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Universal and scaled relaxation of interacting magnetic nanoparticles

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The logarithmic relaxation rate of the thermoremanent magnetic moment $m(t)$ of interacting magnetic nanoparticles in discontinuous $Co_{80}Fe_{20}/Al_2O_3$ multilayers follows a universal power law, whose exponent n increases with increasing particle concentration as predicted by recent simulations [Ulrich *et al.*, Phys. Rev. B **67**, 024416 (2003)]. While $n<1$ characterizes the stretched exponential decay of the dilute superspin glass (SSG) regime, $n>1$ refers to algebraic decay with finite remanence for $t \to \infty$ as observed in more concentrated superferromagnets (SFM). In the crossover regime from SSG to SFM, an increase from $n<1$ at low temperature to $n>1$ at $T \lessapprox T_c$ violates $T\ln(t/\tau_0)$ scaling and seems to indicate a crossover from random-field domain state to SFM behavior.

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Magnetic nanoparticle systems have attracted a lot of interest in fundamental research as well as in applications like future high-density data storage. In this technology the temporal stability of the magnetization of single particles is a crucial prerequisite and worth being investigated. Useful predictions based on size and anisotropy of the particles are provided by the Néel-Brown model.[1] It describes the high-temperature superparamagnetic (SPM) and the low-temperature blocking behavior of magnetic single domain particles (superspins) and predicts an exponential decay of the magnetization, $M(t)=M_0\exp(-t/\tau)$, for monodisperse particles, where M_0 is the magnetization at time $t=0$ and τ is the characteristic relaxation time dependent on the temperature T. Deviations from this simple law (e.g., logarithmic decay[2]) are expected for a finite particle-size distribution[3] and for interacting nanoparticle systems.[4] When forming a cooperative superspin glass (SSG) phase at high dilution[5–8] a stretched exponential relaxation of thermoremanent magnetization has been observed[9] similarly as in atomic spin glass systems.[10,11] At higher concentrations, superferromagnetic (SFM) behavior may be encountered,[12,13] where a finite remanence appears after, again, nonexponential decay.[14,15]

Recently Monte Carlo (MC) simulations were focused on the relaxation of an ensemble of superspins with random spatial distribution, anisotropy, and spin sizes by Ulrich *et al.*[16] They found that the logarithmic relaxation rate $W(t)$, defined as

$$W(t) = -(d/dt)\ln m(t), \tag{1}$$

follows a universal power law for all particle densities after some crossover time t_0,

$$W(t) = At^{-n} \text{ for } t \geqslant t_0. \tag{2}$$

From Eqs. (1) and (2) one finds three different decay laws of the magnetic moment $m(t)$, depending on the value of n

$$m(t) \simeq m_0 \exp[-(t/\tau)^{1-n}] \text{ for } 0 \leqslant n < 1, \tag{3}$$

$$m(t) \simeq m_1 t^{-A} \text{ for } n = 1, \tag{4}$$

$$m(t) \simeq m_\infty + m_1 t^{1-n} \text{ for } n > 1, \tag{5}$$

where m_0, m_1, m_∞, and τ are parameters linked to A, t_0, $m(t=t_0)$, and n. With increasing particle concentration the exponent n increases monotonically. Therefore, $n<1$ corresponds to dilute systems, while $n>1$ refers to dense systems. While both of these cases $n<1$ (Ref. 9) and $n>1$ (Ref. 15) were successfully evidenced previously, the marginal case $n=1$ (power-law relaxation without remanence) still lacks confirmation.

An excellent test ground for these predictions is offered by the matrix-isolated nanoparticle system $[Co_{80}Fe_{20}(t_n)/Al_2O_3(3\text{ nm})]_{10}$ (Refs. 6–8 and 13), where the nominal thickness t_n is related to the particle concentration as evidenced by the magnetic phase diagram shown in Fig. 1. Due to the heterogeneous nucleation of the nanoparticles during growth,[6,17] their number remains virtually constant at growing t_n, but their mutual distance decreases at increasing size. Below the transition temperatures T_g (glass temperature for $t_n<1.1$ nm) or T_c (SFM Curie temperature for t_n

