Slow Dynamics of Magnetic Nanoparticle Systems: Memory effects

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Outline

- Introduction
  
  Motivation

- Memory Effect in Noninteracting Nanoparticle System
  (Superparamagnetism and Blocking)

- Memory Effect in Interacting Nanoparticle System
  (Spin-Glass Dynamics)

- Other Spin-Glass Behavior in Interacting Nanoparticle System

- Summary
Motivation

Recent experiments by Sun et al. [Phys. Rev. Lett. 91, 167206 (2003)] show peculiar memory effects in a nanoparticle sample. Sun et al. claim that these memory effects give evidence for spin-glass dynamics in the sample. Is it really true?
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- Summary
Memory effect in noninteracting nanoparticle systems (superparamagnetism and blocking)

-- Master equation approach --

The Master-equation approach

One magnetic nanoparticle with uniaxial anisotropy in field applied along the anisotropy direction

Two states description:
Occupation probability of state 1

\[ p_1(t) (= 1 - p_2(t)) \]

Master equation for \( p_1(t) \)

\[
\frac{d}{dt} p_1(t) = p_2(t) W_{2 \rightarrow 1}(t) - p_1(t) W_{1 \rightarrow 2}(t) \\
= -[W_{2 \rightarrow 1}(t) + W_{1 \rightarrow 2}(t)] p_1(t) + W_{2 \rightarrow 1}(t)
\]
transition probability

\[ W_{1\rightarrow 2}(t) = \tau_0^{-1} \exp[(KV + H(t)M_s V)/T(t)] \]
\[ W_{2\rightarrow 1}(t) = \tau_0^{-1} \exp[(KV - H(t)M_s V)/T(t)] \]

solution

\[ p_1(t) = \exp[ - \int_0^t dt' W_{2\rightarrow 1}(t') + W_{1\rightarrow 2}(t')] \]
\[ \times \left( \int_0^t dt' W_{2\rightarrow 1}(t') \exp[ \int_0^{t'} dt'' W_{2\rightarrow 1}(t'') + W_{1\rightarrow 2}(t'')] + C \right) \]

magnetization

\[ M(t) = M_s V [p_1(t) - p_2(t)] = M_s V [2p_1(t) - 1] \]

We can calculate \( M(t) \)
for any experimental protocol specified by \( \{H(t), T(t)\} \)
from a given initial condition
such as \( p_1(t=0) = C = 1/2 \), or \( M(0)=0 \)
Average over Volumes of Nanoparticle

\[ P(V) = \frac{1}{\sqrt{2\pi} \gamma V} \exp\left[-\ln(V)^2/(2\gamma^2)\right] \quad (\gamma = 0.6) \]

\[ \left\langle M(t) \right\rangle_{\text{ave}} = \int dV P(V) M(V, t) \]
Results: Memory Effect

Experiment (Sun et al)

Calculation
$$\langle M(t) \rangle_{\text{ave}} = \int dV P(V) M(V, t)$$
Memory effects

Experimental Protocol:
Cooling: stops at constant temperature (+ field changes)
Reheating: memory effects

- Noninteracting nanoparticle systems
  - Wide distribution of relaxation times, or blocking temperatures

- Spin glasses, Strongly interacting nanoparticle systems

?
Experiments on a strongly interacting Fe-N nanoparticle system

Comparison with noninteracting nanoparticle system
ZFC and FC Magnetizations

Fe-N system

calculation (noninteracting)

Smaller SG domains are blocked at lower temperatures, but not necessarily in the field direction due to frustration

Smaller particles are blocked at lower temperatures in the field direction
Sun et al's Protocol: Memory Effect

Fe-N system

$\chi_{\text{FC}}$ (emu/Oe)

$\chi(T,K)$

$t_w(H=0) = 3000 \text{ s}$

(noninteracting)
Cooling and stop in zero field. The magnetic field is applied at the lowest temperature and reheating.

