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Superspin glass behavior of interacting ferromagnetic nanoparticles in discontinuous magnetic multilayers

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Abstract

Discontinuous magnetic multilayers $[\text{Co}_{80}\text{Fe}_{20}(t)/\text{Al}_2\text{O}_3(3\text{nm})]_{10}$ with $t = 0.9$ and 1.0nm are studied by SQUID magnetometry and *ac* susceptibility. Owing to dipolar interaction the superparamagnetic cluster systems undergo collective glass-like freezing upon cooling. While both samples exhibit very similar glass temperatures $T_g \approx 45$ K and critical exponents $z\mathbf{n} \approx 10$ and $\mathbf{g} \approx 1.4$ as obtained from the temperature dependencies of the relaxation time, τ , and the nonlinear susceptibility, \mathbf{c}_3 , dynamical scaling reveals different critical exponents, $\mathbf{b}(0.9\text{nm}) \approx 1.0$ and $\mathbf{b}(1.0\text{nm}) \approx 0.6$, respectively.

Keywords: Multilayers, *ac* susceptibility, superparamagnetism, dipolar interactions, spin glass behavior

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INTRODUCTION

The magnetic behavior of ensembles of small ferromagnetic particles has found considerable interest during the past 50 years (Néel, 1949; Brown, 1963; Djurberg *et al.*, 1997; Dormann *et al.*, 1997). Most investigations have been focused onto single-domain particles on a nanoscale. They are usually randomly distributed in a diamagnetic metallic or insulating matrix and possess randomly oriented anisotropy axes. In the absence of interactions the dynamics of such a superparamagnetic (*SPM*) system is governed by the magnetic relaxation time of the particles with volume V at a given temperature, T ,

$$\mathbf{t} = \mathbf{t}_0 \exp(KV / k_B T), \quad (1)$$

where K and \mathbf{t}_0 are the anisotropy constant and the inverse attempt frequency, respectively (Néel, 1949; Brown, 1963). In the case of a monodisperse ensemble the real and the imaginary part of the complex *ac* susceptibility, $\mathbf{c} = \mathbf{c}' - i\mathbf{c}''$, become analytical functions of the *ac* frequency, f , and temperature (Andersson *et al.*, 1997). $\mathbf{c}'(T)$ and $\mathbf{c}''(T)$ exhibit peaks at temperatures T_m and T^* , respectively. T_m asymptotically approaches zero with decreasing f and paramagnetic Curie-type static susceptibility is achieved as $f \rightarrow 0$.

This is different in the presence of interactions between the *superspins*, e.g. of RKKY- or dipolar origin. Although conflicting models have been discussed (Dormann *et al.*, 1999b), both experimentally (Djurberg *et al.*, 1997; Jonsson *et al.*, 1998) and theoretically (Andersson *et al.*, 1997) much evidence was found for various systems that a *superspin glass* (*SSG*) phase may exist below a well-defined glass temperature, T_g . The dynamics of the system then follows a critical scaling law of the form (Ogielski, 1985)

$$\mathbf{t} = \mathbf{t}_0 (T / T_g - 1)^{-\mathbf{n}} \quad (2)$$

in the ergodic regime at $T > T_g$. While \mathbf{n} is the critical exponent of the correlation length, $\mathbf{x} \propto (T/T_g - 1)^{-\mathbf{n}}$, the exponent z relates the relaxation time to the correlation length, $\mathbf{t} \propto \mathbf{x}^z$. The

relaxation time t usually refers to the peak temperature, T_m , of the ac susceptibility, where $t = 1/2\pi f$ is connected with the ac frequency f of the experiment (Dormann *et al.*, 1999a).

Another crucial test of collective glassy freezing relies on the criticality of the nonlinear susceptibility, c_3 , as defined by the expansion of the magnetization M with respect to a weak magnetic field H (Djurberg *et al.*, 1997),

$$M = c_1 H - c_3 H^3 + O(H^5). \quad (3)$$

A power law

$$c_3 = c_3^0 (T/T_g - 1)^{-g} \quad (4)$$

with a critical exponent $g \geq 1$ is expected to hold close to T_g in the ergodic *SPM* regime.