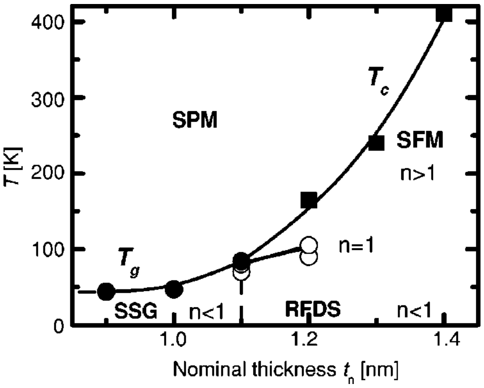

FIG. 1. Magnetic phase diagram of $[Co_{80}Fe_{20}(t_n)/Al_2O_3(3\text{ nm})]_{10}$ nanoparticle systems indicating the stability ranges of the phases SPM, SSG, SFM, and RFDS by the lines T_g (solid circles), T_c (solid squares) [Ref. 6] and $n=1$ (open circles) vs t_n and the corresponding rate exponents n (see text).

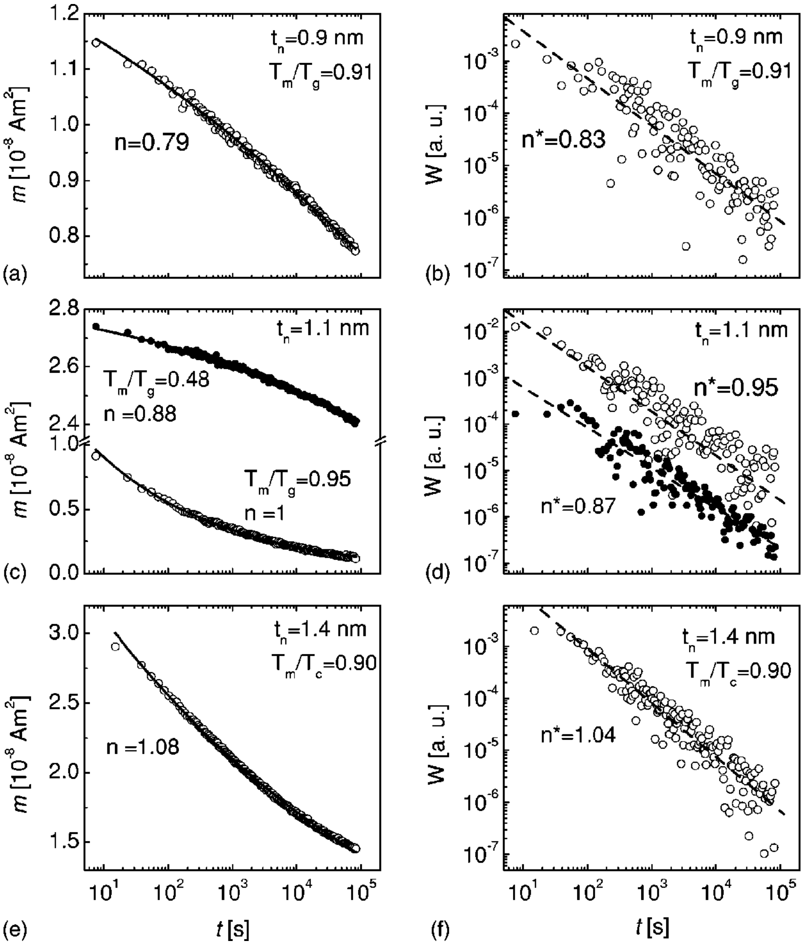

FIG. 2. Relaxation curves $m(t)$ [(a), (c), (e)] and $W(t)$ [(b), (d), (f)] of $[\mathrm{Co}_{80}\mathrm{Fe}_{20}(t_n)/\mathrm{Al}_2\mathrm{O}_3(3\ \mathrm{nm})]_{10}$ obtained for $t_n = 0.9$ at 40 K [(a), (b)], for $t_n = 1.1$ at 40 and 80 K [(c), (d), solid and open circles, respectively], and for $t_n = 1.4$ nm at 370 K [(e), (f)], and best-fitted to Eqs. (3)–(5) and (2) with solid and broken curves, respectively (see text).

