Low temperature dynamics of magnetic nanoparticles

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Service de Physique de l'Etat Condensé

CEA IRAMIS / SPEC (*CNRS URA 2464)*

CEA Saclay (France)

Spin Physics and Nanomagnetism, 60th birthday of E. Chudnovsky

New-York, March 2009

1. Low-temperature dynamics: thermal or quantum ?

2. Super-spin glass state

1. Low-temperature dynamics: thermal or quantum ?

2. Super-spin glass state

The beginning of the story

The starting point of my first adventure with Eugene...

PHYSICAL REVIEW LETTERS

VOLUME 60 | 22 FEBRUARY 1988 | NUMBER 8

Quantum Tunneling of Magnetization in Small Ferromagnetic Particles

E. M. Chudnovsky and L. Gunther

Department of Physics and Astronomy, Tufts University, Medford, Massachusetts 02155

(Received 29 October 1987)

The probability of tunneling of the magnetization in a single-domain particle through an energy barrier between easy directions is calculated for several forms of magnetic anisotropy. Estimated tunneling rates prove to be large enough for observation of the effect with the use of existing experimental techniques.

There were other adventures with Eugene…

South Manhattan in 1998, seen from the
Tampico driven by captain Eugene

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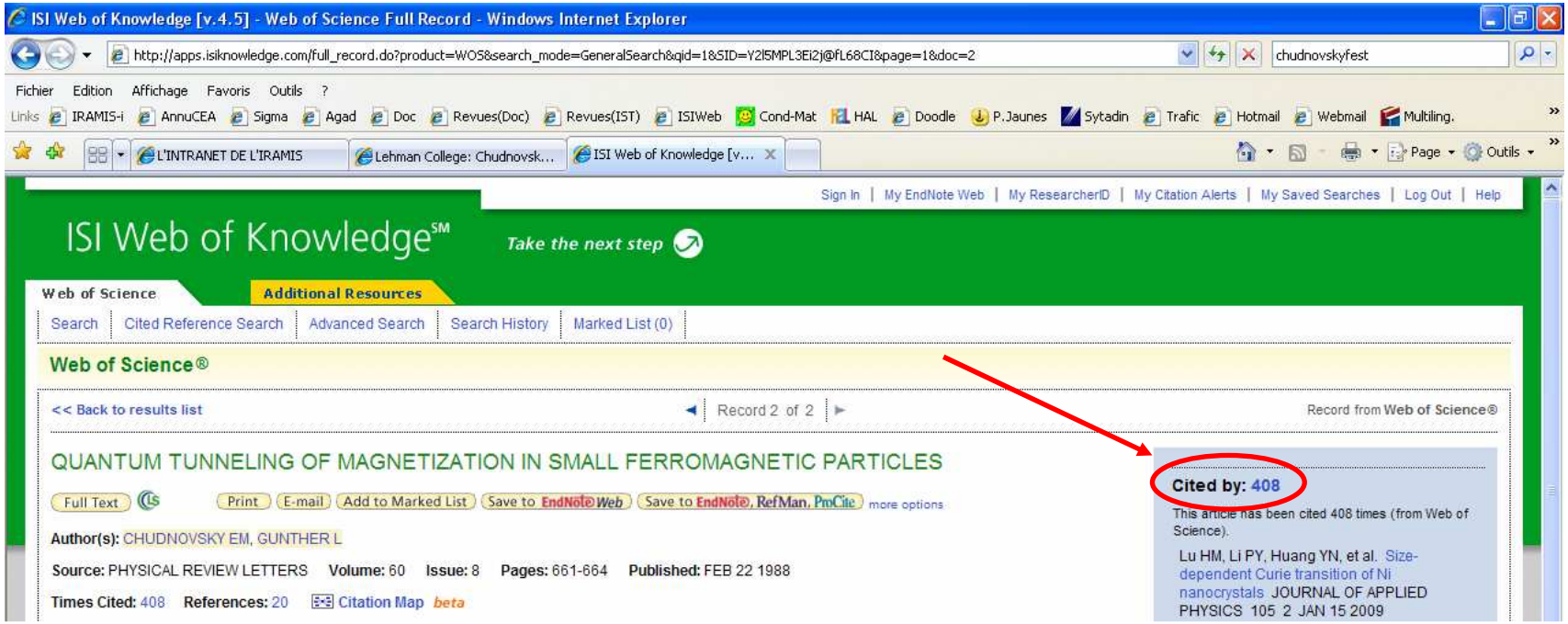
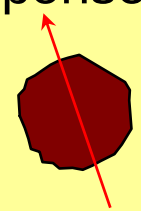


Dynamics of “super-spins”

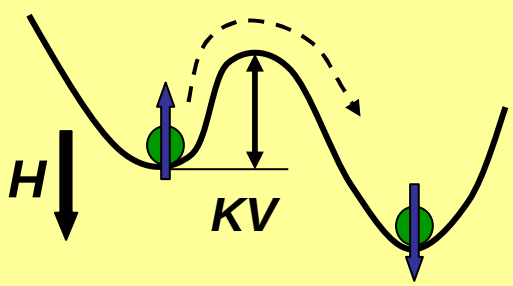
- Small enough ferromagnetic nanoparticle→ single domain
- $T \ll T_{Curie}$: response of single nanoparticle ~ response of single macro-spin

  → a ‘superspin’



- Easy axis → anisotropy barrier U~K.V

  T<<KV → blocking of magnetization



to $\theta=\theta_2$ at $\tau=0$, and then back to $\theta=0$ at $\tau=\infty$. Calculating the action for this trajectory, we obtain the WKB exponent for the tunneling from $\theta=0$ to $\theta=\pi$,

