November 2001

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ELSEVIER

Journal of Magnetism and Magnetic Materials 226–230 (2001) 1825–1827

Journal of magnetism and magnetic materials

www.elsevier.com/locate/jmmm

AC susceptibility studies of discontinuous $Co_{80}Fe_{20}/Al_2O_3$ multilayers

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Abstract

Discontinuous metal–insulator multilayers $[Co_{80}Fe_{20}(t)/Al_2O_3(3\,nm)]_{10}$ are prepared by ion-beam sputtering and investigated by SQUID magnetometry and AC susceptometry. For $1.0 \leqslant t \leqslant 1.6$ nm, two coexisting subsystems of dipolar coupled nanoparticles are encountered. While a spin-glass-like freezing particulate fraction dominates at $t = 1.0$ nm, a percolating dipolar ferromagnetic fraction becomes increasingly important at higher nominal thickness of t.

Keywords: Multilayers; AC susceptibility; Superparamagnetism; Dipolar interactions; Spin glass behavior

Discontinuous magnetic metal–insulator multilayers (DMIMs) have become an interesting new class of granular magnetoresistive (MR) materials [1–4]. Consisting of layers with closely spaced ferromagnetic (FM) granules intercalated by insulating spacer layers, they do not suffer from pinhole short circuits when used as tunneling MR elements. Room temperature saturation MR values up to 4% were reported in the pioneering work on $CoFe/HfO_2$ and $CoFe/SiO_2$ [2–4], while a recent study on $[Co_{80}Fe_{20}(t)/Al_2O_3(3\,nm)]_n$ DMIMs yielded MR = 6.5% for current-in-plane geometry and a CoFe layer thickness $t = 1.0$ nm [5].

In the latter system, two different percolation limits were found when analyzing transport and magnetic properties, respectively. While the change from insulating to metallic behavior occurs at $t > 1.8$ nm [5], FM long-range order appears readily for $t > 1.3$ nm [6]. It has been argued that this behavior might be promoted by long-range dipolar interactions.

Within this context, it appears interesting to investigate the dynamics of the nanoparticle systems embedded in DMIMs. It is well known that noninteracting particles should follow the predictions of the Néel–Brown theory [7,8] of superparamagnetic relaxation, which primarily depends on the temperature, volume and anisotropy of the particles. While some predictions of this theory were recently confirmed by AC susceptibility studies on dilute granular films of γ-Fe_2O_3/Al_2O_3 [9] and Ni/SiO_2 [10], a study of interacting magnetic $Fe_{1-x}C_x$ nanoparticles evidenced collective magnetic dynamics, critical slowing down at finite temperatures [11] and low-temperature spin-glass-like behavior [12]. The present paper is, hence, motivated by the search for dipolar glassy behavior of FM granules in DMIMs and the possible appearance of a dipolar FM state.

To this end $[Co_{80}Fe_{20}(t)/Al_2O_3(3\,nm)]_{10}$ DMIMs were prepared by Xe ion-beam sputtering on glass substrates using methods described elsewhere [5,6]. While the Al_2O_3 layer thickness was fixed at 3.0 nm, the nominal thickness of the CoFe layers was varied between $1.0 \leqslant t \leqslant 1.6$ nm. Magnetization and magnetic

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0304-8853/01/$ - see front matter
PII: S0304-8853(00)00866-0

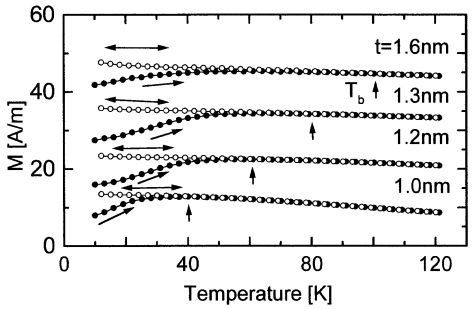

Fig. 1. M vs. T of DMIMs of $[CoFe(t)/Al_2O_3(3\,nm)]_{10}$ with $1.0 \leqslant t \leqslant 1.6$ nm measured on ZFC (solid symbols) and FC (open symbols). Vertical arrows mark blocking temperatures.

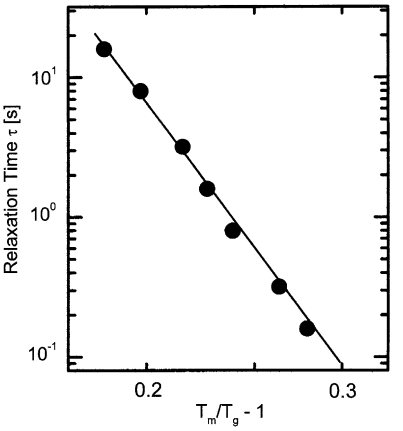

Fig. 3. Double-log plot of $\tau = (2\pi f)^{-1}$ vs. reduced peak temperature $T_m/T_g - 1$ for the DMIM with $t = 1.0$ nm (Fig. 2a), where $T_g = 47.1$ K from a best-fit to a power law (see text).

susceptibility were measured by using a SQUID magnetometer (Quantum Design MPMS-5S) at temperatures, $4 \leqslant T \leqslant 300$ K, magnetic fields, $|\mu_0 H| \leqslant 5$ T and frequencies, $10^{-3} \leqslant f \leqslant 500$ Hz.