>1.2 nm, but below the physical percolation threshold $t_n = 1.8$ nm[17]), we observe well-characterized SSG phases[7–9] and SFM domain states,[13,15] respectively. Indeed, while stretched exponential relaxation, hence, $0 < n < 1$ was observed for $t_n = 0.9$ nm,[9] a power-law decay with an asymptotic finite remanence, hence, $n > 1$, describes the $t_n = 1.4$ nm data.[15] According to the MC simulations[16] one would then expect the marginal case $n = 1$ to be realized in the crossover regime ($t_n \approx 1.1$ nm). Experimentally, however, reentrance from SFM to SSG-like (RSSG) behavior was inferred from *ac* susceptibility data of the $t_n = 1.1$ and 1.2 nm samples at low temperature.[6] In contrast with theory,[16] this would imply temperature-dependent decay exponents n. In this paper, we will corroborate this inference, but revise the interpretation of the RSSG state.

The discontinuous magnetic metal-insulator multilayers $[\mathrm{Co}_{80}\mathrm{Fe}_{20}(t_n)/\mathrm{Al}_2\mathrm{O}_3(3\ \mathrm{nm})]_{10}$ samples were prepared by focused Xe-ion beam sputtering on glass substrates.[17] High-resolution transmission electron micrographs of $t_n = 0.9$ and 1.3 nm samples[8,17] have shown that nearly spherical CoFe granules are embedded in the amorphous $\mathrm{Al}_2\mathrm{O}_3$ matrix. The relaxation of the thermoremanent magnetic moment $m(t)$ was measured by a superconducting quantum interference device (SQUID) magnetometer (Quantum Design, MPMS-5S) after flux gate controlled zero-field conditions within $|\mu_0 H| < 2\ \mu T$ were achieved by quenching the superconducting solenoid and compensating its remanent field.

The samples were cooled in a field of $\mu_0 H_0 = 0.46$ mT supplied by a fast switching copper coil from high temperatures (above the respective transition temperature, see Fig. 1) to the measurement temperature T_m, where the field was switched off and $m(t)$ was recorded. Note that for the $t_n = 1.4$ nm sample, in order to reduce the influence of the temperature-induced superspin alignment process, 17 h of waiting time was necessary before the cooling field was switched off.[15]

Figure 2 shows the relaxation of the thermoremanent magnetic moment $m(t)$ (left column) and its relaxation rate $W(t)$ (right column) as calculated from Eq. (1) for $t_n = 0.9$ [(a), (b)], 1.1 [(c), (d)], and 1.4 nm [(e), (f)] at T_m/T_g (or T_c) $\approx 0.4 - 0.9$. The solid lines in (a), (c), and (e) are best-fitted curves based on Eqs. (3)–(5). Here the $m(t)$ data were first fitted by all three laws, then the appropriate functional dependence was accepted after removing unphysical results like $m_0 \gg m(t=0)$ or $m_\infty < 0$. By this method we have obtained the exponents n without ambiguity. Note that the marginal case $n = 1$ emerges only indirectly in case that fitting to Eq. (4) yields the best result. On the other hand, the dashed lines in (b), (d), and (f) are best fits of $W(t)$ to Eq. (2). By this method exponents n^* emerge, which agree within errors ($|\Delta n| < 0.01$, $|\Delta n^*| < 0.05$) with the values of n obtained from $m(t)$ (see above). Table I presents more results of n and n^* for different t_n and T_m.

