $T_{eff} = f(C)$

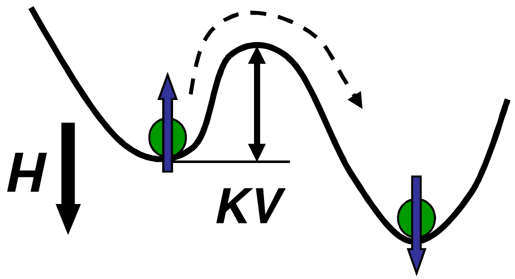
- not domain growth-like ($1/T_{eff}=0$, horizontal lines)
- 1-step RSB type models: *straight lines of slope $1/T_{eff}$ - compatible*
- continuous RSB models (SK, mean-field spin glass): $\chi=1-\sigma=(1-C)^{0,47}$ *(dashed line)*

Super-Spins, Superspin Glass (SSG)

- Small enough ferromagnetic nanoparticle → single domain
- $T \ll T_{Curie}$: response of single nanoparticle ~ response of single spin
  → a 'superspin'



- Easy axis → anisotropy barrier ~K.V
- T<<KV → blocking of magnetization below T_B ~ KV



- Varying concentration of nanoparticles changes interparticle interaction
  Case of ferrofluid (liquid suspension - frozen): dipole-dipole interaction

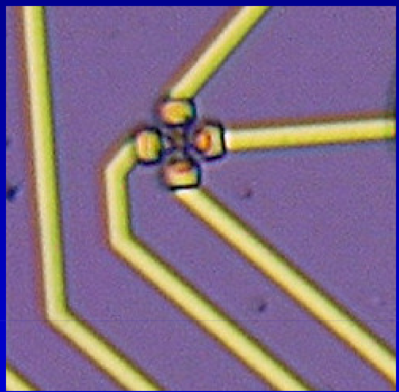
Dilute nanoparticle system
↓
Non-interacting superspins
Superparamagnet

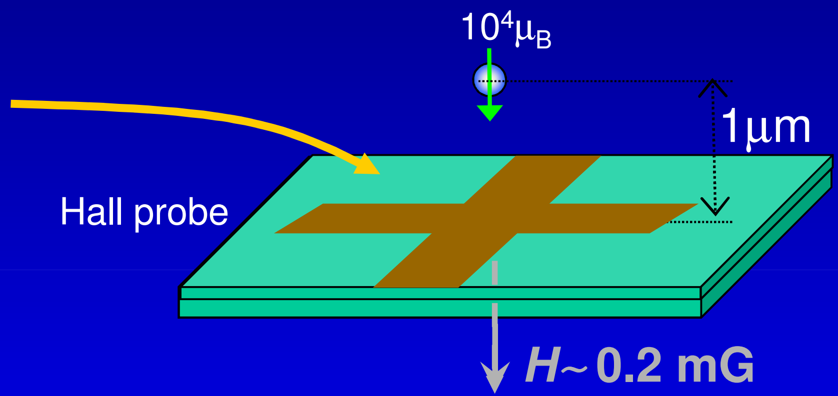
Concentrated nanoparticle system
↓
Interacting superspins
« Superspin glass »

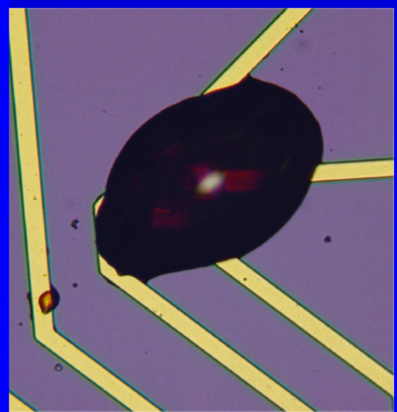
Noise measurements of interacting magnetic particles with high resolution Hall microprobes

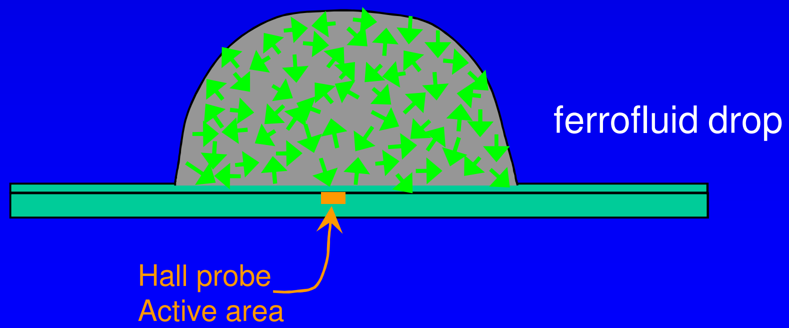
Hall probes: 2DEG in AlGaAs/InGaAs/GaAs heterojunction. Size of active area ~ 1-10 μm^2

From 4 to 300K, from low to high fields, good resolution (2mG/Hz$^{1/2}$)





