October 2000

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Christian Binek
University of Nebraska-Lincoln, cbinek@unl.edu

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O. Petracic, W. Kleemann* and Ch. Binek

Laboratorium für Angewandte Physik, Gerhard-Mercator-Universität,

47048 Duisburg, Germany

G. N. Kakazei, Yu. G. Pogorelov and J. B. Sousa

IFIMUP, Departamento de Fisica, Universidade de Porto, 4169-007 Porto, Portugal

S. Cardoso and P. P. Freitas

INESC, Rua Alves Redol 9-1, 1000, Lisbon, Portugal

Received October 2000

Abstract

Discontinuous magnetic multilayers $[Co_{80}Fe_{20}(t)/Al_2O_3(3nm)]_{10}$ 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 $zn \approx 10$ and $g \approx 1.4$ as obtained from the temperature dependencies of the relaxation time, τ, and the nonlinear susceptibility, c_3, dynamical scaling reveals different critical exponents, b(0.9nm) ≈1.0 and b(1.0nm) ≈ 0.6, respectively.

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

* Corresponding author: Fax : ++49-203-1965, email: wolfgang@kleemann.uni-duisburg.de

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,

$$t = t_0 \exp(KV / k_B T), \tag{1}$$

where K and 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, $c = c' - \mathrm{i}c''$, become analytical functions of the *ac* frequency, f, and temperature (Andersson *et al.*, 1997). $c'(T)$ and $c''(T)$ 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 $f \to 0$.

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.*, 1999*b*), 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)

$$t = t_0 (T / T_g - 1)^{-zn} \tag{2}$$

in the ergodic regime at $T > T_g$. While n is the critical exponent of the correlation length, $x \propto (T/T_\mathrm{g} - 1)^{-n}$, the exponent z relates the relaxation time to the correlation length, $t \propto x^z$. 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 susceptbility, c_3, as defined by the expansion of the magnetization M with respect to a weak magnetic field H (Djurberg *et al*., 1997),

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

A power law

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

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, $c''(w,T)$, measured at various frequencies w is expected to yield a data collapse onto a single function $H(w,t)$ according to (Gunnarsson *et al.*, 1988)

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

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

In the present paper the magnetic behavior of discontinuous multilayers (*DMIM*s) $[Co_{80}Fe_{20}(t)/Al_2O_3(3\text{ nm})]_{10}$ with t = 0.9 and 1.0nm 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 < 60K, despite the fairly wide size distribution as determined by high resolution electron microscopy, e.g. $\langle 2r \rangle \approx$ 3nm and s = 2.7 for t = 1.3nm (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≤ t < 1.8nm at room temperature (Kakazei et al., 2000) and for 1.1≤ t ≤ 1.3nm 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 $[Co_{80}Fe_{20}(t)/Al_2O_3(3nm)]_{10}$ are prepared on a glass substrate covered by a $Al_2O_3(3nm)$ 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 $\mu_0 H_{ac}$ = 0.05mT in zero external field and frequencies 0.01Hz ≤ f ≤ 1 Hz. The real and the imaginary parts, c' and c'', 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 = 300K at temperatures 55 ≤ T ≤ 65K in fields –5 ≤ $\mu_0 H$ ≤ 5mT 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 > 60K and ≤ 60K, respectively.

RESULTS AND DISCUSSION

Figure 1 shows the temperature dependence of the real and imaginary part of c for seven different *ac* frequencies, f = 0.01 - 1 Hz obtained on both samples, t = 0.9 (a) and 1.0 nm (b) in virtually zero-field ($|\mu_0 H|$ < 0.02 mT) after zero-field cooling (*ZFC*) from T = 200 K. It is seen