$$P \propto \exp[-(8M_0/3\hbar\gamma)(K_1/K_2)^{1/2}\epsilon^{3/2}]$$

$$=\exp(-U/k_B T_c), \tag{26}$$

where

$$T_c = 3\hbar\gamma(K_1K_2)^{1/2}\sqrt{\epsilon}/8k_B M_0. \tag{27}$$

QTM

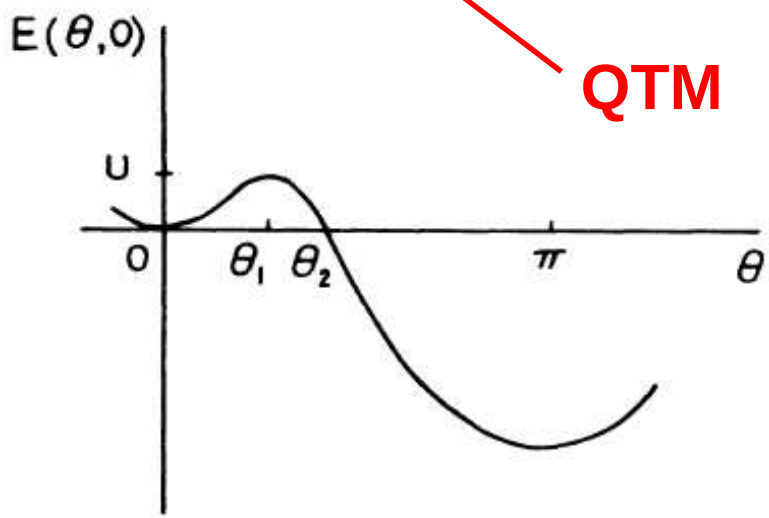
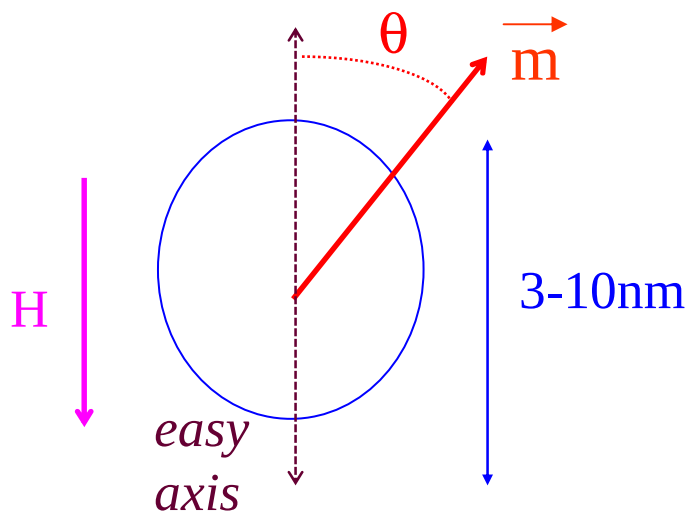


FIG. 3. Energy $E(\theta,\phi=0)$ for a model (model III) with an easy axis, a magnetic field along this axis, and transverse anisotropy.

Chudnovsky, PRL **60**, 661 (1988)

Thermal activation, Quantum tunneling

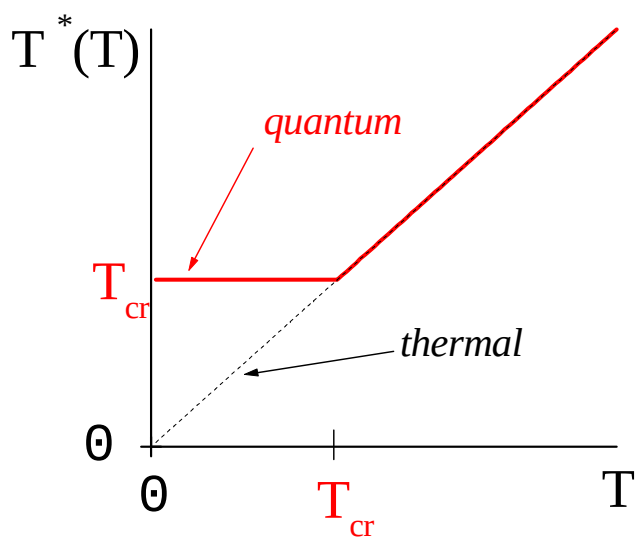


Flipping time τ of the nanoparticle magnetic moment:

$$\tau = \tau_0 \exp\left\{ \frac{U}{k_B T^*(T)} \right\}$$

$$U \sim K_{//} V$$

$T^*(T)$ ≡ "effective temperature" for the dynamics:



$$T_{cr} \propto \frac{\sqrt{K_{//} K_{\perp}}}{M_s}$$

E.M. Chudnovsky & L. Gunther,
PRL 60 (1988) 661

Observation of macroscopic quantum tunneling of magnetization in nanoparticles ?

1. In nanoparticle assemblies:

many difficulties related to particle size distribution (to be discussed in this talk)

2. In *individual* nanoparticles :

many difficulties also… but nice results

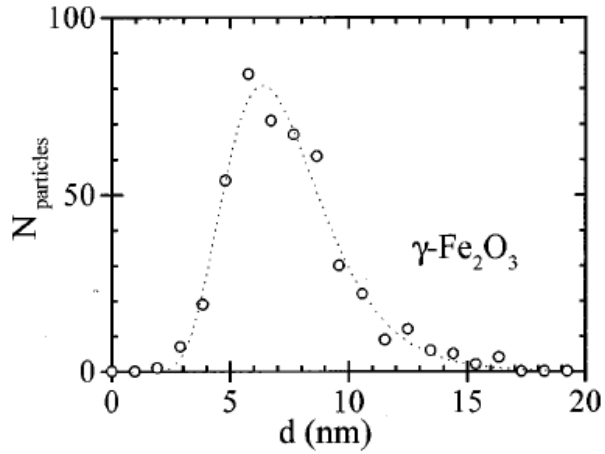


FIG. 1. Histogram of the γ-Fe_2O_3 particle diameters, as observed in TEM imaging (symbols). 454 particles have been

VOLUME 79, NUMBER 20 PHYSICAL REVIEW LETTERS 17 NOVEMBER 1997

Macroscopic Quantum Tunneling of Magnetization of Single Ferrimagnetic Nanoparticles of Barium Ferrite

W. Wernsdorfer,[1] E. Bonet Orozco,[1] K. Hasselbach,[2] A. Benoit,[2] D. Mailly,[3] O. Kubo,[4] H. Nakano,[5] and B. Barbara[1]

[1]*LL Néel, CNRS, B.P. 166, 38042 Grenoble, France*
[2]*CRTBT, CNRS, B.P. 166, 38042 Grenoble, France*
[3]*LMM, CNRS, 196 Avenue H. Ravera, 92220 Bagneux, France*
[4]*Toshiba Corporation, 8 Shinsugita-cho, Isogo-ku, Yokohama 235, Japan*
[5]*Toshiba Glass Corporation, Yoshida-cho, Haibara-gun, Shizuoka 421, Japan*