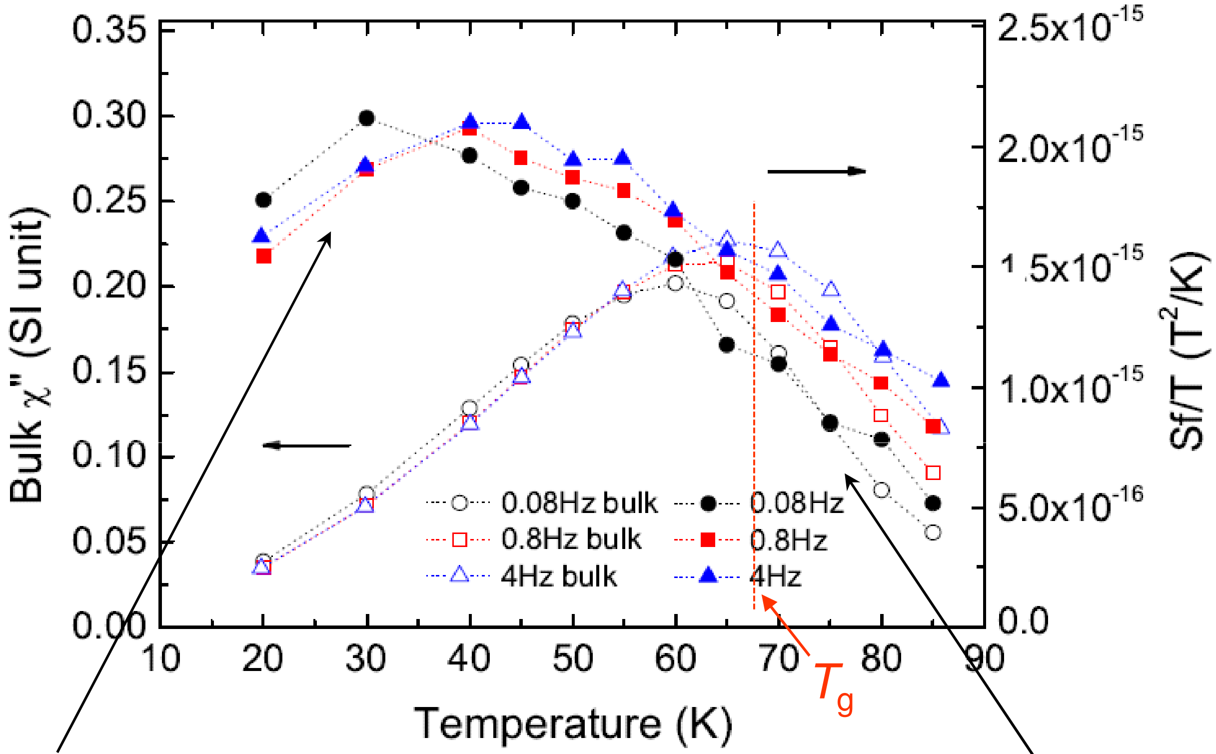




L'Hôte, Nakamae, Ladieu, Mosser, Kerlain, Konczykowski, J. Stat. Mech. P01027 (2009)

First results

γ-Fe_2O_3 nanoparticles
d=8nm,
15% volume fraction



FDT is violated below T_g

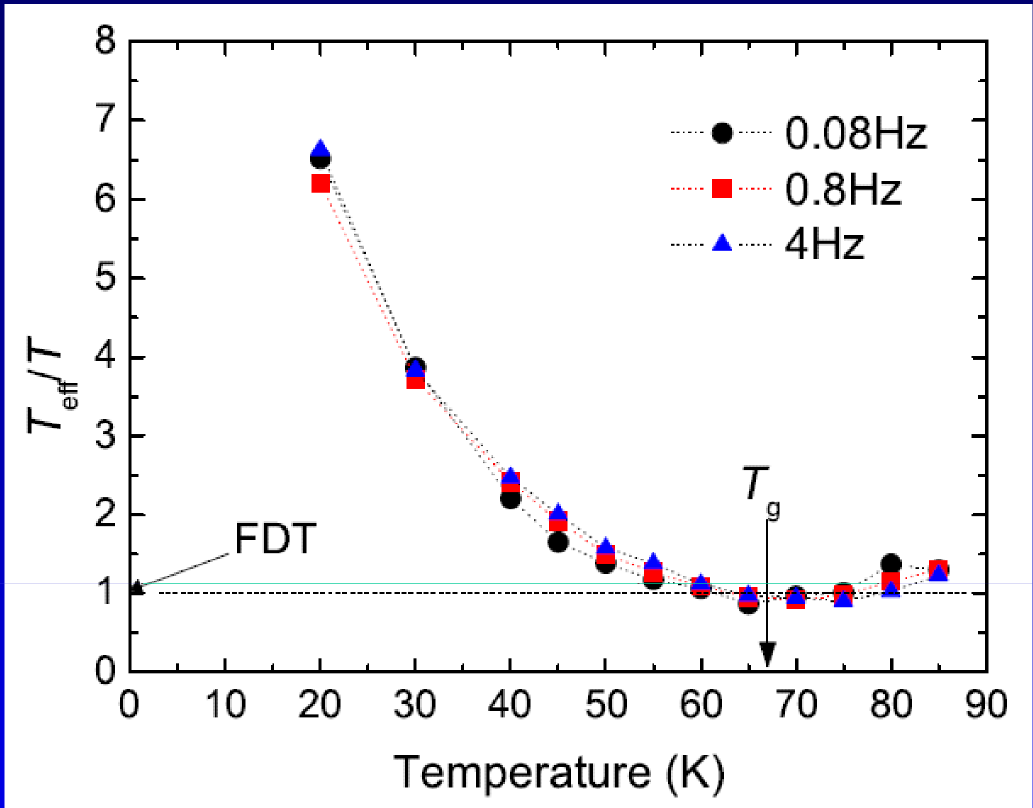
FDT is confirmed at high temperatures above T_g

χ'': Imaginary part of susceptibility

S_m: magn. noise power, f: frequency, T: temperature

$$\chi'' \propto \frac{S_m f}{T}$$

First results



In $CdCr_{1.7}In_{0.3}S_4$ spin glass:
T_{eff}/T_g from 1.5 (T/T_g=0.6) to 4 (0.9) –
opposite T-dependence ???