As expected, the exponent n increases as the particle con-

TABLE I. Selected values of the relaxation exponents n and n^* obtained by fitting the experimental data obtained for different t_n at different temperatures T_m to Eqs. (3)–(5) and (2), respectively (see text).

t_n [nm]	T_g (T_c) [K]	T_m [K]	n	n^*
0.9	44	40	0.79	0.83
		30	0.91	0.91
1.0	47	42	0.94	0.94
1.1	84	80	1	0.95
		70	1	0.94
		50	0.88	0.89
		40	0.88	0.87
		30	0.90	0.85
1.2	(164)	135	1.57	1.21
		120	1.23	1.13
		105	1	1.02
		90	1	1.05
		75	0.92	0.92
		60	0.88	0.88
1.4	(410)	370	1.08	1.04
		200	1.04	1.03

centration increases. In the SSG regime [Figs. 2(a) and 2(b)], i.e., for small t_n, we obtain $n<1$, while $n>1$ in the SFM regime [Figs. 2(e) and 2(f)], i.e., for large t_n. Intermediate values close to $n=1$ as expected from the *MC* simulations[16] appear at the border line between SSG and SFM [see Figs. 2(c) and 2(d) for $t_n=1.1$ nm and $T_m/T_g=0.95$]. Interestingly, in this marginal case a slight decrease to $n=0.88$ is encountered on cooling to $T_m/T_g=0.48$ [Figs. 2(c) and 2(d) and Table I]. Hence, a crossover from stretched exponential to power-law relaxational behavior seems to occur upon heating. This is compatible with the behavior of conventional spin glasses, where the time stretch exponent n also increases as $T\rightarrow T_g$.[10]

A more drastic change of n, namely, from $n>1$ at $T \lesssim T_c$ to $n<1$ at low temperature occurs for $t_n=1.2$ nm (Table I). Here one might infer reentrance of SSG behavior on cooling as conjectured previously.[6] As will be argued below, however, we rather believe the low-T relaxation of this marginal composition to be basically controlled by quenched random fields rather than by glassy disorder. This random-field induced domain state (RFDS) occurs below the phase line $n=1$ as indicated in the revised phase diagram in Fig. 1.

Principal differences of the relaxation behavior when increasing t_n from 1.1 to 1.2 nm become evident from scaling properties. As is well known from systems with thermally activated dynamics, e.g., spin glasses[18] or polydisperse Néel-Brown particle systems,[19] any change in temperature corresponds to a change in the time scale of the system. Within a general droplet picture involving Arrhenius-type activation, it is then plausible that the magnetic relaxation of such systems is governed by virtual energy barriers $E_c(t,T) = k_B T \ln(t/\tau_0)$, where τ_0 is a typical attempt time. As a consequence, $M(t,T)$ is a function of $E_c(t,T)$. Hence, all of the thermally activated relaxation curves are expected to collapse onto one master curve under $T\ln(t/\tau_0)$ scaling,[18,19] where the master curve relaxes toward a thermal equilibrium state.

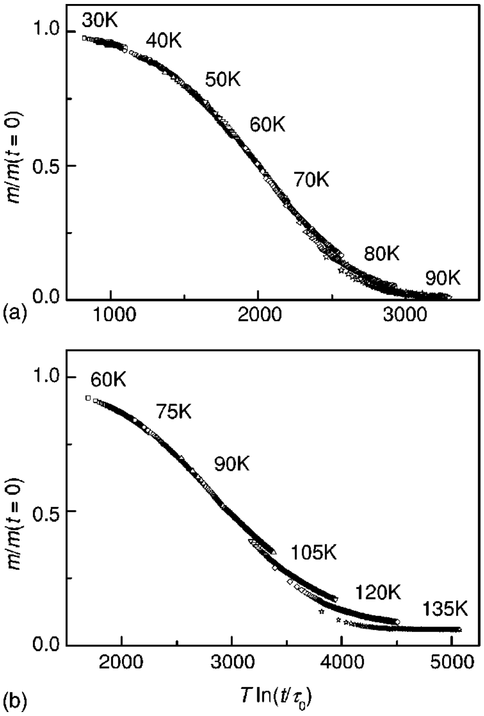

FIG. 3. Scaling plots of the normalized relaxation curves, $m(t)/m(t=0)$ vs $T\ln(t/\tau_0)$ for $[\mathrm{Co}_{80}\mathrm{Fe}_{20}(t_n)/\mathrm{Al}_2\mathrm{O}_3(3\text{ nm})]_{10}$ with $t_n=1.1$ (a) and 1.2 nm (b), where $\tau_0=1\times10^{-11}$ and 0.4×10^{-11} s, respectively. The measurement temperatures T are indicated at the particular data sets.

