Comment on “Memory Effects in an Interacting Magnetic Nanoparticle System"

著者	SASAKI Munetaka, Jonsson Petra, Takayama Hajime, Nordblad Per
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In a recent Letter, Sun *et al.* [1] study and discuss memory effects in an interacting nanoparticle system with specific temperature and field protocols. The authors claim that the observed memory effects originate from spin-glass dynamics and that the results are consistent with the hierarchical picture of the spin-glass phase. In this Comment, we argue their claims to be premature by demonstrating that all their experimental curves can be reproduced qualitatively using only a simplified model of *isolated* nanoparticles [2] with a temperature dependent distribution of relaxation times.

The ith magnetic moment in this model occupies one of two states with energies $-KV_i \pm HM_sV_i$, where K is the anisotropy constant, M_s the saturation magnetization, H the applied field, and V_i the volume of the ith nanoparticle. The superparamagnetic relaxation time is $\tau_i = \tau_0 \exp(KV_i/T)$. The occupation probability of one of the states is $p_i(t)$, which is solved by the master equation approach for any temperature and field protocol from a given initial condition [2]. The magnetization of the particle system is evaluated by averaging over the volume distribution $P(V) = \exp[-\ln(V)^2/(2\gamma^2)]/(\gamma V\sqrt{2\pi})$ with $\gamma = 0.6$.

Figure 1 shows field-cooled (FC) magnetization vs temperature measured on cooling—with temporary stops under zero field—and the subsequent reheating. Since the field is cut at T_s for t_s, $\{p_i(t)\}$ of moments which are active on the present time scale relax to 1/2. Among them, moments of particles fulfilling $t_s \approx \tau_0 \exp(KV_i/T_s)$ are frozen in certain values when the cooling is restarted. Those frozen states are reactivated when the system is reheated to T_s, causing a dip in χ. The time evolutions of the thermo-remanent-magnetization (TRM) shown in Fig. 2 can similarly be understood; an energy barrier specifies quite sharply a temperature, below (above) which the moment is blocked (superparamagnetic).

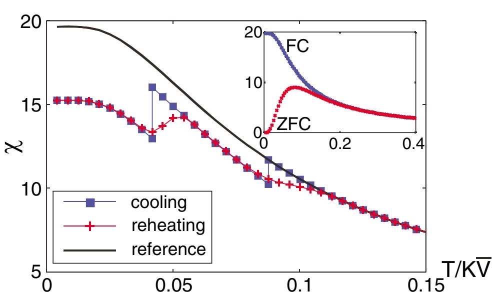

FIG. 1 (color online). FC susceptibility vs temperature using the same protocol as in Fig. 2 of Sun *et al.* [1]. The field is cut during the temporary stops of the cooling at $T = 0.088$ and at $T = 0.042$ for $10^{14}\tau_0$. The cooling (and reheating) rate is $2.4 \times 10^{13}\tau_0$ per temperature unit. The inset shows ZFC and FC susceptibility vs temperature.

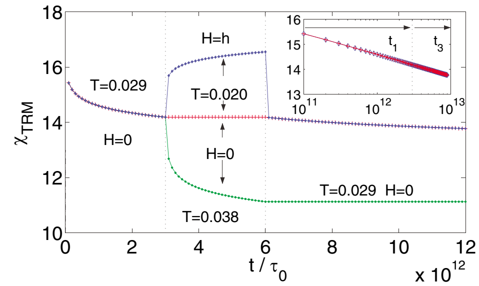

FIG. 2 (color online). χ_{TRM} vs time using the same protocols as in Figs. 3–5 of Sun *et al.* [1]. The system is cooled to $T = 0.029$ at the same rate as in Fig. 1 under a field which is cut just before recording χ_{TRM}. After a time of $3 \times 10^{12}\tau_0$, the temperature is changed for a period of $3 \times 10^{12}\tau_0$ either in $H = 0$ or $H = h$, and then it is shifted back in $H = 0$.

An appropriate protocol to confirm memory effects due to spin-glass dynamics is a zero-field-cooled (ZFC) process with a stop during cooling under a zero field [3]. In a spin glass the correlation length of spin-glass order grows during the stop and a memory dip shows up upon reheating, but not in a noninteracting nanoparticle system. This protocol, however, has not been examined in [1].

We have argued that a distribution of (free-)energy barriers is a sufficient origin of the memory effects discussed in [1]. In noninteracting nanoparticle systems the distribution of relaxation times originates only from that of the particle volumes, and is thus extrinsic and static. In spin glasses, on the other hand, it is the consequence of the cooperative nature of spins with randomly frustrated interaction, and is intrinsic and dependent on the age of the system. To conclude, only through the memory effects studied by Sun *et al.* [1] one cannot draw any conclusion whether a nanoparticle system is a noninteracting superparamagnet or an interacting spin glass [4].

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M. Sasaki,[1] P. E. Jönsson,[1] H. Takayama,[1] and P. Nordblad[2]
[1]ISSP, University of Tokyo
Kashiwa, Chiba 277-8581, Japan
[2]Å-lab, Uppsala University
Box 534, SE-751 21 Uppsala, Sweden

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