Komatsu, L'Hôte, Nakamae, Mosser, Konczykowski, Dubois, Dupuis, Perzynski (2010)
forthcoming paper

Ongoing experiments: t_w dependence

1. Response: slow relaxations, aging

2. Fluctuations (noise): comparison with response

2.a in magnetic systems

2.b in other glassy materials

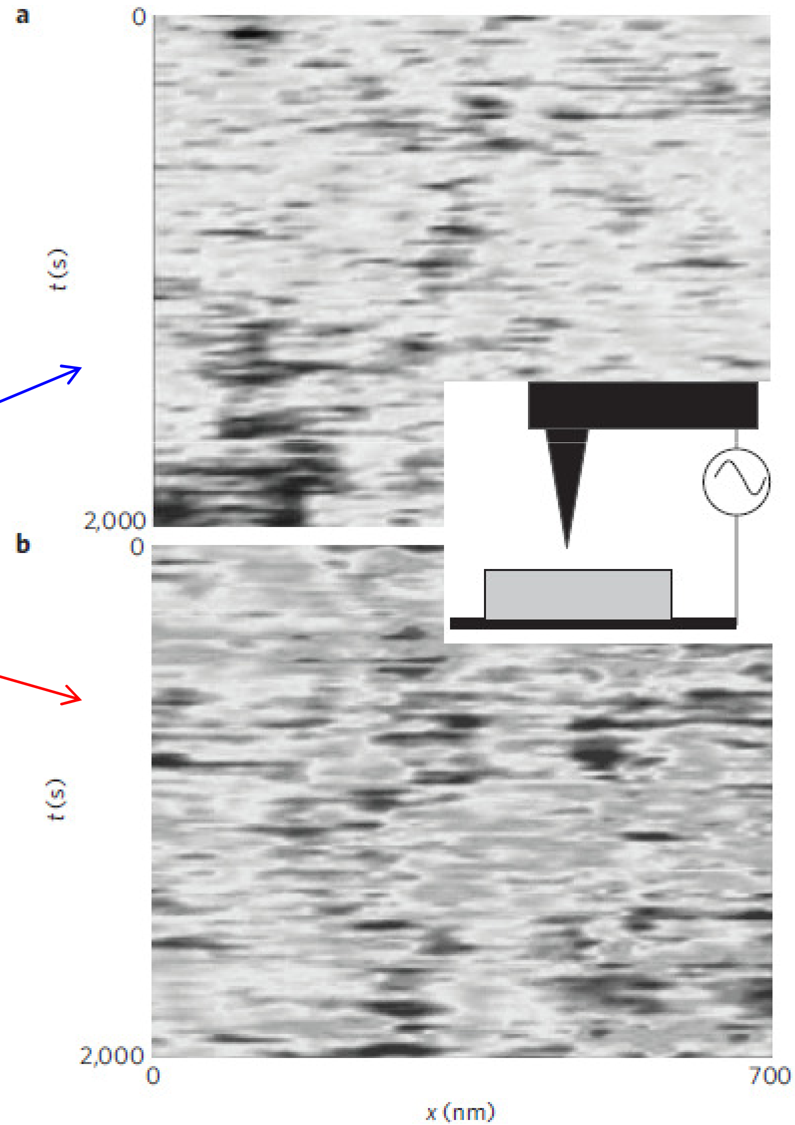
nature physics
LETTERS
PUBLISHED ONLINE: 20 DECEMBER 2009 | DOI: 10.1038/NPHYS1482

Nanoscale non-equilibrium dynamics and the fluctuation–dissipation relation in an ageing polymer glass

(Boston group)

Hassan Oukris and N. E. Israeloff*

Electric Force Microscopy on PVAc film T_g=304K
Measurement of time dependent polarization $V_p(t)$



Space/time fluctuations:

Long-lasting correlations at 301.5K

In contrast with image at 305.5K

local measurement (~100nm)
→ large polarization fluctuations

but average on sample surface (32 regions) to improve statistics
(⇔ 32 samples in parallel)

nature physics
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Nanoscale non-equilibrium dynamics and the fluctuation–dissipation relation in an ageing polymer glass

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Hassan Oukris and N. E. Israeloff*

Response to bias voltage:

$$\Delta\chi_{ex}(t) = \Delta V_P(t)/V_{d.c.}$$

Autocorrelation of the fluctuations :