Finally, dynamic critical scaling can be used to test the universality of the system. The imaginary part of the susceptibility, $c''(\omega, T)$, measured at various frequencies ω is expected to yield a data collapse onto a single function $H(\omega, t)$ according to (Gunnarsson *et al.*, 1988)

$$c'' / c_{eq} = (T/T_g - 1)^b H(\omega, t), \quad T > T_g, \quad (5)$$

where b is the order parameter exponent and c_{eq} denotes the equilibrium susceptibility in the limit $\omega \rightarrow 0$.

In the present paper the magnetic behavior of discontinuous multilayers (*DMIMs*) $[\text{Co}_{80}\text{Fe}_{20}(t)/\text{Al}_2\text{O}_3(3 \text{ nm})]_{10}$ with $t = 0.9$ and 1.0 nm is investigated. In these systems the CoFe layers are known to form isolated quasi-spherical ferromagnetic (*FM*) nanoclusters embedded in the alumina matrix (Kakazei *et al.*, 1999). It will be shown that generic *SSG* behavior emerges at low temperatures, $T < 60 \text{ K}$, despite the fairly wide size distribution as determined by high resolution electron microscopy, e.g. $\langle 2r \rangle \approx 3 \text{ nm}$ and $s = 2.7$ for $t = 1.3 \text{ nm}$ (Kakazei *et al.*, 2000). Previously it has been shown that *superferromagnetism* occurs for slightly higher CoFe thickness but still below site percolation for $1.4 \leq t < 1.8 \text{ nm}$ at room temperature (Kakazei *et al.*, 2000) and for $1.1 \leq t \leq 1.3 \text{ nm}$ below room temperature (Kleemann *et al.*, 2000a, b).

Under these conditions magnetic percolation takes place in a particularly dense fraction of the nanocluster system, while another, more dilute one still undergoes *SSG* freezing.

EXPERIMENT

DMIM systems of the structure $[\text{Co}_{80}\text{Fe}_{20}(t)/\text{Al}_2\text{O}_3(3\text{nm})]_{10}$ are prepared on a glass substrate covered by a $\text{Al}_2\text{O}_3(3\text{nm})$ buffer layer by ion-beam sputtering (Kakazei *et al.*, 1999). The thickness of the alumina layers were kept fixed.

The measurements were performed by use of a commercial Superconducting Quantum Interference Device (*SQUID*) magnetometer (*MPMS-5S*, Quantum Design). The *ac* susceptibility was measured by applying a small oscillating field with amplitude $m_0 H_{ac} = 0.05\text{mT}$ in zero external field and frequencies $0.01\text{Hz} \leq f \leq 1\text{ Hz}$. The real and the imaginary parts, χ' and χ'' respectively, are extracted as in- and out-of-phase components of the time-dependent magnetization response, respectively.

For nonlinear susceptibility studies the magnetization curves M vs. H were recorded after zero-field cooling (*ZFC*) from $T = 300\text{K}$ at temperatures $55 \leq T \leq 65\text{K}$ in fields $-5 \leq m_0 H \leq 5\text{mT}$ with 0.01mT resolution. In order to warrant thermal equilibrium, the critical slowing down has been overcome by isothermal waiting times between data points, $t_w = 100$ and 500s at $T > 60\text{K}$ and $\leq 60\text{K}$, respectively.

RESULTS AND DISCUSSION

Figure 1 shows the temperature dependence of the real and imaginary part of χ for seven different *ac* frequencies, $f = 0.01 - 1\text{ Hz}$ obtained on both samples, $t = 0.9$ (a) and 1.0 nm (b) in virtually zero-field ($|m_0 H| < 0.02\text{ mT}$) after zero-field cooling (*ZFC*) from $T = 200\text{ K}$. It is seen

that the peaks of both χ'' and χ' shift gently towards lower T on decreasing the frequency f , while the χ' curves peak on the low- T branches of the corresponding χ'' curves as expected for spin glass-like systems (Djurberg *et al.*, 1997). $T_m(f)$ values are extracted from the interpolated peak positions of $\chi''(T)$. Fig. 2 shows double-logarithmic plots of $t = (2\pi f)^{-1}$ versus the reduced temperature $T_m/T_g - 1$ for both samples together with best-fitted power laws, Eq. (2) (solid lines). The fits are seen to be satisfactory with essentially the same slopes, i.e. equal critical exponents $z\nu$. The following sets of data are obtained by best-fit procedures: $T_g = (45.6 \pm 4.6)\text{K}$, $t_0 = (2.8 \pm 1.3) \cdot 10^{-7}\text{s}$, $z\nu = 10.2 \pm 4.6$ for $t = 0.9$ nm and $T_g = (47.1 \pm 5.3)\text{K}$, $t_0 = (6.7 \pm 1.4) \cdot 10^{-7}\text{s}$, $z\nu = 10.0 \pm 3.6$ for $t = 1.0$ nm. Similar results, albeit with shorter t_0 values, were obtained on FeC and $\text{g-Fe}_2\text{O}_3$ nanoparticle systems (Djurberg *et al.*, 1997; Dormann *et al.*, 1999b). While the value of $z\nu$ agrees with that predicted for 3D spin glasses (Gunnarsson *et al.*, 1988), the large "spin-flip" time t_0 accounts for the cluster nature of the "superspins" (Djurberg *et al.*, 1997; Pfannes *et al.*, 2000).