that the peaks of both c' and c'' shift gently towards lower T on decreasing the frequency f, while the c'' curves peak on the low-T branches of the corresponding c' curves as expected for spin glass-like systems (Djurberg *et al.*, 1997). $T_m(f)$ values are extracted from the interpolated peak positions of $c''(T)$. 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 zn. The following sets of data are obtained by best-fit procedures: T_g = (45.6±4.6)K, t_0 = (2.8±1.3)·10^{-7}s, zn = 10.2±4.6 for t = 0.9 nm and T_g = (47.1±5.3)K, t_0 = (6.7±1.4)·10^{-7}s, zn = 10.0±3.6 for t= 1.0 nm. Similar results, albeit with shorter t_0 values, were obtained on FeC and g-Fe_2O_3 nanoparticle systems (Djurberg *et al.*, 1997; Dormann *et al.*, 1999b). While the value of zn 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 = 1.0nm. To this end magnetization curves M vs. H were recorded after *ZFC* from T = 300K at temperatures $55 \le T \le 65$K in fields $-5 \le m_0H \le 5$mT with 0.01mT

resolution. Isothermal waiting times between data points, t_w = 100 and 500s at T > 60K and ≤ 60K, respectively, were employed. The data were fitted to a polynomial, $M = c_1H - c_3H^3 + c_5H^5$, 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±2.3)K, g = 1.36±0.53 and c_3^0 = (2.5±1.3)·10^{-9} $(m/A)^2$. 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 $c''(w,T)$ 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 $w(T/T_g - 1)^{-zn}$. The function c_{eq}(T) is approximated by a Curie-Weiss hyperbola, c'(*f* = 0.01Hz, T) = $c_0/(T - T_0)$, best-fitted to the respective low-*f* data within the range 60 < T < 90K, while trial values of *zn* 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 = 44K, *zn* = 10.0, b = 1.0 for t = 0.9 nm and T_g = 46K, *zn* = 10.0, b = 0.6 for *t*= 1.0 nm, respectively. Obviously quite different values of the order parameter exponent b emerge in parallel to remarkably different shapes of the scaling functions $H(w,t)$. While b = 1.0 for t = 0.9 nm complies with observations on *SSG* systems like $Fe_{0.88}C_{0.22}$ (Jönsson *et al.*, 2000), the low value b(1.0 nm) = 0.6 reminds of that found in the Ising spin glass $Fe_{0.5}Mn_{0.5}TiO_3$, b= 0.45 (Gunnarsson *et al.*, 1988). However, this agreement is probably fortuitous. Owing to the unusual non-monotonic shape of $H(w,t)$ 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.*, 2000*b*). Very likely a subsection of nanoparticles within the broad distribution of particle sizes is close to ferromagnetic percolation (Kleemann *et al.*, 2000*b*). This gives rise to an extra contribution to the loss function $c''(w,T)$, which is assumed to be at the origin of the unusual peak structure discovered in $H(w,t)$. Hence, we claim that only the more diluted sample, t = 0.9 nm, might represent a generic *SSG* system, whose critical exponent $b \approx 1.0$ 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 $k = (1/T_f)(\Delta T_f / \Delta \log w)$ (Sandlund *et al.*, 1989). When setting the freezing temperatures $T_f \approx T_m$ (see Fig. 2*a*), 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.*, 2000*b*) 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.

We gratefully acknowledge stimulating discussions with P. Nordblad and valuable help by M.M.P. de Azevedo and S. Sahoo. Thanks are due to *DFG*, *DAAD* (Germany), *CRUP* and *PRAXIS XXI* (Portugal) for financial support.

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Figure Captions

Fig. 1 $c'(f,T)$ and $c''(f,T)$ vs. T of CoFe/Al_2O_3 *DMIM*s 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 *DMIM*s with t = 0.9 (*a*) and 1.0nm (*a* and *b*), where T_g = (45.6 ± 4.6), (47.1 ± 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. 1*a* and *b* for t = 0.9 (a) and 1.0 nm (b) best-fitted by the parameter sets T_g = 44K, zn = 10.0, b = 1.0, $c_{eq} = 0.115/(\mathrm{T} - 42.5\mathrm{K})$ and T_g = 46K, zn = 10.0, b = 0.6, $c_{eq} = 0.269/(\mathrm{T} - 45\mathrm{K})$, 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. 1*a* and *b*.