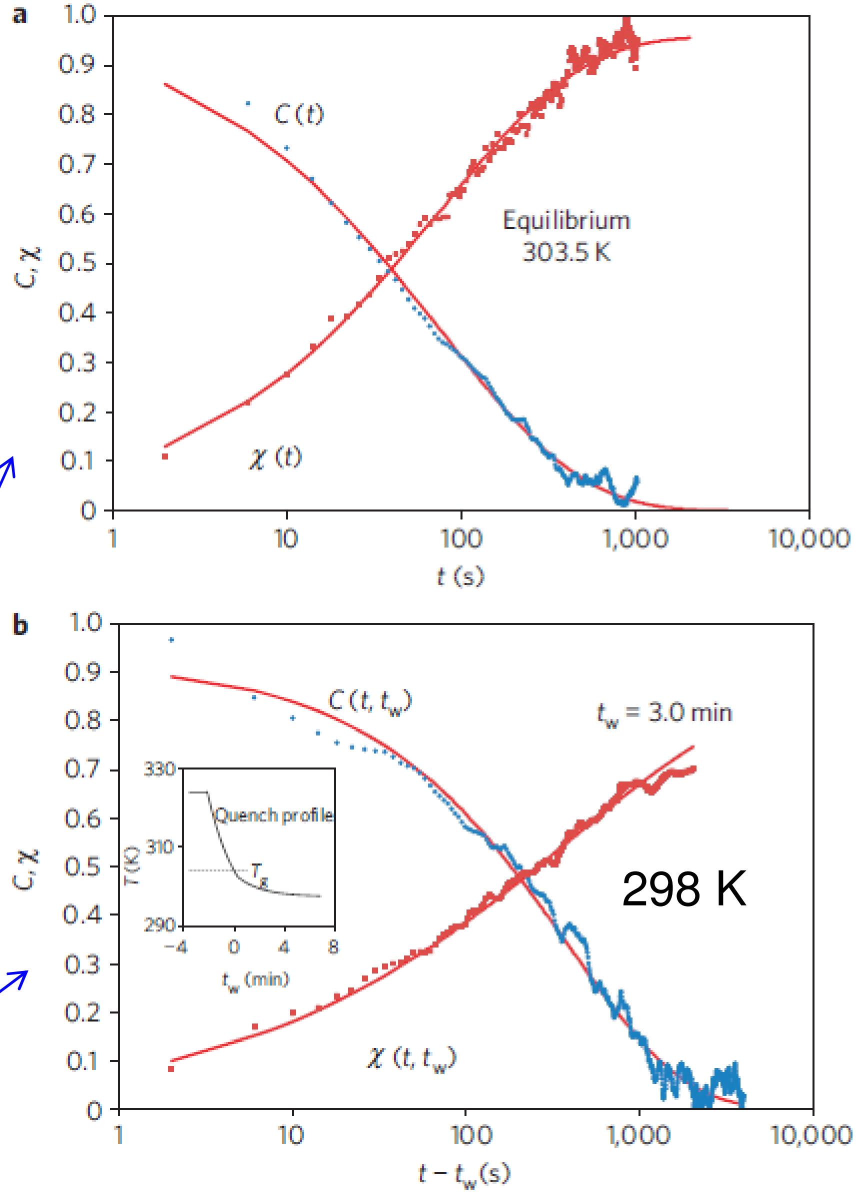
$$C(t) = \langle V_P(t)V_P(t+t')\rangle$$

Fluctuation-Dissipation Relation :

$$\Delta\chi_{ex}(t) = \frac{c_{eff}}{k_B T}[C(0) - C(t)]$$

obeyed around Tg

violated below Tg during aging



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Nanoscale non-equilibrium dynamics and the fluctuation–dissipation relation in an ageing polymer glass

(Boston group)