**Fe-N system**

\[ T_{stop} = 40 \text{ K} \]
\[ t_w = 3000 \text{ s} \]

\[ M(t) = 0, \text{ or } p1(t) = p2(t) = 1/2, \text{ in zero field irrespectively of } T(t) \]
Growth and Blocking of SG Domains

After rapid quench to \((T, h)\), or at an intermittent stop at \((T, h)\), spin-glass system ages to the corresponding equilibrium state:

\[ R_{T,h}(t) \] : mean size of SG domains

By simulation it is explicitly measured:

3D Ising EA model \((T_C \approx 0.95J)\)

\[ R_T(t) \approx b_T (t/t_0)^{1/z(T)} \]

\[ 1/z(T) \approx 0.16 (T/T_C) \]

at \((T/T_C) \lesssim 0.7\)

When cooling is resumed, longer SG correlations developed are 'effectively blocked' at lower temperatures:

Memory Effect in Spin Glass !!
Memory effects

Experimental Protocol:
Cooling: stops at constant temperature (+ field changes)
Reheating: memory effects

Noninteracting nanoparticle systems

Spin glasses, Strongly interacting nanoparticle systems

Wide distribution of relaxation times, or blocking temperatures

Growth of SG order by aging: Time dependent distribution of relaxation times (energy barriers)
Experiments on a strongly interacting Fe-N nanoparticle system

Some other spin-glass properties
Interest of Interacting Nanoparticle System in Spin-Glass Study

Time scale of observation relative to microscopic time

\[ R_T(t) = 0.635 \left( t/\tau_{\text{micro}} \right)^{0.169T/T_c} \]

$t_{\text{micro}} = \tau_m = 1 \text{ mcs}$

$\tau_m = 10^{-12} \text{ s}$
Ac Susceptibility in an Isothermal Aging

$\chi''(t; \omega) = f(L_\omega / R_T(t))$

$L_\omega$ : max. size of SG droplets that can respond to ac field of frequency $\omega$ (independent of $t$)

$R_T(t)$ : mean size of SG domains at time $t$ in aging of $T$

$\chi''(t; \omega) \Rightarrow R_T(t)$

Cooling-rate dependence of aging in nanoparticle SG system
Temperature-shift protocol

Using growth low,

\[ R_T(t) \simeq b_T(t/t_0)^{1/z(T)} \]

we obtain:

\[ R_{T_m}(t_{\text{eff}}) = R_{T_i}(t_w) + \text{transient effect} \]

(generalized Kovacs effect)

Cumulative Aging Scenario
Twin Protocol

\[ R_{T_1}(t_{\text{eff}}) = R_{T_2}(t_w) \]

negative shift:
\[ t_{\text{eff}} \rightarrow t_1, t_w \rightarrow t_2 \]

positive shift:
\[ t_w \rightarrow t_1, t_{\text{eff}} \rightarrow t_2 \]

\[ t_2 = \tau_0 (t_1/\tau_0)^{T_1/T_2} \]

even with
\[ \tau_m = \tau_0 \exp(KV/k_B T) \]
Twin Protocol. II

\[ t_2 = \tau_0 \left( t_1 / \tau_0 \right)^{T_1 / T_2} \], or Cumulative Memory Scenario in fact holds when \( \Delta T \) is relatively small.
Deviation from Cumulative Memory Scenario

-- Rejuvenation and Chaos Effect --

Twin protocol on 3D Ising EA model (simulation): $T_c \approx 0.95J$

Deviation from

$t_2 = \tau_0 (t_1/\tau_0)^{T_1/T_2}$

for $0.7 \leftrightarrow 0.4$

Precursor of Chaos Effect
Summary

- **Memory Effect in Noninteracting Nanoparticle Systems** can be explained by distribution of energy barriers (Superparamagnetism and Blocking).

- **Memory Effect in Interacting Nanoparticle System (Spin Glass)** are interpreted by the growth of SG domains in aging and their blocking by cooling to lower temperatures.

- Typical Spin-Glass behavior of Interacting Nanoparticle System is explained by *Cumulative Memory Scenario*. 