Magnetic viscosity measurements

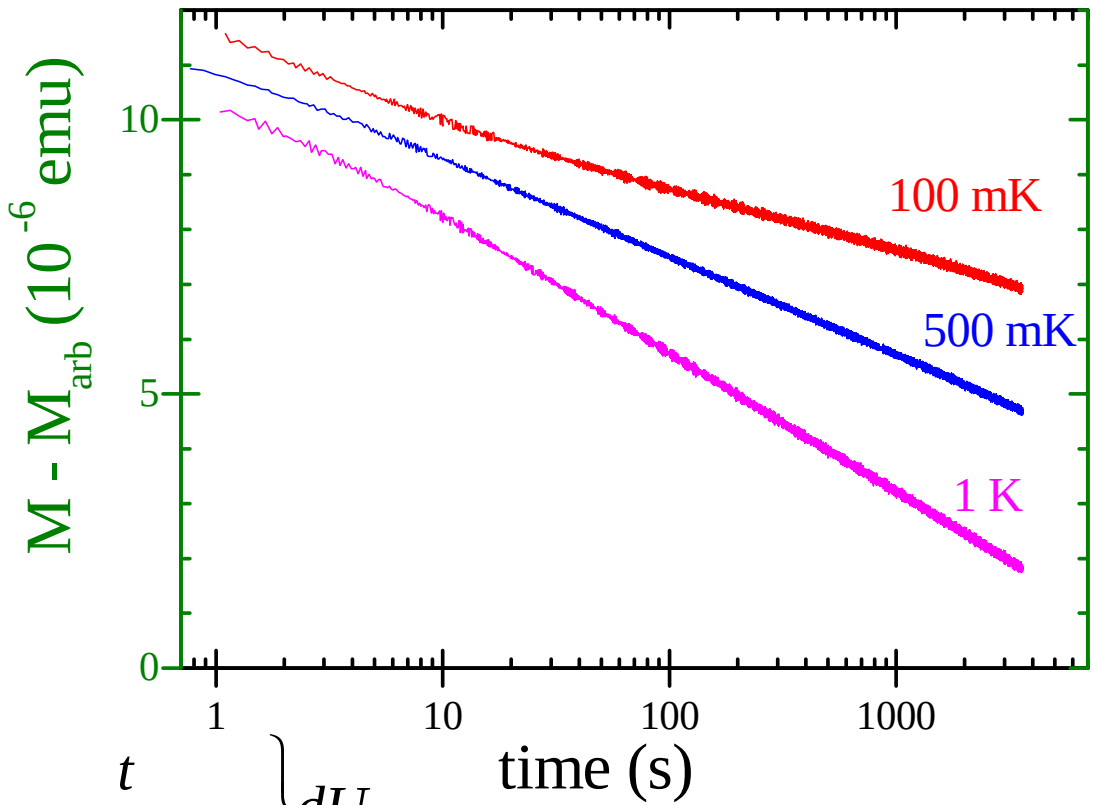
H=60 Oe, $T>T_b$

↓

Field-cool down to $T_0<T_b$

↓

H=0 at t=0 s



U=KV

Given $P(U)m_i(U)$,

$$M(t,T)=\int_0^{\infty} P(U)m_i(U)\exp\left\{-\frac{t}{\tau(U,T^*(T))}\right\}dU$$

Magnetic viscosity: $$S(T)=-\frac{\partial M(t,T)}{\partial \ln t}\approx k_B T^* P(U_c)m_i(U_c)$$ with $U_c=k_B T^* \ln\left(\frac{t}{\tau_0}\right)$

nature distribution

Nonthermal viscosity in magnets: Quantum tunneling of the magnetization (invited)

J. Tejada, X. X. Zhang, and Ll. Balcells
Department Física Fonamental, Universitat de Barcelona, Diagonal, 647, 08028 Barcelona, Spain

6709 J. Appl. Phys. **73** (10), 15 May 1993

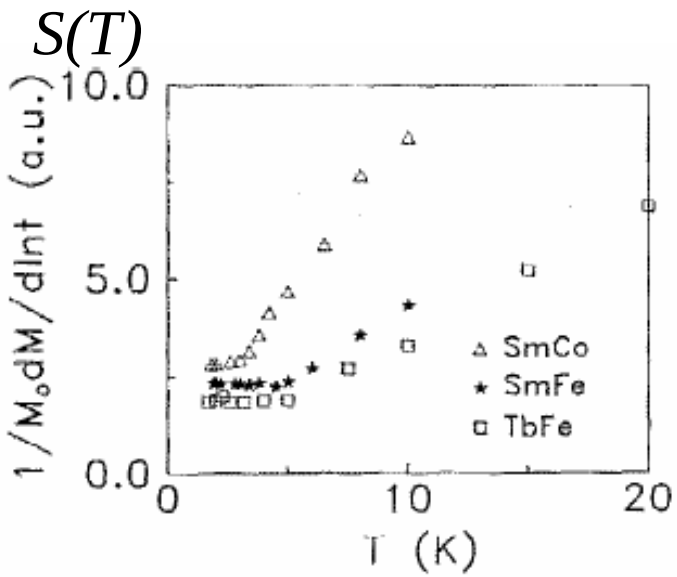


FIG. 10. Temperature dependence of the magnetic viscosity $S(T)$ for the three random magnet samples.

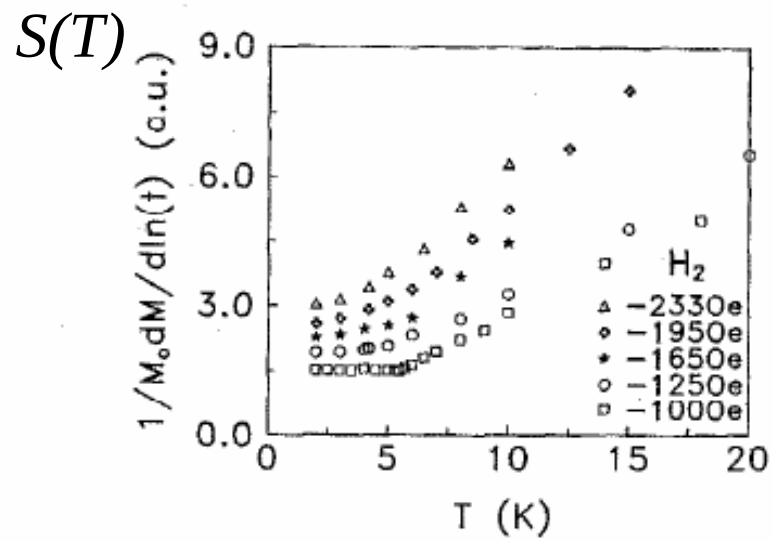


FIG. 12. Variation of the $S(T)$ and T_c with the value of the applied magnetic field H_2. In all cases $H_1 = 100$ Oe.

Low-T anomalies observed in many magnetic systems : quantum origin ?

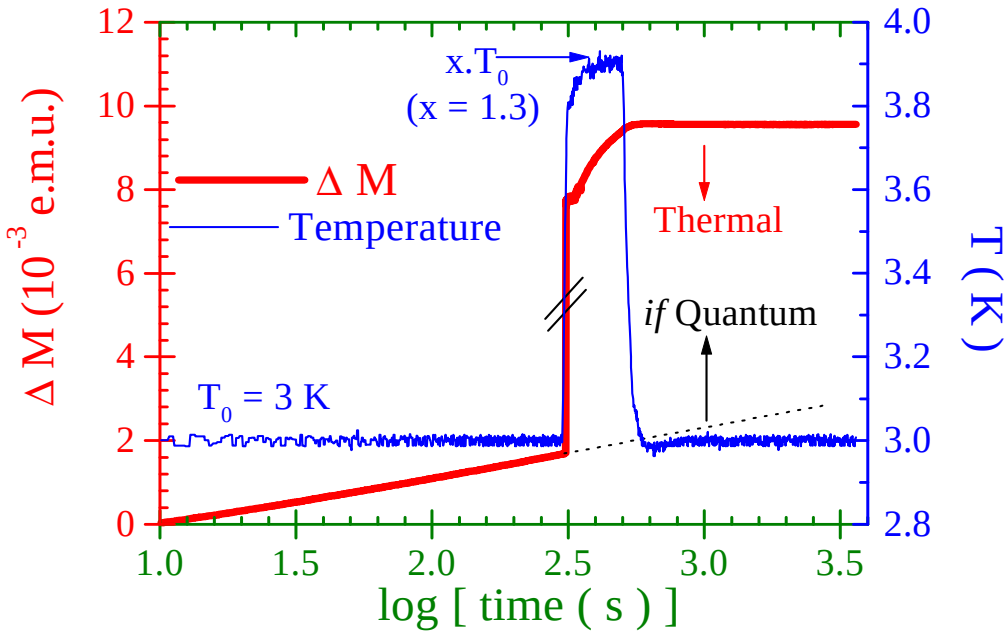
$$S(T) = -\frac{\partial M(t,T)}{\partial \ln t} \approx k_B T^* \; P(U_c) m_i(U_c)$$