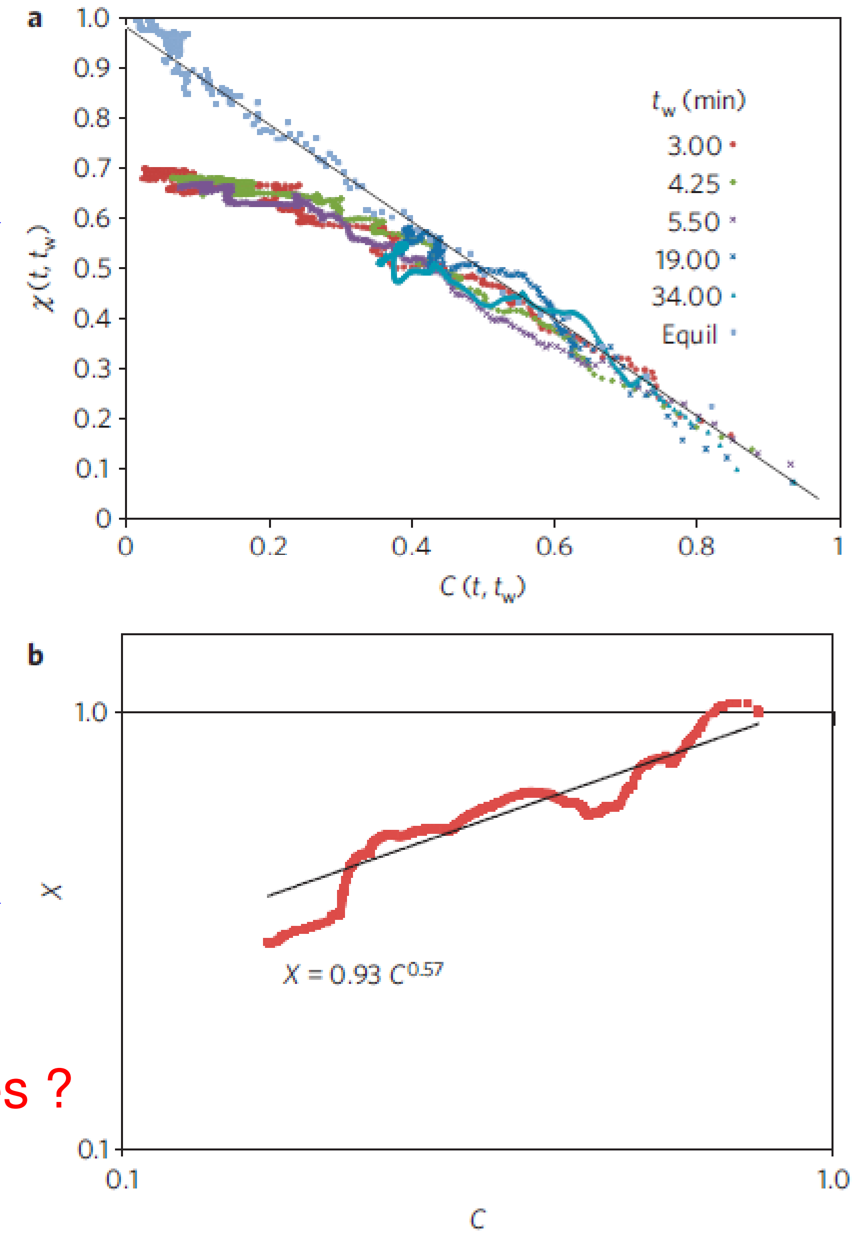
Hassan Oukris and N. E. Israeloff*

FDR graph :

The susceptibility χ is a function of only C in the aging regime
shape similar to SK class of mean-field spin-glass models

FDR-violation factor :
$X = T/T_{eff} \propto -d\chi/dC$
($X=1$ at equilibrium)

SK spin-glass: a model for polymer glasses ?



PRL 102, 130601 (2009) PHYSICAL REVIEW LETTERS

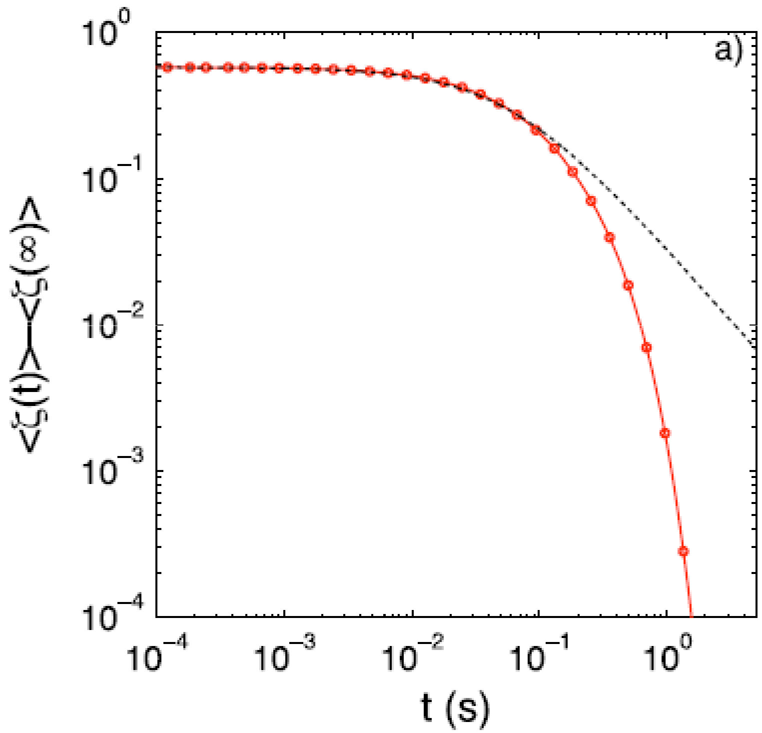
Aging and Effective Temperatures Near a Critical Point

S. Joubaud, B. Percier, A. Petrosyan, and S. Ciliberto
Université de Lyon, Laboratoire de Physique, Ecole Normale Supérieure de Lyon, CNRS,

Liquid crystal
confined between 2 plates (9µm), with surface treatment → molecules aligned in a unique direction

Fréedericksz transition:
Apply a perpendicular electric field (⇔ quench) → new alignment along the perpendicular direction (slowly over ~10s)

Measurement of the mean alignment by polarization interferometry



Relaxation of the mean alignment

Aging and Effective Temperatures Near a Critical Point

S. Joubaud, B. Percier, A. Petrosyan, and S. Ciliberto
Université de Lyon, Laboratoire de Physique, Ecole Normale Supérieure de Lyon, CNRS,

Correlation function

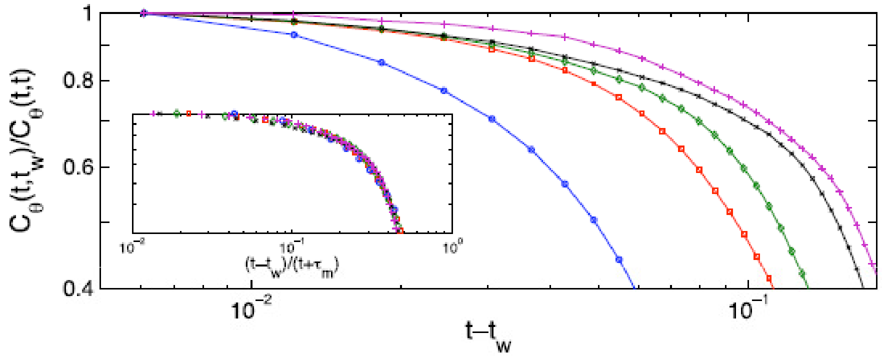


FIG. 2 (color online). (a) Correlation functions $C_\theta(t, t_w)$ as a function of $t - t_w$ for different fixed $t = 0.06$ s 0.2 s (□), 0.26 s (◇), 0.28 s (×), and 0.31 s (+) after the same quench of Fig. 1. Inset: The correlation functions have a simple master curve obtained by plotting $C_\theta(t, t_w)/C_\theta(t, t_w)$ versus $(t - t_w)/(t + \tau_m)$. Notice that $(t + \tau_m)$ is the reduced time used in Fig. 1.

