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# Superspin-glass nature of discontinuous Co<sub>80</sub>Fe<sub>20</sub>/Al<sub>2</sub>O<sub>3</sub> multilayers

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## Superspin-glass nature of discontinuous $\text{Co}_{80}\text{Fe}_{20}/\text{Al}_2\text{O}_3$ multilayers

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Ferromagnetic single domain particles of CoFe in discontinuous magnetic multilayers  $[\text{Co}_{80}\text{Fe}_{20}(0.9 \text{ nm})/\text{Al}_2\text{O}_3(3 \text{ nm})]_{10}$  reveal spin-glass ordering below  $T_g = 43.6 \text{ K}$  as evidenced by the criticality of the nonlinear susceptibility. Dynamic critical scaling yields exponents  $z\nu = 8.0$  and  $\beta = 1.0$  in both zero and weak applied field, where an Almeida-Thouless line is encountered. In fields exceeding 2 mT a crossover into a chiral glass regime seems to occur.

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### I. INTRODUCTION

In recent years much attention has been focused on understanding the superspin-glass (SSG) behavior of single domain ferromagnetic nanoparticle systems.<sup>1-5</sup> Three-dimensional (3D) random distributions of such nanoparticles in an insulating matrix with high enough density and sufficiently narrow size distribution exhibit properties similar to those of conventional spin glasses. It is now widely accepted that interparticle dipole-dipole interactions in such SSG systems can lead to collective spin-glass ordering at low enough temperature.<sup>1-5</sup> Typical spin-glass characteristics such as critical slowing down of the relaxation and the divergence of the nonlinear susceptibility at a finite glass transition temperature  $T_g$  have strengthened the evidence of SSG transition in ferromagnetic fine-particle systems.<sup>1,4,5</sup> Some work has been addressed to the understanding of the nonequilibrium nature of the SSG phase,<sup>6</sup> where properties such as aging and memory effects have been observed and discussed within the framework of existing models.<sup>6,7</sup>

One of the most crucial tools to investigate the spin-glass phase transition is to study the nonlinear susceptibility. The magnetization in a spin-glass system can be expressed as an odd power series in  $H$  as<sup>8</sup>

$$M = \chi_1 H - \chi_3 H^3 + \chi_5 H^5 - \dots, \quad (1)$$

where  $\chi_1$  is the linear and  $\chi_3$ ,  $\chi_5$ , etc. are the nonlinear susceptibilities. At the phase transition temperature  $\chi_1$  is nondivergent, whereas the higher-order terms diverge, e.g.,  $\chi_3 \propto \varepsilon^{-\gamma}$  and  $\chi_5 \propto \varepsilon^{-(2\gamma+\beta)}$ , where  $\varepsilon = T/T_g - 1$  with the critical exponents  $\gamma$  and  $\beta$ .

Superspin-glass ordering in the presence of an externally applied magnetic field is a yet unexplored issue. In conventional spin-glass physics it has been matter of a long-standing debate.<sup>7,9</sup> A positive answer has been found within the framework of the Sherrington-Kirkpatrick mean-field model,<sup>10</sup> where de Almeida and Thouless<sup>11</sup> (AT) found stable solutions and have shown that replica symmetry breaking is

a characteristic feature of the spin-glass phase both in zero and nonzero field. Similar results were recently obtained for two different 3D spin-glass models, namely, the Edwards-Anderson Ising-type spin glass with short-range interactions<sup>12</sup> and the fully isotropic Heisenberg spin glass undergoing a chiral glass (CG) transition.<sup>13</sup> Gabay and Toulouse (GT)<sup>14</sup> generalized the mean-field theory to vector spin glasses, in which the freezing-in of transverse and longitudinal spin components successively occurs at two different temperatures in the presence of a field. At decreasing temperature the transverse components freeze-in first at  $T_{GT}$  as indicated by a weak difference of the zero-field-cooled (ZFC) and field-cooled (FC) magnetization, whereas the longitudinal components freeze-in at a lower temperature  $T_{AT}$ , where a strong irreversibility in the magnetization occurs. In the case of the Heisenberg model with random anisotropy a crossover in the high-field limit from AT to GT-type behavior was predicted<sup>15</sup> and recently been reinterpreted as an AT-to-CG crossover.<sup>13</sup>

While both lines,  $T_{GT}(H)$  and  $T_{AT}(H)$ , have experimentally been found in Heisenberg-like spin glasses,<sup>16,17</sup> in Ising-like spin glasses such as  $\text{Fe}_{0.5}\text{Mn}_{0.5}\text{TiO}_3$  only the  $T_{AT}(H)$  line seems to occur.<sup>18</sup> However, even the latter was disputed within the framework of the droplet model<sup>19</sup> where two thermodynamic equilibrium states are related to each other by a global spin reversal. Mattsson *et al.*<sup>20</sup> considered, again,  $\text{Fe}_{0.5}\text{Mn}_{0.5}\text{TiO}_3$  and argued that there is no spin-glass phase transition in a magnetic field although similar downward shifts of both the droplet freezing temperature  $T_f$  and  $T_{AT}$  are predicted for increasing field.