Figure 3 shows the normalized ralaxation curves, $m(t)/m(t=0)$, of the samples with $t_n=1.1$ (a) and 1.2 nm (b) in $T\ln(t/\tau_0)$ scaled representation. The empirical best-fit parameters $\tau_0=10^{-11}$ and 0.4×10^{-11} s, respectively, comply with typical inverse spin-wave frequencies. In both cases, the data sets collapse onto well-defined master curves for low temperatures, i.e., for $n<1$. However, in the higher-temperature regime, $0.85<T_m/T_g<1$ (a) and $0.7 <T_m/T_c<1$ (b), respectively, where $n\geqslant1$ (see Table I), the relaxation curves deviate from the master curve. Their decay is faster than predicted from the master curves.

For $t_n=1.1$ nm, we believe that only a slight acceleration of τ_0 apart from the transition into power-law relaxation ($n \simeq 1$; see above) might be responsible for the observed negative shifts on the t-axis at high T. Basically, however, the system decays into a SSG-like ground state with zero remanence after SSG droplet growth, which obeys Arrhenius activation.[20] Very probably this is different for $t_n=1.2$ nm, where, despite their sizable downward jumps, the decay curves end up with finite remanence, i.e., $n>1$, as $T\rightarrow T_c$. This hints at SFM behavior with additional dynamical processes besides the low-T ones. At higher T mesoscopic magnetized regions (domains) are expected to rearrange spontaneously as to minimize the magnetostatic energy in zero external field. These processes remind of Barkhausen-type domain wall relaxation (governed by nucleation and and growth) as observed in the magnetization decay or reversal in ordinary ferromagnets. A similar competition between fast relaxation of SFM domain walls toward lower total magnetization with slow relaxation within the SFM domains toward higher local magnetization was evidenced previously on the SFM multilayer with $t_n = 1.4$ nm at a fixed temperature.[15] In the present case, again, both domain wall relaxation on a mesoscale and dropletlike growth processes on a nanoscale are observed to compete.

Since processes with $n > 1$ and, hence, finite remanence at equilibrium, $m(t \to \infty) > 0$, dominate at large $T \ln(t/\tau_0)$, the system is basically SFM-like. In this situation, reentrance into a SSG-like disordered system at low temperatures as conjectured previously[6] is not easily understood. Here we propose an alternative mechanism, which might explain the observed $n < 1$ relaxation at small $T \ln(t/\tau_0)$ more physically. As is well known, an important part of the disorder in granular magnets is due to quenched random fields (RFs). They may be attributed to the small fraction of large particles[8,17] that become blocked at temperatures higher than the ordering ones.[13,21] Starting from the homogeneously magnetized initial state, it is then plausible that local fluctuations of the RFs give rise to a dropletlike formation of SFM domains on a nanoscale with a nonexponential decay of the order parameter autocorrelation function.[22] This mechanism requires low activation energy and should be efficient at low T, while mesoscopic domain wall processes with higher activation energies are more likely as $T \to T_c$. Thus, we propose that the low-T phase previously referred to as reentrant SSG[6] should rather be named the *RF domain state* (RFDS), showing SFM order on a nanoscale. The vertical phase line between SSG and RFDS (Fig. 1) then separates regions with dominating random bond (low t_n) and RF-induced disorder (high t_n), respectively.

In conclusion, the magnetic relaxation of interacting nanoparticle systems in discontinuous multilayers $[\mathrm{Co}_{80}\mathrm{Fe}_{20}(t_n)/\mathrm{Al}_2\mathrm{O}_3(3\ \mathrm{nm})]_{10}$ shows different decay laws as t_n increases. In agreement with predictions from MC simulations on related superspin systems,[16] stretched exponential, simple power, and power laws with finite remanence are identified according to the values of the relaxation rate exponents n. Strong temperature dependence of n is found in the marginal regime, $t_n \approx 1.2$ nm. It crosses the value $n = 1$ at the SFM/RFDS phase line,[6] which separates a RF-controlled $T \ln(t/\tau_0)$ scaling regime at low T from a region with preponderant domain wall relaxation dynamics at $T \lesssim T_c$.

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