FDR plot

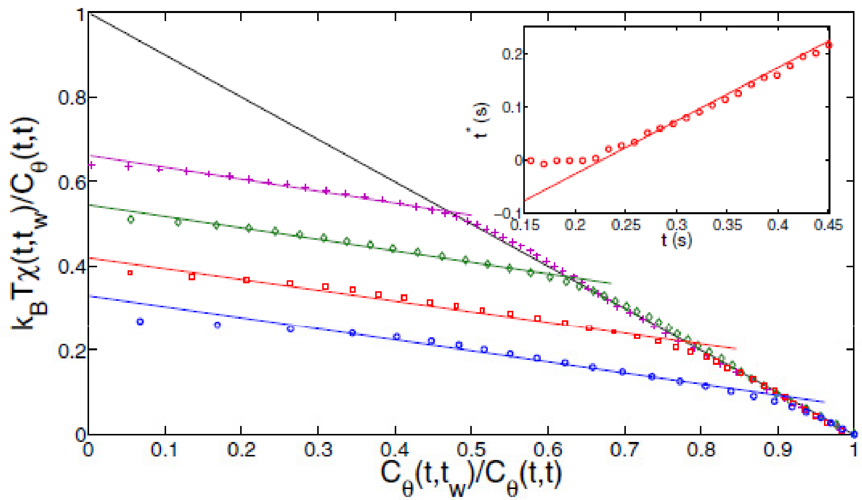


FIG. 3 (color online). (a) Parametric plot of integrated response versus correlation for $t = 0.4$ s (○), 0.5 s (□), 0.6 s (◇), and 0.7 s (+). Continuous lines are linear fits. Dark line represents the FDT at equilibrium. Inset: The characteristic time t^* is plotted as a function of t.

$$C_\theta(t, t_w) \equiv \langle \delta\theta_0(t)\delta\theta_0(t_w)\rangle$$

$$\chi(t, t_w) = \frac{X(t, t_w)}{k_B T}[C_\theta(t, t) - C_\theta(t, t_w)],$$

X=T/T$_{eff}$=0.31 (T_{eff} = effective temperature)

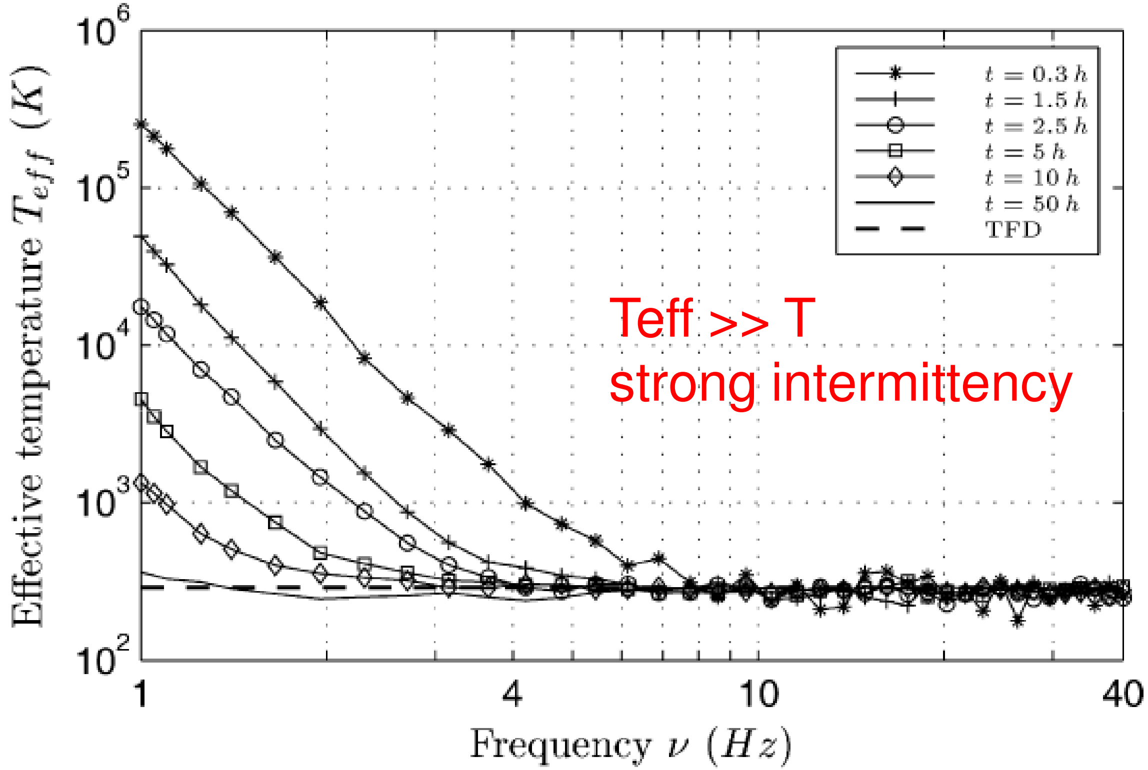
Insert: time after which $X \neq 1$
crossover from equilibrium to aging regime

