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Magnetic states of discontinuous $Co_{80}Fe_{20}$–Al_2O_3 multilayers

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Abstract

Discontinuous metal–insulator multilayers $[Co_{80}Fe_{20}(t)/Al_2O_3(3\,nm)]_n$ were studied by SQUID magnetometry and AC susceptometry. CoFe forms ferromagnetic particles in the Al_2O_3 matrix. In this paper, we focus on the field dependence of the AC susceptibility of samples with $t = 1.3$ nm. We find strong evidence for transitions from a superpara- to a superferromagnetic and, finally, to a reentrant superspin glass state.

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

In the past decade the magnetic behavior of ensembles of *interacting* nanoparticles in a diamagnetic matrix has intensely been studied. It is now well established that dipolar interactions can give rise to a superspin glass state (SSG) in three-dimensional (3D) disordered systems of nearly monodisperse nanoparticles with low enough interparticle distances [1–3]. On the other hand, ferromagnetic (FM) ordering has been reported to exist in one- (1D) and two-dimensional (2D) self-organized [4,5] or regular [6] arrays of FM nanoparticles.

Recently, we discovered [7,8] superferromagnetism (SFM) and reentrant superspin glass (RSSG) behavior in 3D discontinuous metal–insulator multilayers (DMIMs) of $[Co_{80}Fe_{20}(t)/Al_2O_3(3\,nm)]_{10}$ with the nominal thickness $1.0 < t < 1.3$ nm [9,10]. While samples with a low concentration of CoFe particles, $t < 1.05$ nm, show a transition from a superparamagnetic (SPM) to an SSG state upon cooling [7,11], at intermediate concentrations, $1.05 \leqslant t \leqslant 1.3$ nm, reentrance into an SSG phase is encountered when cooling the SFM phase to low temperatures. In this paper, we primarily study the nature of the SFM transition by measuring magnetization and AC susceptibility in different applied magnetic fields. We chose the DMIM sample with $t = 1.3$ nm, which shows multiple magnetic phases in a most pronounced fashion [8]. A comparison between the 3D ($n = 10$) and the 2D case ($n = 1$) will also be presented.

The samples were prepared by Xe ion beam sputtering on glass substrates [9,10]. Transmission electron microscopy on DMIMs with $n = 1$ and $t = 1.3$ nm reveals quasispherical CoFe particles of face-centered cubic structure of average diameter $\bar{d} = 3$ nm, inter-granule distance of about 2 nm and log-normal size distribution width $\sigma = 2.7$ [10]. The *DC* magnetization and the *AC* susceptibility at frequencies $0.01 \leqslant f \leqslant 500$ Hz were measured with a superconducting quantum interference device magnetometer (Quantum Design MPMS-5S) at temperatures $4 \leqslant T \leqslant 400$ K. For zero-field measurements and for zero-field cooling experiments the remanent field of the superconducting coil and the earth magnetic field, $|\mu_0 H_{rem}| \approx 0.6$ and $|\mu_0 H_e| \approx 0.046$ mT, respectively, were compensated to below 0.03 mT.

Fig. 1 shows the real and imaginary parts of the AC susceptibility, $\chi'(T)$ and $\chi''(T)$, respectively, of

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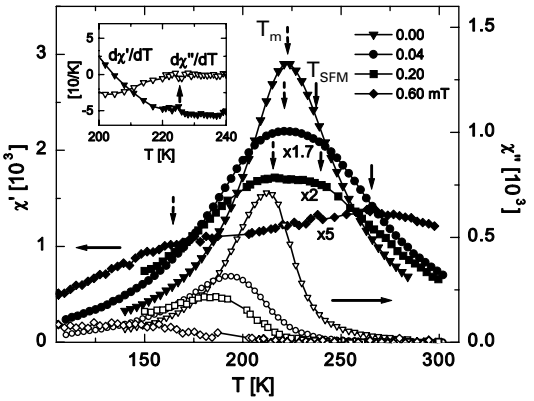