nature distribution

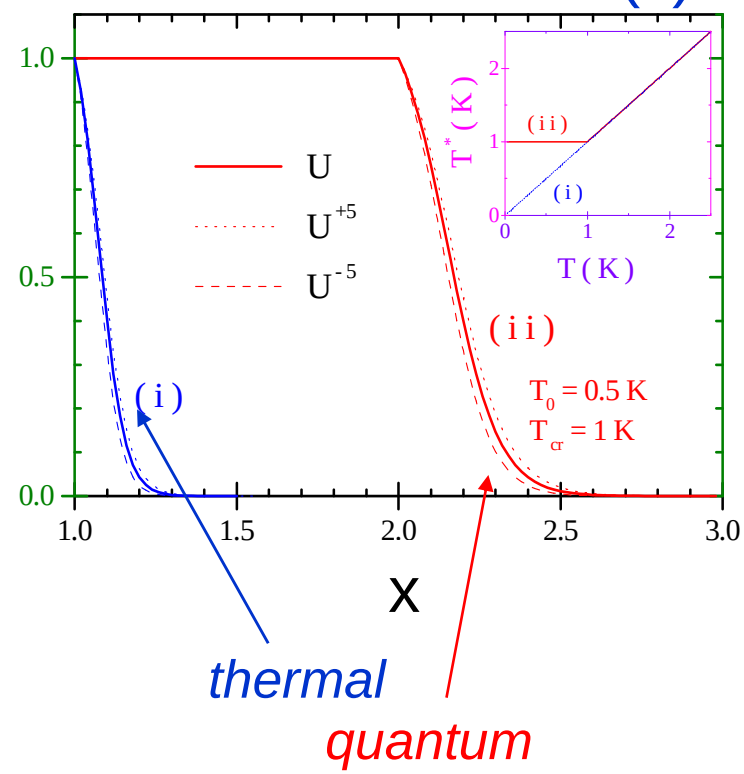
Eugene Chudnovsky in Barcelona, actively participating in the experiments on macroscopic Quantum Tunneling

A new method: the “Residual Memory Ratio” (RMR)

Relaxation with a heating cycle



Calculated RMR (x)



We define: $$RMR(x) \equiv \frac{S(T_0, x)}{S(T_0)}$$

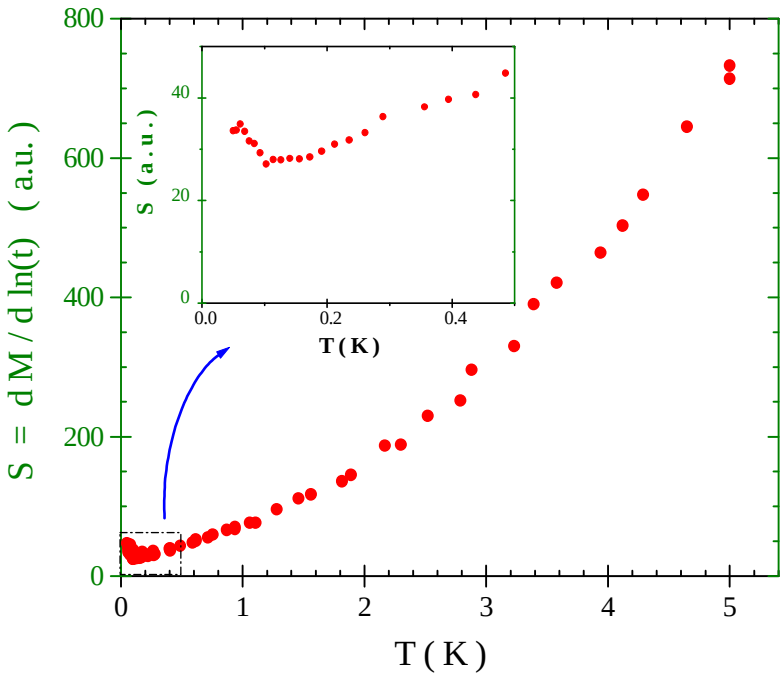
RMR is sensitive to $T^*(T)$ while being extremely insensitive to other poorly known quantities.

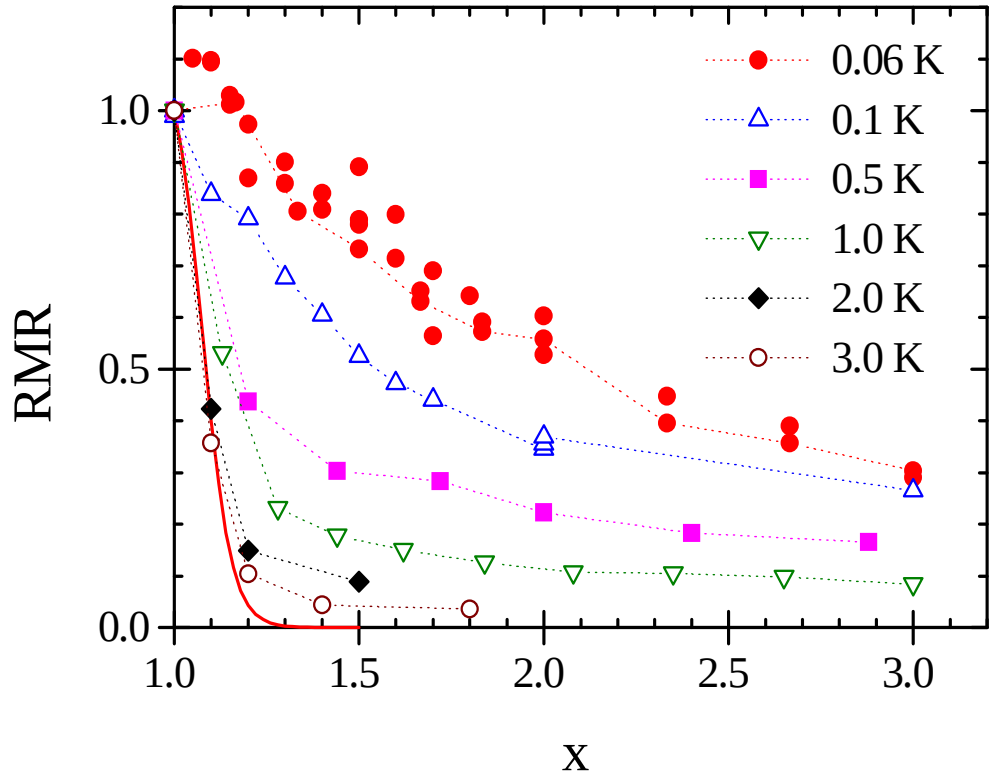
Sappey et al, *Europhys. Lett.* **37**, 639 (1997)