FDR in colloidal glasses: intriguing results on Laponite

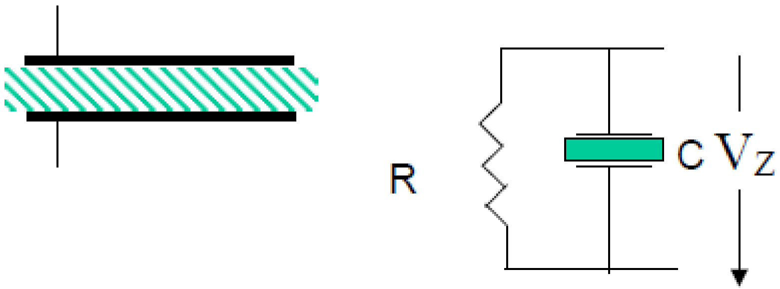
Laponite: a synthetic clay
suspension of electro-charged discs 25nmx1nm

Powder in water: → visco-elastic glass (in a few hours)

L. Bellon, S. Ciliberto / Physica D 168–169 (2002) 325–335



Dielectric measurements:
electric noise ↔ impedance measurement
Out of equilibrium FDR = generalized Nyquist relation:



$$Z(t_w, \omega) = \frac{R}{(1 + i\omega\ R\ C)}$$

$$S_Z(t_w, f) = 4\ K_B\ T_{eff}(\omega, t_w)\ Real[Z(t_w, \omega)]$$

FDR in colloidal glasses: intriguing results on Laponite

Rheological measurements: *(Bellon Ciliberto 2002)*

A rheometer sensitive to fluctuations:

- rotor inserted in a cylindrical cell, filled with laponite
- very sensitive observation of rotation by light interferometry (10^{-10} rad !)

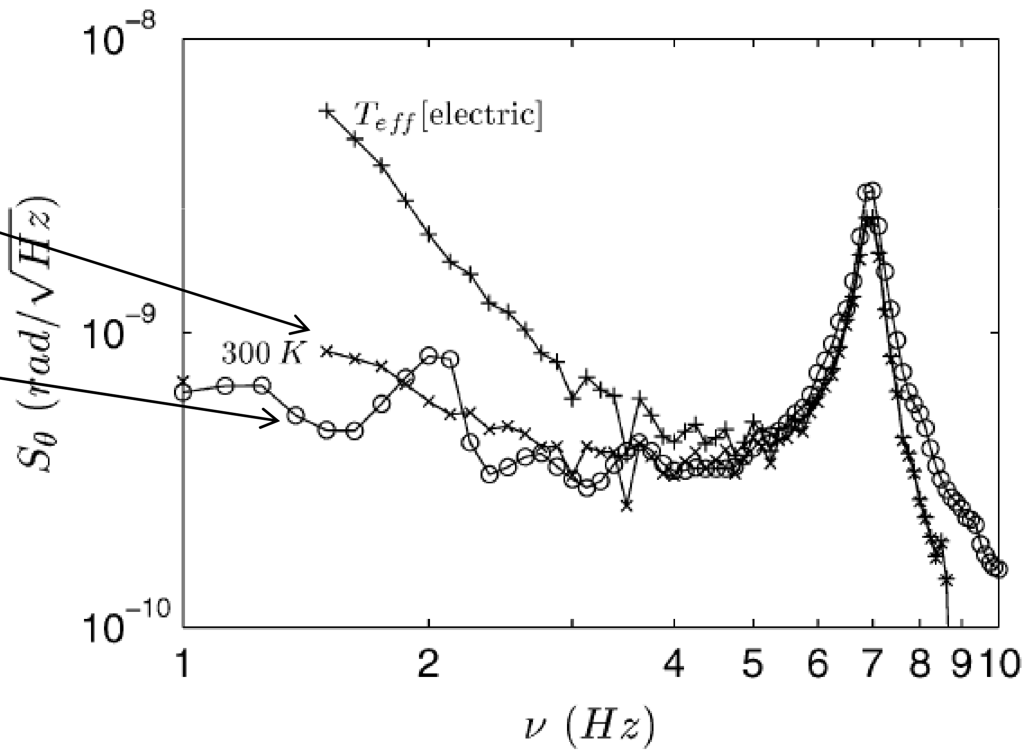
FDR prediction from viscoelastic response (x)

$$S_\theta = \frac{4k_B T}{\omega} \mathrm{Im}(\chi_{\theta\Gamma_{ext}})$$

Direct noise measurement (o)

No difference ! $T_{eff} = T$

1.5Hz peak: resonance of the table
7HZ peak: resonance of the torsion pendulum



Spectrum of the thermal fluctuations of θ

Different T_{eff} for different observables ?