Interestingly, the glass temperature T_g decreases but little as the CoFe layers become thinner. This agrees with observations on spin glass-like dilute FM systems (Fischer and Hertz, 1991; Mydosh, 1993). T_g is virtually independent of the concentration below a critical concentration, x_c , as soon as the average interaction $\langle J \rangle$ becomes smaller than the width dJ of its distribution. Since $T_g \approx dJ/k_B$, one does not expect important changes upon decreasing the average interspin distance, although $\langle J \rangle$ is further decreased. Similarly, when assuming heterogeneous nucleation in our $DMIM$ s (Kakazei *et al.*, 1999) and thus virtually constant particle density, the clearance between nearest-neighbor particles is enhanced at decreasing t . Hence, $\langle J \rangle$ will decrease, while dJ remains virtually unaffected.

Nonlinear susceptibility studies corroborate the above conjectured SSG nature of the $DMIM$ system with $t = 1.0\text{nm}$. To this end magnetization curves M vs. H were recorded after ZFC from $T = 300\text{K}$ at temperatures $55 \leq T \leq 65\text{K}$ in fields $-5 \leq m_0 H \leq 5\text{mT}$ with 0.01mT

resolution. Isothermal waiting times between data points, $t_w = 100$ and 500 s at $T > 60$ K and ≤ 60 K, respectively, were employed. The data were fitted to a polynomial, $M = c_1 H - c_3 H^3 + c_5 H^5$, where c_3 is expected to diverge at T_g in case of a collective spin-glass-like phase transition (Gunnarsson *et al.*, 1988). The results are plotted in Fig. 2b together with a best-fitted power law, $c_3 = c_3^0 (T/T_g - 1)^{-g}$ revealing $T_g = (50.7 \pm 2.3)$ K, $g = 1.36 \pm 0.53$ and $c_3^0 = (2.5 \pm 1.3) \cdot 10^{-9} \text{ (m/A)}^2$. Within errors T_g agrees with the value obtained from dynamic scaling (see above). The critical exponent $g \approx 1.4$ is smaller than that observed on spin glasses, $g \approx 4$ (Gunnarsson *et al.*, 1991). This seems to hint either at proximity to mean-field behavior, $g = 1$ (Fischer and Hertz, 1991; Mydosh, 1993) owing to the long-range nature of the dipolar interaction, or at spurious blocking processes of large particles within the relatively broad log-normal particle size distribution in our samples (Kakazei *et al.*, 2000).

Dynamic scaling analysis of $c''(\mathbf{w}, T)$ is shown in Figure 3 for $t = 0.9$ (a) and 1.0 nm (b). Here the data from Figure 1 are best-fitted to Eq. (5) when plotting $(T/T_g - 1)^{-b} c'' / c_{eq}$ versus $\mathbf{w}(T/T_g - 1)^{-z\mathbf{n}}$. The function $c_{eq}(T)$ is approximated by a Curie-Weiss hyperbola, $c'(f = 0.01\text{Hz}, T) = c_0 / (T - T_0)$, best-fitted to the respective low- f data within the range $60 < T < 90$ K, while trial values of $z\mathbf{n}$ and T_g are chosen to be close to the above dynamic power law fits. It is seen that data sets are reasonably collapsing in both cases when choosing consistent parameter sets, $T_g = 44$ K, $z\mathbf{n} = 10.0$, $\mathbf{b} = 1.0$ for $t = 0.9$ nm and $T_g = 46$ K, $z\mathbf{n} = 10.0$, $\mathbf{b} = 0.6$ for $t = 1.0$ nm, respectively. Obviously quite different values of the order parameter exponent \mathbf{b} emerge in parallel to remarkably different shapes of the scaling functions $H(\mathbf{w}, t)$. While $\mathbf{b} = 1.0$ for $t = 0.9$ nm complies with observations on SSG systems like $\text{Fe}_{0.88}\text{C}_{0.22}$ (Jönsson *et al.*, 2000), the low value $\mathbf{b}(1.0 \text{ nm}) = 0.6$ reminds of that found in the Ising spin glass $\text{Fe}_{0.5}\text{Mn}_{0.5}\text{TiO}_3$, $\mathbf{b} = 0.45$ (Gunnarsson *et al.*, 1988). However, this agreement is probably fortuitous. Owing to the unusual non-monotonic shape of $H(\mathbf{w}, t)$ we rather believe that a