Maghemite nanoparticles

γ-Fe_2O_3
d~7nm, σ_d=0.3
x_v=0.043%, in silica

estimated T_{cr}~0.1 K





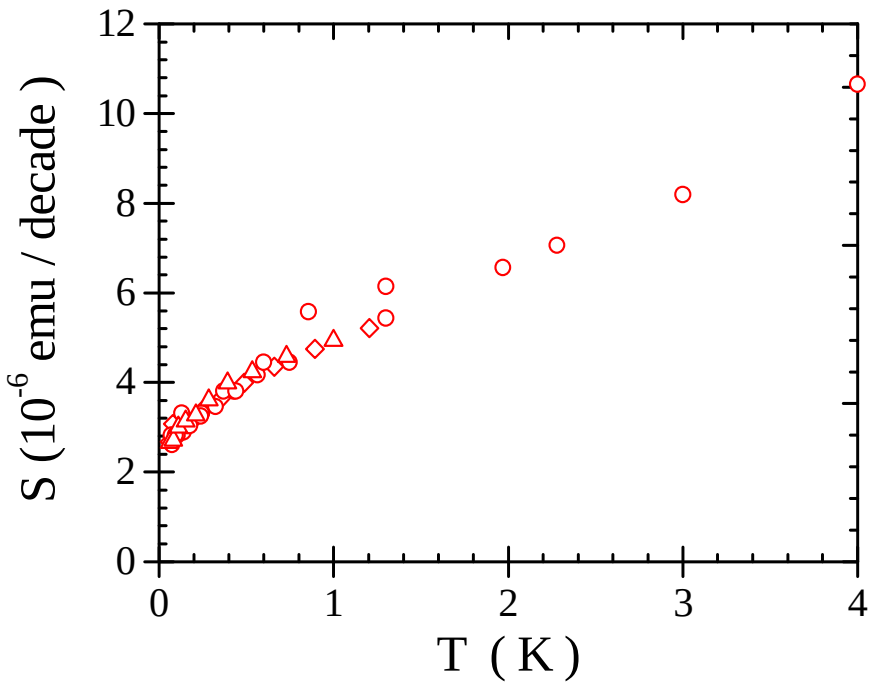
Sappey et al, EPL **37**, 639 (1997) and JMMM **221**, 87 (2000)

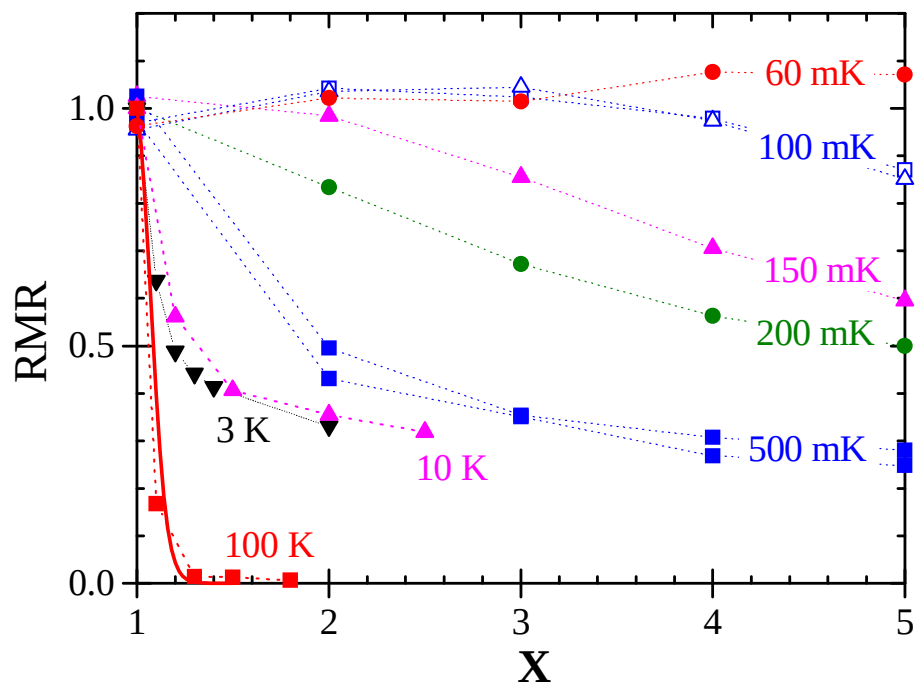
- Clear anomalies below 0.5K
- Intermediate between pure quantum and thermal
- Distribution of crossover temperatures

Cobalt ferrite nanoparticles

$CoFe_2O_4$
d ~ 5.7 nm, σ_d=0.2
x_v=2.3%, in water

estimated T_{cr}~5K





Sappey et al, JMMM **221**, 87 (2000)

- Clear QTM-like for T<150mK (flat RMR)
- But at T~3-10K RMR does not go to zero: more complex behaviour than simple model (defects ? surface effects ?)