Fig. 1. Real and imaginary parts of the AC susceptibility, $\chi'(T)$ and $\chi''(T)$, respectively, of the $[CoFe(1.3\,nm)/Al_2O_3(3\,nm)]_{10}$ sample at different *DC* fields, $\mu_0 H = 0$, 0.04, 0.2 and 0.6 mT (± 0.03 mT) with constant AC frequency $f = 10$ Hz and *AC* amplitude $\mu_0 H_{AC} = 0.05 \pm 2 \times 10^{-5}$ mT . Different scale factors as indicated apply to both the real and the imaginary parts. The vertical arrows mark the positions of the two peaks, T_m (broken) and T_{SFM} (solid), in the χ' data. The inset shows the derivatives of the $f = 0.01$ Hz data.

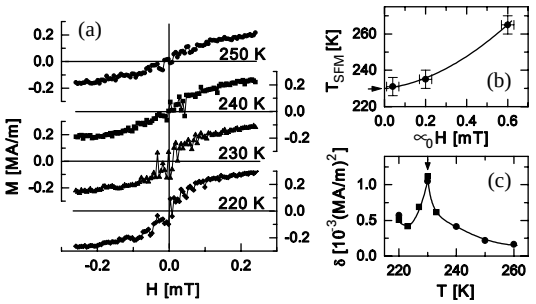

Fig. 2. (a) Magnetization vs. DC field, $M(H)$, at different temperatures, $T = 220$, 230, 240 and 250 K. (b) Peak positions, T_{SFM}, from Fig. 1. The arrow marks the extrapolated value, $T_{SFM}(H \to 0) \approx 230$ K. (c) Integrated square noise, $\delta(T)$, from two independent series of $M(H)$ isotherms (solid circles and squares). The arrow marks the peak at $T \approx 230$ K. Solid lines are guides to the eye.

$[Co_{80}Fe_{20}(1.3\,nm)/Al_2O_3(3\,nm)]_{10}$ in different DC fields, $\mu_0 H = 0$, 0.04, 0.2 and 0.6 mT (± 0.03 mT), taken after zero-field cooling (ZFC) from $T = 300$ K. The AC frequency, $f = 10$ Hz, and the AC field amplitude, $\mu_0 H_{AC} = 0.05$ mT are kept constant. The real part in zero field, $\chi'(T)[H = 0]$, shows a single peak at $T_m = 223$ K. This peak shows a characteristic frequency dependence, $T_m(f)$ (not shown), which is attributed to SSG dynamics as reported previously [7,8,11]. An additional anomaly is seen in the derivatives of the $f = 0.01$ Hz data (inset of Fig. 1) at $T_a = 225$ K, while the peak of $\chi'(T)$ shifts to $T_m = 205$ K.

In finite DC field the SSG peak at T_m shifts to lower temperatures (Fig. 1, broken arrows) in accordance with the Almeida–Thouless line shift observed on conventional spin glass systems [12]. An additional, frequency-independent peak emerges on the high-temperature branch of the $\chi'(T)$ curve at the SFM transition temperature T_{SFM} (solid arrows) [8]. As shown in Fig. 2(b) it strongly shifts with increasing field from $T_{SFM} \approx (230 \pm 5)$ to 265 K at $\mu_0 H = 0.6$ mT. Remarkably, the imaginary part, $\chi''(T)$, becomes flat with increasing DC field and shows no additional feature at T_{SFM}. Obviously, the SFM peak cannot simply be attributed to superparamagnetic blocking or spin glass freezing.

In order to study the SFM transition more thoroughly we measured magnetization isotherms within $-0.25 \leqslant \mu_0 H \leqslant 0.25$ mT in steps of 0.005 mT close to the expected transition temperature, $T_{SFM}(H = 0)$ (Fig. 2(a)). At $T = 230$ K one clearly observes strong fluctuations near $H = 0$. They are absent at other temperatures, e.g. at $T = 250$ K. The integral noise, $\delta = (1/\Delta H) \int (M - M_{sm})^2 \, dH$, as calculated for several curves within $220\,K \leqslant T \leqslant 260$ K, where $M_{sm}(H)$ is the smoothed magnetization curve, is shown in Fig. 2(c). A sharp peak at $T \approx (230 \pm 2)$ K resulting from two independent series of measurements corroborates the value of $T_{SFM}(H \to 0)$.