crossover situation is encountered for the $t = 1.0$ nm sample, which is close to exhibit superferromagnetism (Kleemann *et al.*, 2000b). Very likely a subsection of nanoparticles within the broad distribution of particle sizes is close to ferromagnetic percolation (Kleemann *et al.*, 2000b). This gives rise to an extra contribution to the loss function $\mathbf{c}''(\mathbf{w}, T)$, which is assumed to be at the origin of the unusual peak structure discovered in $H(\mathbf{w}, t)$. Hence, we claim that only the more diluted sample, $t = 0.9$ nm, might represent a generic *SSG* system, whose critical exponent $\mathbf{b} \approx 1.0$ agrees with results on related systems (Jönsson *et al.*, 2000).

It should be stressed, however, that apart from these subtle differences both of our systems dominantly exhibit *SSG* freezing as evidenced by the virtually identical parameters T_g and zn . In this context one might consider another useful criterion, which clearly evidences the collective freezing process. It is well-known that the decrease of the gap between logarithmically equally spaced susceptibility curves with decreasing frequency (Fig. 1) can be characterized by specific values of the modulus $k = (1/T_f)(\Delta T_f / \Delta \log \mathbf{w})$ (Sandlund *et al.*, 1989). When setting the freezing temperatures $T_f \approx T_m$ (see Fig. 2a), k is found to decrease to small values as $T \rightarrow T_g$, $k \approx 0.01$ in both cases (Fig. 4). As discussed previously (Kleemann *et al.*, 2000b) these values seem to hint at three-dimensional glass-like freezing rather than at the two-dimensional one, where larger k values are expected (Sandlund *et al.*, 1989). In future experiments it will therefore be interesting to reduce the number n of *DMIM* bilayers towards the two-dimensional limit, $n = 1$, where significant changes, viz. $k \approx 0.04$ and $T_g \rightarrow 0$, are expected.

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Figure Captions

Fig. 1 $c'(f,T)$ and $c''(f,T)$ vs. T of CoFe/Al₂O₃ DMIMs with $t = 0.9$ (a) and 1.0 nm (b) measured at frequencies $f = 0.01, 0.02, 0.05, 0.1, 0.2, 0.5$ and 1 Hz (data points interpolated by spline functions; symbols explained in Fig. 2). Glass temperatures T_g are marked by arrows.

Fig. 2 Double-logarithmic plots of (a) $t = (2pf)^{-1}$ vs. $T_m/T_g - 1$ and (b) c_3 vs. $T/T_g - 1$ (obtained after waiting times t_w as indicated) for DMIMs with $t = 0.9$ (a) and 1.0nm (a and b), where $T_g = (45.6 \pm 4.6), (47.1 \pm 5.3)$ and (50.7 ± 2.3) K, respectively, from best-fits to power laws (straight lines).

Fig. 3 Dynamic scaling plots $(T/T_g - 1)^{-b} c''/c_{eq}$ vs. $w(T/T_g - 1)^{-zn}$ of the susceptibility data shown in Fig. 1a and b for $t = 0.9$ (a) and 1.0 nm (b) best-fitted by the parameter sets $T_g = 44$ K, $zn = 10.0$, $b = 1.0$, $c_{eq} = 0.115/(T - 42.5$ K) and $T_g = 46$ K, $zn = 10.0$, $b = 0.6$, $c_{eq} = 0.269/(T - 45$ K), respectively.

Fig. 4 Variation of the modulus $k = (1/T_f)(\Delta T_f / \Delta \log w)$ with the freezing temperature $T_f \approx T_m$ as obtained from c' vs. T as shown in Fig. 1a and b.