FDR in colloidal glasses: intriguing results on Laponite

Micro-rheological measurements : confirmation of $T_{eff} = T$

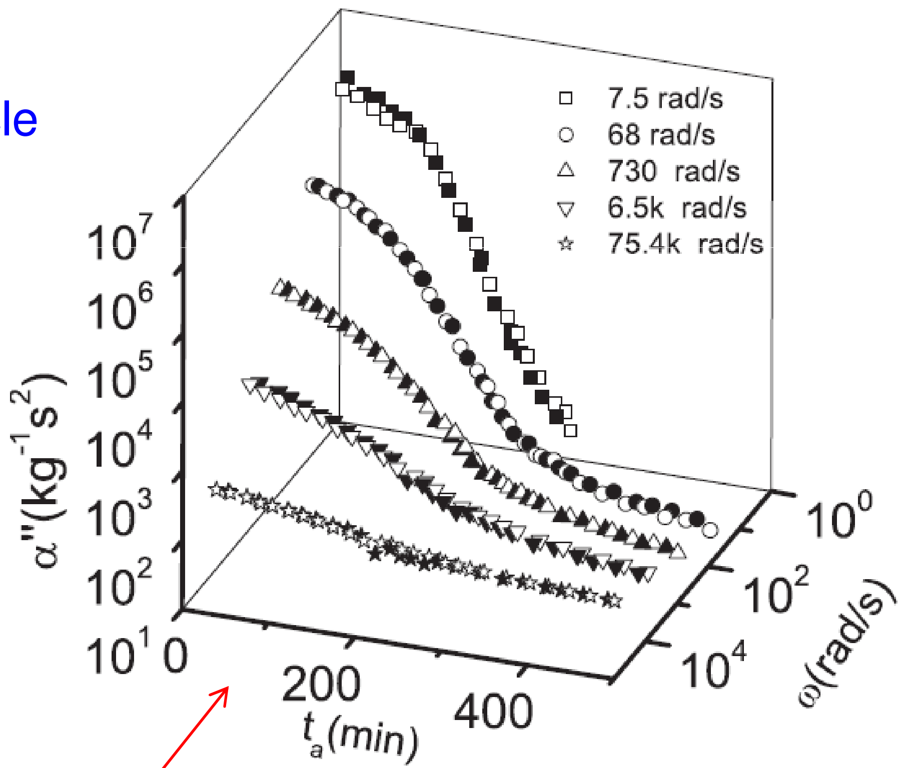
Measurement of the position (resolution 0.1nm !) of a silica bead trapped in Laponite by optical tweezers (2 lasers focused to 'diffraction-limited spots')

Active micro-rheology:
another laser drives oscillations of the particle in the optical trap → α''

Passive micro-rheology:
recording of the spontaneous fluctuations of the particle position → $\langle |x| \rangle^2$

FDR :

$$\langle |x(\omega)|^2 \rangle = \int_0^\infty \langle x(t)x(0)\rangle e^{i\omega t}\, dt = \frac{2k_B T}{\omega}\alpha''(\omega)$$



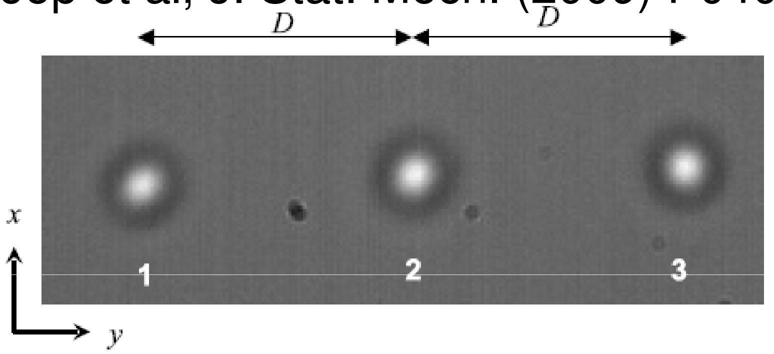
α'' from passive (open symbols) and active (solid) is identical: $T_{eff} = T$ for rheological measurements

Jabbari-Farouji et al, PRL 98, 108302 (2007) *Amsterdam and ENS Paris groups*

FDR in colloidal glasses: intriguing results on Laponite

New micro-rheological results :
simultaneous measurements of three beads in Laponite !

Jop et al, J. Stat. Mech. (2009) P04012 *ENS Lyon group*



simultaneous active and passive measurements

confirms :
(by 4 techniques)
no FDR violation
$T_{eff} = T$

Figure 2. Configuration of three optical traps separated by a distance $D = 9.3 \times 10^{-6}$ m. The bright spots in the image correspond to three probe particles of 2 μm diameter trapped by them.

Abou et al PRL 2004: observation of time varying T_{eff} (up to 1.8T) in microrheology of Laponite, explained by Jop et al by the difference in optical trap strengths in active and passive measurements

but $T_{eff} >> T$ in dielectric measurements ?

Not fully understood. Dissolution of aggregated particles → ions in the solution, effect on dielectric properties and not on rheology

Conclusions

- Spin glasses, structural and polymer glasses, gels : aging effects

  After quench, waiting time (t_w) dependence of the slow relaxations following a field change

  Scaling of the relaxation curves with ~ t / t_w^{μ} , $\mu \leq 1$ (usually…)

- Spontaneous fluctuations (noise) : can be related to response functions
  - ergodic systems : Fluctuation-Dissipation Relation (FDR)
  - aging systems : generalized FDR, « effective temperature T_{eff} »

- Magnetic systems (spin glass, superspin glass) :

  not many data – clear violations of equilibrium FDR

- Structural and polymer glasses, gels, liquid crystals :

  interesting differences between dielectric and rheological properties *(more data needed !)*

spin glasses → 'simple' models for other glassy systems ?