Eugene Chudnovsky in Barcelona, discussing with the experimentalists

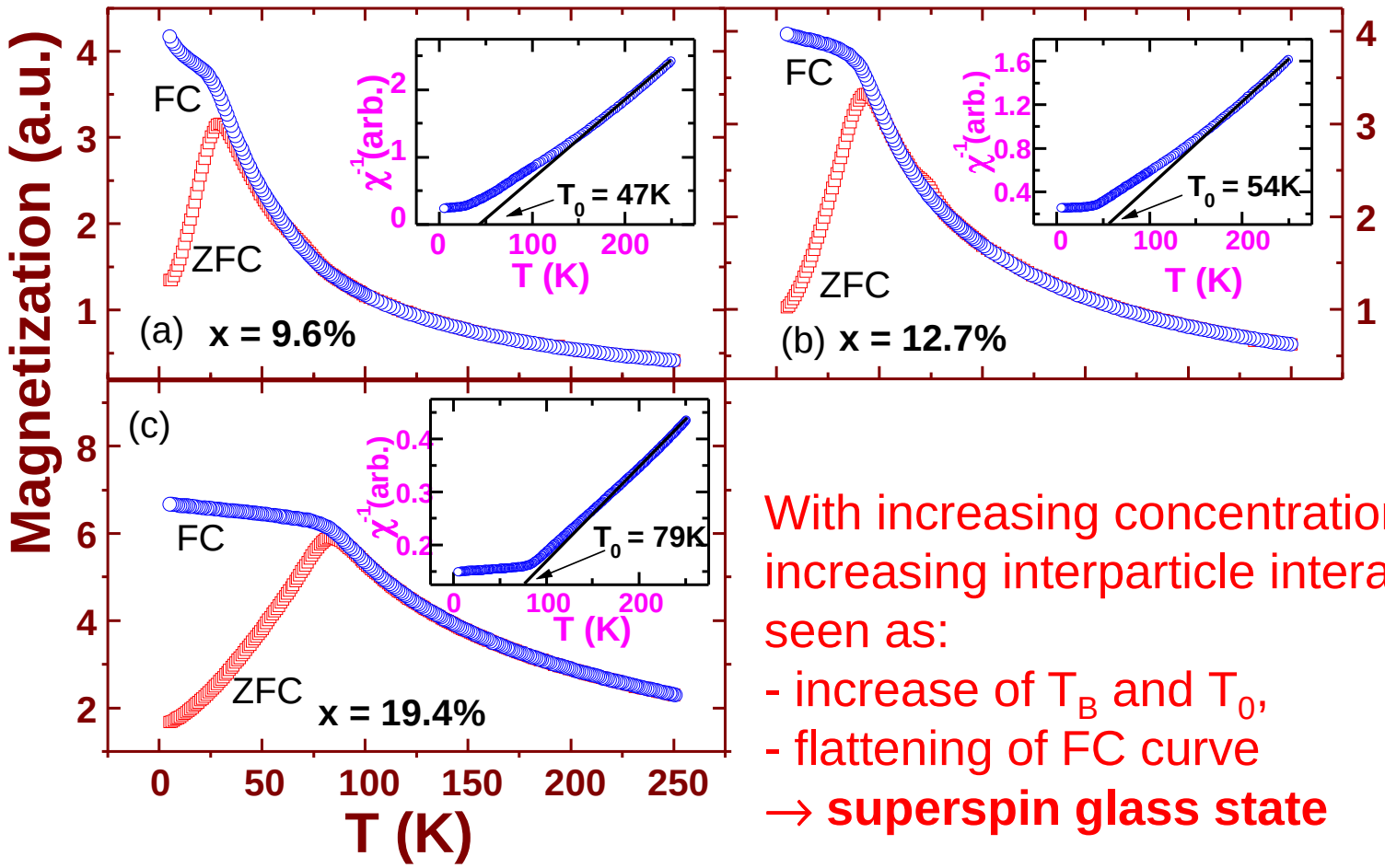
1. Low-temperature dynamics: thermal or quantum ?

2. Super-spin glass state

Ineracting Co nanoparticles in Ag matrix: superspin glass state

(Co_xAg_{1-x} , metal matrix → RKKY interactions)

X.X. Zhang group, Phys. Rev. B75, 014415 (2007)

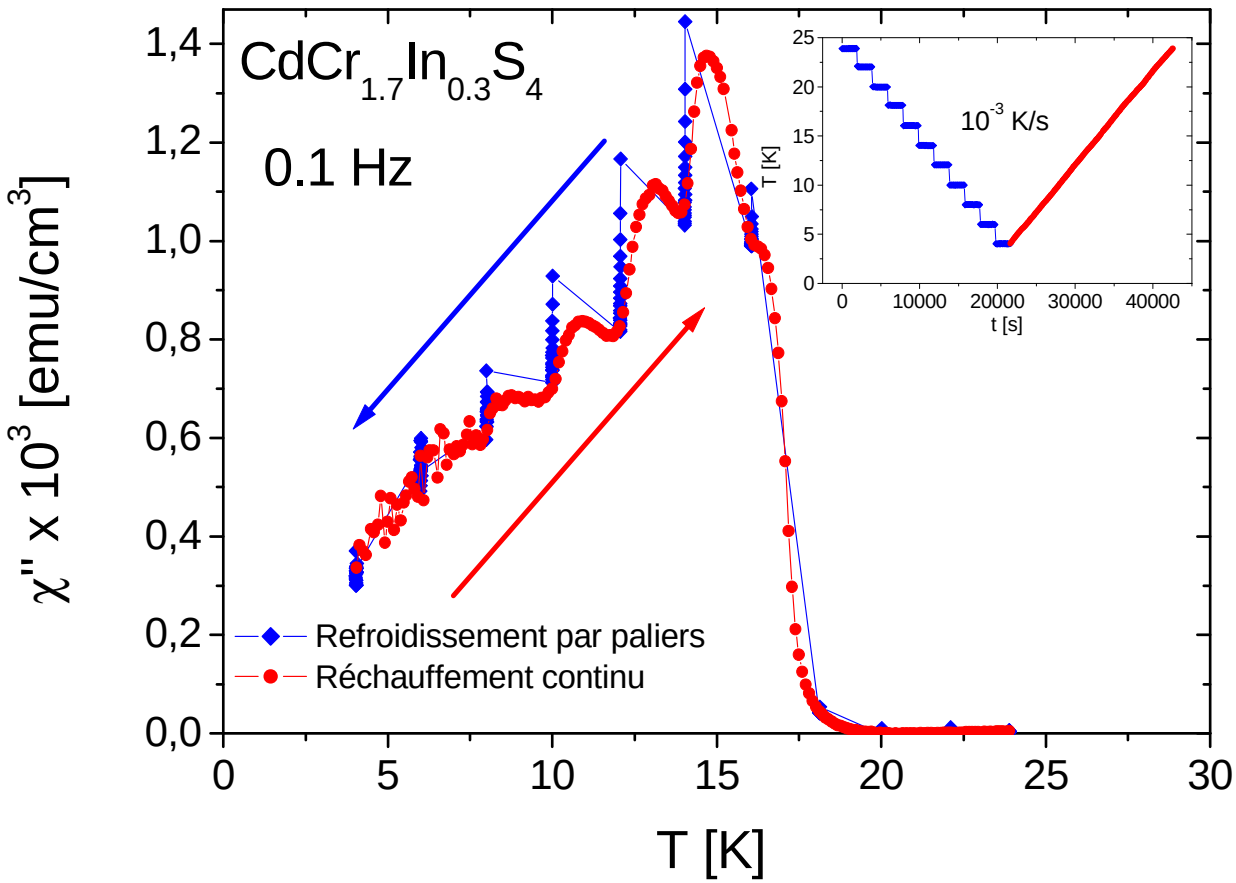


With increasing concentration x : increasing interparticle interactions, seen as:

- increase of T_B and T_0,
- flattening of FC curve

→ **superspin glass state**

Spin glass: aging, rejuvenation and memory effects

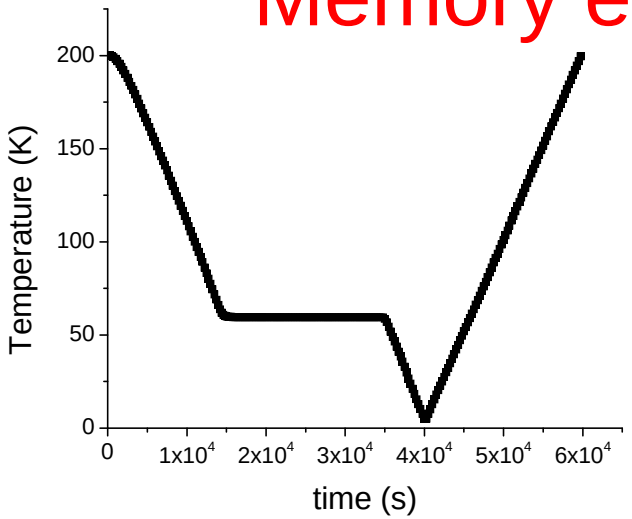


χ''-relaxation:
aging

T↓ : *rejuvenation*
T↑ : *memory*

« memory dips » experiments:
Uppsala / Saclay *PRL* **81**, 3243 (1998)
Bouchaud et al, *Phys. Rev. B* **65**, 024439 (2001)
See details and references in e.g. cond-mat/0603583

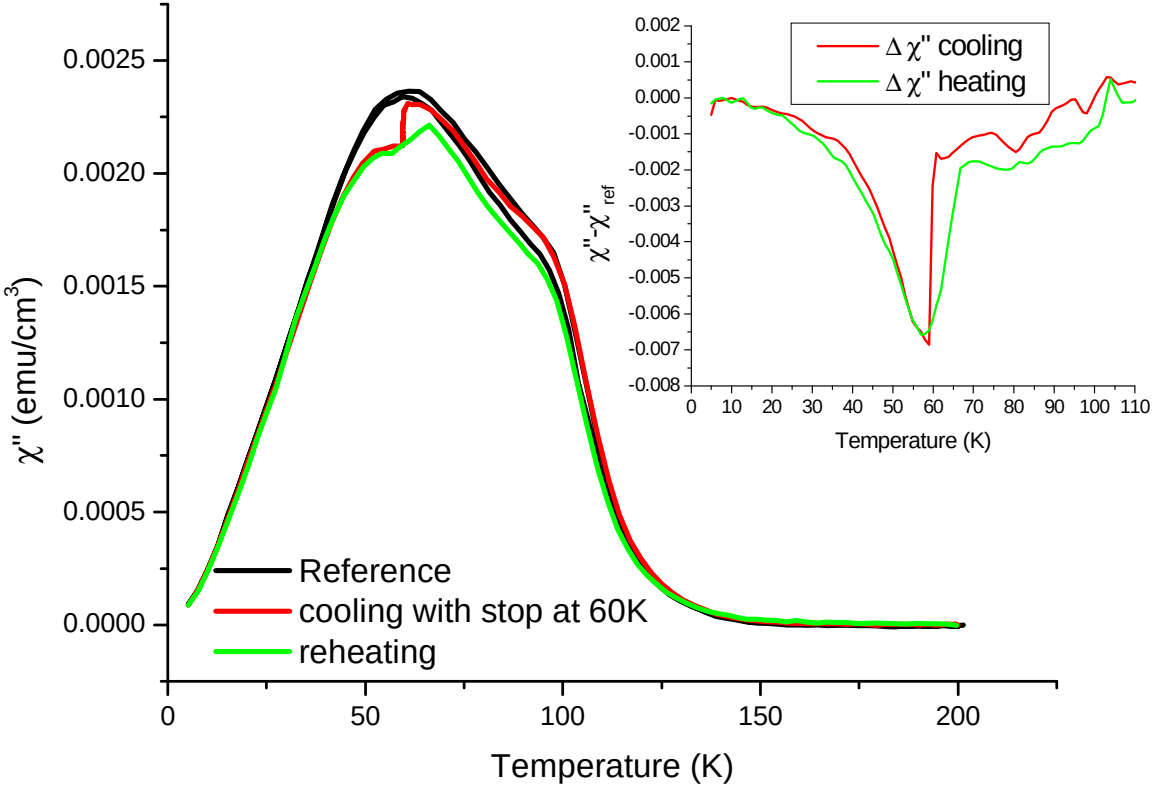
Memory effect in a superspin glass



γ-Fe_2O_3 nanoparticles in water

d=8.5nm

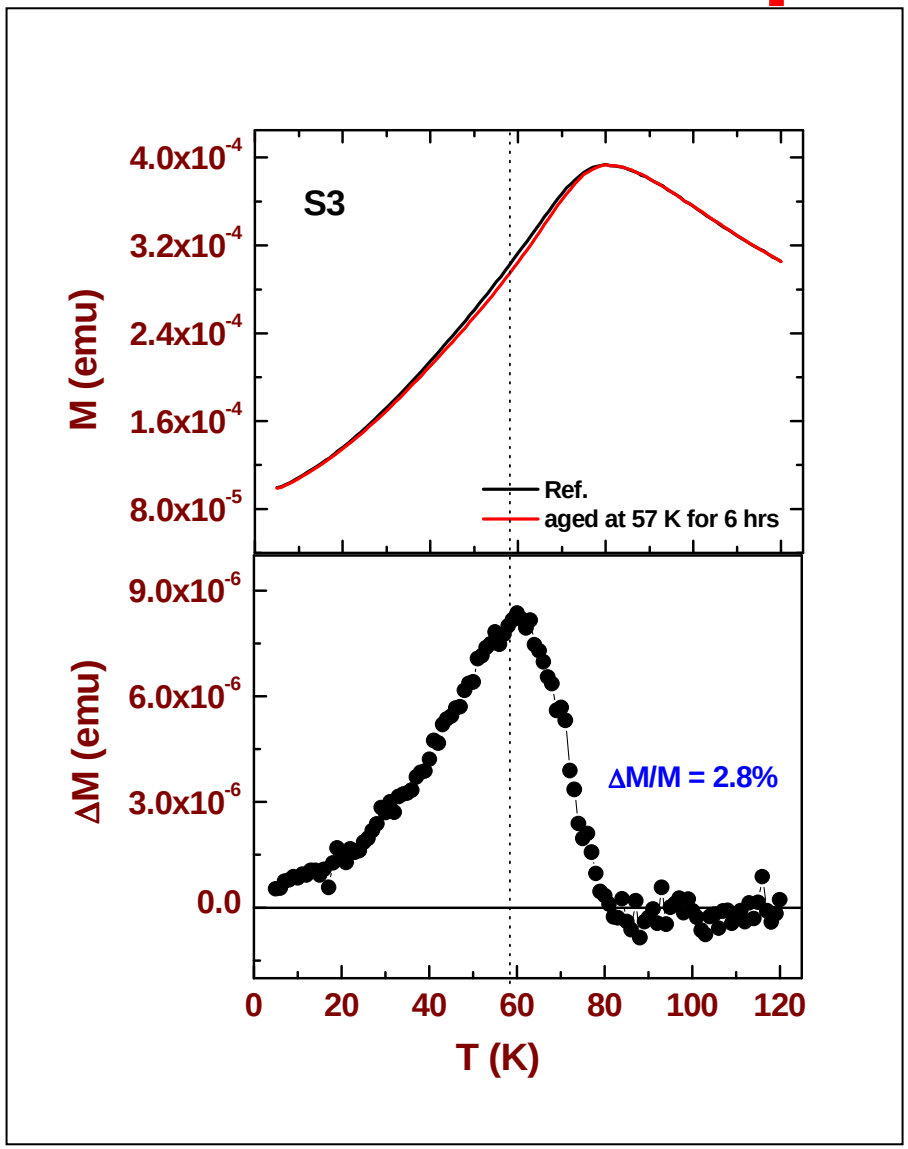
Φ=35%



- no visible rejuvenation
- but clear memory effect

V. Dupuis et al, AIP Conf. Proc. 832, 295 (2006)

Memory effect in Co nanoparticles



$Co_{20}Ag_{80}$ granular film

aging at $T_w \approx 0.7\ T_B$ for 6 hrs

$$\Delta M/M \approx 2.8\%$$

J. Du et al, Phys. Rev. B **75**, 014415 (2007)

(*X.X. Zhang group, HKUST*)