Remarkably, the fluctuations around T_{SFM} (Fig. 2(a)) occur on a time scale of several seconds. This hints at critical slowing down near the SFM phase transition, probably enhanced by the large superspin moments ($m \sim 1000 \mu_B$). As argued previously [8] the magnetically percolated 'backbone' of closely packed granules orders like a soft ferromagnet with vanishing remanence at $T_{SFM}(H = 0)$. Obviously, the extent and stability of the network can be controlled by the DC field. In view of the large field-induced shift of T_{SFM} (Fig. 2(b)) the field seems to have a strong cooperative effect on the FM phase transition apart from the usual smearing tendency. This might be explained by quantum mechanical inter-particle exchange [13], which is sensitively enhanced by the external field [14].

In order to study the influence of dimensionality, we repeated the AC susceptibility measurements on the 2D system $[Co_{80}Fe_{20}(1.3\,nm)/Al_2O_3(3\,nm)]_1$. Fig. 3 shows $\chi'(T)$ and $\chi''(T)$ measured under identical conditions as in the 3D case. A similar set of curves as in Fig. 1 is observed. The single peak as found in zero field at $T_m \approx 273$ K (broken arrow) shows a kink in the high-temperature branch at $T_{SFM} \approx 288$ K (solid arrow). With increasing *DC* field T_m shifts to lower and T_{SFM} to higher temperatures. Although the exact determination of T_{SFM} is difficult, its existence is unquestionable. Hence, SFM ordering is encountered similar to that found in 2D arrays of Fe particles on $CaF_2/Si(1\,1\,1)$ [13].

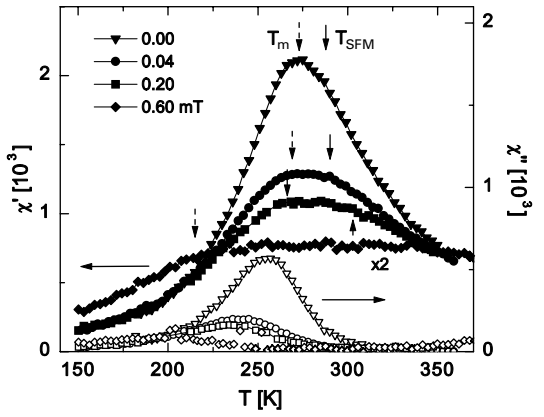

Fig. 3. Real and imaginary parts of the AC susceptibility, $\chi'(T)$ and $\chi''(T)$, respectively, measured on the [CoFe(1.3 nm)/Al_2O_3(3 nm)]$_1$ sample as described in Fig. 1. The real and imaginary parts for $\mu_0 H = 0.6$ mT were multiplied by a factor of 2. The vertical arrows mark the positions of the peaks, T_m (broken) and T_{SFM} (solid), in the χ' data.

Since dipolar coupling alone cannot lead to long-range order [15], additional anisotropy or quantum mechanical exchange [14] is required. Being unaware of sources of anisotropy during the DMIM growth process, we assume quantum mechanical inter-particle exchange to play an important role in our system.

In conclusion, the SFM transition temperature of the reentrant system $[Co_{80}Fe_{20}(1.3\,nm)/Al_2O_3(3\,nm)]_n$ is found to increase with increasing field. This is attributed to a field-induced stabilization of the SFM network, which is created upon cooling to below T_{SFM} from the SPM state. While $T_{SFM}(H = 0) \approx 287$ K for the 2D system ($n = 1$), it is shifted towards 230 K in the 3D system ($n = 10$). Probably, this can be attributed to a destabilizing weak antiferromagnetic interlayer interaction in the 3D multilayer.

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