Fig. 1 shows the temperature dependence of the magnetic moments of various DMIMs within the range $10 \leqslant T \leqslant 180$ K upon field-heating in $|\mu_0 H| = 10$ mT after zero-field cooling, (ZFC) to $T = 10$ K (solid symbols, up arrows) and upon subsequent field cooling (FC) back to $T = 10$ K (open symbols, bi-directional arrows). It is seen that the curves split apart below blocking temperatures $T_b \approx 40$, 60, 80 and 100 K (vertical arrows) for $t = 1.0$, 1.2, 1.3 and 1.6 nm, respectively. Obviously, all the samples exhibit the blocking phenomenon and are thus still distant from complete bond percolation expected at $t > 1.8$ nm [5].

Within the framework of the Néel–Brown theory [7,8], this would signify that the CoFe nanoparticles are frozen after ZFC in arbitrary anisotropy directions on the time scale of the SQUID experiment ($\tau \approx 1$ s) unless their magnetic moment becomes mobile via thermally activated processes above $T_b = \Delta E/[k_B \ln(\tau/\tau_0)]$. Assuming spin-flip times $\tau_0 \approx 10^{-8}$s, one calculates activation energies $\Delta E \approx 70$, 105, 140 and 180 meV, respectively. It is noticed that, contrary to the expectation of the conventional model [7,8], the relationship $\Delta E \propto t$ does not hold, where t varies essentially as the particle volume. Most probably, this indicates non-negligible dipolar interactions, which will be considered below.

Susceptibility data in zero external field are shown in Fig. 2 for two DMIMs and various frequencies, f. For the $t = 1.0$ nm sample, (a), $\chi'(f, T)$ and $\chi''(f, T)$ are similar to those observed previously on a granular Ni/SiO_2 film system [10]. While sizeable dispersion characterizes the range $40 \leqslant T \leqslant 80$ K, non-dispersive Curie–Weiss-type decay of $\chi'(f, T)$ with an extrapolated FM Curie temperature $\Theta \approx 58$ K is encountered at $T > 80$ K.

Convergence of the peak temperatures T_m of $\chi'(f, T)$ towards a finite freezing temperature T_g at low-f values is shown in Fig. 3 in a double-logarithmic plot of $\tau = (2\pi f)^{-1}$ vs. $T_m/T_g - 1$. A best fit of the data to the power law of critical dynamics, $\tau = \tau_0(T_m/T_g - 1)^{-z\nu}$, is

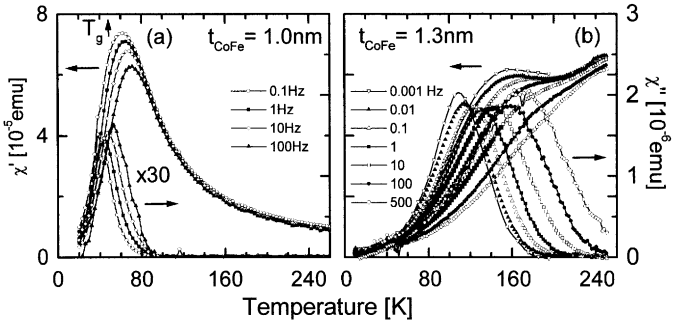

Fig. 2. $\chi'(f, T)$ and $\chi''(f, T)$ vs. T of DMIMs with $t = 1.0$ nm (a) and 1.3 nm (b) measured at frequencies $0.001 \leqslant f \leqslant 500$ Hz. Note the magnification factor of 30 in (a).

obtained with $T_g = 47.1 \pm 5.3\,\mathrm{K}$, $\tau_0 = (6.7 \pm 0.4) \times 10^{-7}\,\mathrm{s}$ and $zv = 10.0 \pm 3.6$. The large error margins are probably due to the non-uniformity of cluster sizes in the DMIM system (log-normal distribution [6]). While the value of the exponent zv is compatible with those predicted for spin glasses [13], the large spin-flip time τ_0 accounts for the cluster nature of the spins. It is noticed that also the loss peaks, $\chi''(f, T)$ tend to converge to $\chi'(f, T)$ at a finite temperature owing to their scaling relationship. Similar results apply to interacting $Fe_{1-x}C_x$ nanoparticle systems [11].

At higher nominal thickness, $t \geqslant 1.2\,\mathrm{nm}$, a dispersionless background is superimposed to the response curves of the polydispersive glassy subsystem. Fig. 2b shows a monotonically rising background signal for $t = 1.3\,\mathrm{nm}$. Tentatively it is attributed to a dipolar coupled, percolating cluster system. It behaves like a ferromagnet with finite anisotropy, whose susceptibility decreases upon cooling from its ordering temperature to low T. The appearance of the background signal unfortunately hampers an exact evaluation of the glass transition in this case. From the T-dependence of the $\chi'(f, T)$ shoulders in Fig. 2b a freezing temperature $T_g \approx 115\,\mathrm{K}$ is estimated after subtracting the background as approximated by the response function at the highest frequency, $f = 500\,\mathrm{Hz}$.

In summary, our analysis clearly shows that dipolar interaction has to be taken into account in all our DMIM-type nanoparticle systems. In the low-concentration range, $t < 1.2\,\mathrm{nm}$, it gives rise to a magnetic dipolar glass state at low temperatures. At higher concentrations, $t \geqslant 1.2\,\mathrm{nm}$, it coexists with a virtually percolating cluster giving rise to a dispersionless susceptibility background. Detailed studies of this novel phase are presently underway.

Thanks are due to DFG and DAAD (Germany) and CRUP (Portugal) for financial support.

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