PHYSICAL REVIEW B **71**, 104404 (2005)

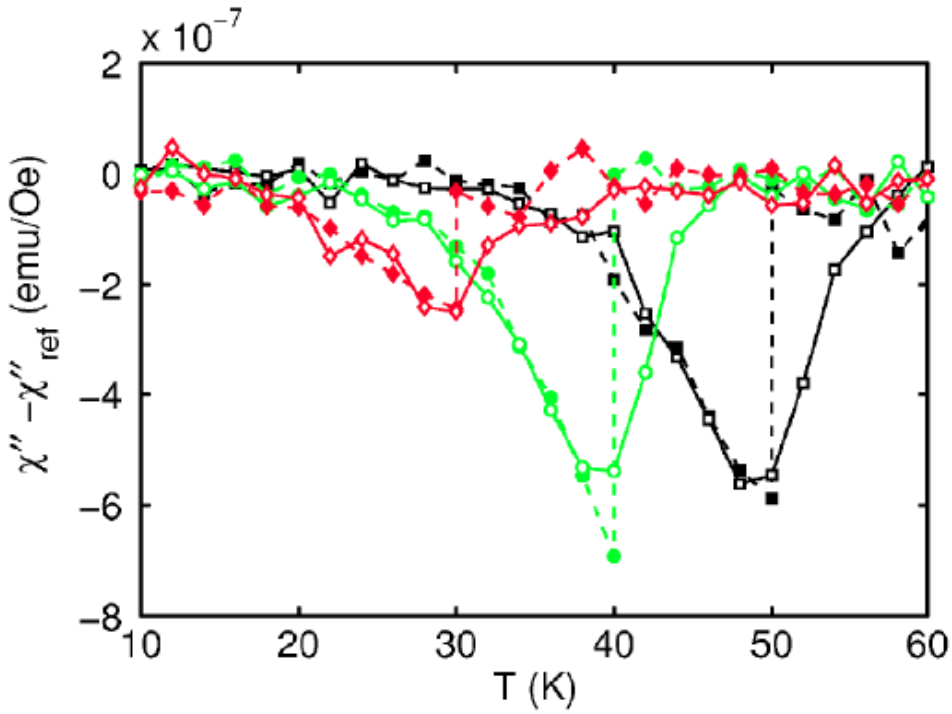
Absence of strong rejuvenation in a superspin glass

P. E. Jönsson,[1] H. Yoshino,[2] H. Mamiya,[3] and H. Takayama[1]

Concentrated Fe_3N nanoparticle system

Clear T-specific memory effect, although not so well-marked as in atomic SG's

Time scales are different:
SG $\tau_0 \approx 10^{-12}$ s
SSG $\tau_0 \approx 10^{-9}$ s or much longer
Longer $\tau_0 \rightarrow$ shorter accessible time scale t_{exp}/τ_0

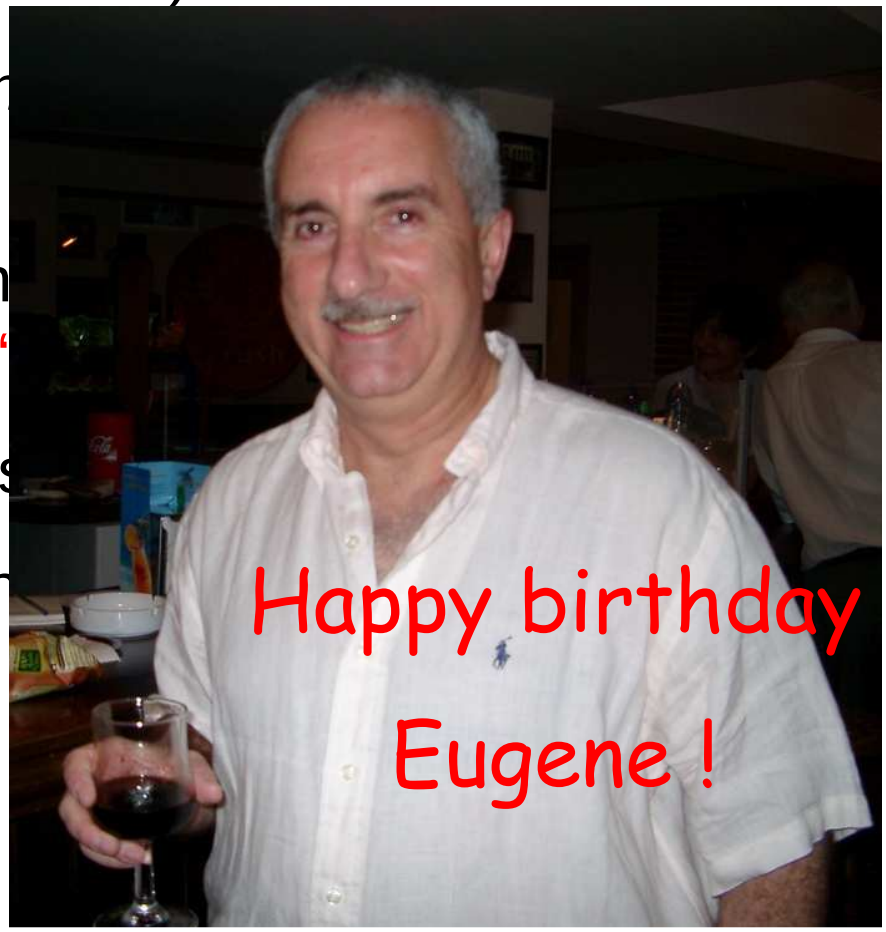


→ bridge the time gap between *real* and *numerical* spin glasses

see Wandersman et al, EPL **84**, 37011 (2008)

Conclusions

- Despite the problem of size distribution, strong deviations from thermal activation have been identified in assemblies of magnetic nanoparticles (RMR method)

  → *macroscopic Quantum Tur*

- Concentrated suspensions of (in
  nanoparticles may behave as a "
- Memory effects are seen like in s
- Will bridge the gap between spin
  experiments