In view of this rather unsettled situation it seems useful to collect experimental information on spin glasses coming close to the randomly anisotropic Heisenberg model. It is most promising to exhibit AT-type behavior in the low-field regime and to cross over into the chiral glass phase at higher fields.<sup>13</sup> To this end we have investigated the magnetic phase boundary of a novel SSG, the discontinuous metal-insulator multilayer (DMIM) system  $[\text{Co}_{80}\text{Fe}_{20}(0.9 \text{ nm})/\text{Al}_2\text{O}_3(3 \text{ nm})]_{10}$ .<sup>4</sup> It is shown that this disordered magnetic

nanoparticle system with random anisotropy shows typical spin-glass properties, e.g., the divergence of nonlinear susceptibilities,  $\chi_3$  and  $\chi_5$ , and dynamic scaling of the magnetic-loss function,  $\chi''$  vs frequency  $\omega$ , with reasonable values of critical exponents  $z\nu$  and  $\beta$ . Remarkably, dynamic scaling remains valid in weak magnetic field,  $\mu_0 H = 1$  mT, where an AT-like phase line with an exponent  $\alpha \approx 3/2$  is observed. A strong increase of  $\alpha$  in higher fields seems to corroborate the crossover into the chiral glass regime.

## II. EXPERIMENTAL DETAILS

Measurements of ac susceptibility and dc magnetization in different fields have been performed on the DMIM system  $[\text{Co}_{80}\text{Fe}_{20}(t=0.9 \text{ nm})/\text{Al}_2\text{O}_3(3 \text{ nm})]_{10}$  by use of a commercial superconducting quantum interference device magnetometer (Quantum Design MPMS-5S). The CoFe forms nearly spherical granules in the  $\text{Al}_2\text{O}_3$  matrix. It is worth mentioning that these superparamagnetic nanoparticles reveal randomly distributed magnetic anisotropy axes thus allowing for the realization of a SSG with random anisotropy. The details of sample preparation are described elsewhere.<sup>21</sup> High-resolution transmission electron micrographs on a related sample with CoFe thickness  $t=1.3$  nm reveal an average diameter  $d \approx 3$  nm within a log-normal distribution width of  $\sigma=2.7$ .

The ac susceptibility was measured by applying a small oscillating field of amplitude  $\mu_0 H_{ac}=0.05$  mT superimposed to various dc fields in the range  $0 \leq \mu_0 H \leq 1$  mT. In all measurements the sample was mounted with the external dc field and the probing ac field oriented along the sample plane. The ZFC magnetization ( $M^{\text{ZFC}}$ ) and the FC magnetization ( $M^{\text{FC}}$ ) were measured as follows: (i) The sample was cooled in zero field from 140 K to 10 K. (ii) The field  $\mu_0 H$  was applied and  $M^{\text{ZFC}}$  was measured on heating up to 140 K. (iii)  $M^{\text{FC}}$  was measured on cooling from 140 K without changing the field. For zero-field measurements, and for ZFC experiments the remanent field of the superconducting coil and the earth magnetic field,  $\mu \approx 0.05$  and 0.046 mT, respectively, were compensated to within an accuracy of a constant (positive) field less than 0.03 mT.

## III. RESULTS AND DISCUSSION

In order to test static criticality, magnetization isotherms  $M$  vs  $H$  (not shown) were recorded after ZFC from  $T = 150$  K to temperatures  $52 \leq T \leq 64$  K in fields  $0 \leq \mu_0 H \leq 0.8$  mT at steps of 0.01 mT. In order to warrant thermal equilibrium, the critical slowing down has been overcome by isothermal equilibration times between data points,  $t_e = 200$  and 500 s at  $T \geq 60$  K and  $\leq 60$  K, respectively. The data were fitted to a polynomial,  $M = \chi_1 H - \chi_3 H^3 + \chi_5 H^5$ , where  $\chi_3$  should diverge at  $T_g$  in case of a collective spin-glass-like phase transition. The results are plotted in Fig. 1 together with a best-fitted power law,  $\chi_3 = \chi_3^0 (T/T_g - 1)^{-\gamma}$ , yielding  $T_g = (43.6 \pm 1.5)$  K,  $\gamma = 1.47 \pm 0.20$ , and  $\chi_3^0 = (6.0 \pm 0.2) \times 10^{-5} (\text{m/A})^2$ . Within errors  $T_g$  agrees with the value obtained from previous dynamic scaling analysis.<sup>4</sup> The critical exponent  $\gamma$  is smaller than that observed on spin glasses

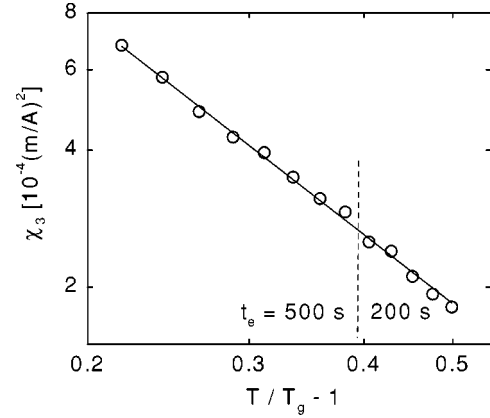


FIG. 1. Double logarithmic plot of  $\chi_3$  vs  $T/T_g - 1$  (obtained after equilibration times  $t_e$  as indicated), best fitted to the power law  $\chi_3 \propto \varepsilon^{-\gamma}$  with  $T_g = 43.6 \pm 1.5$  K and  $\gamma = 1.47 \pm 0.20$  (straight line).

( $\gamma=4$ ).<sup>1</sup> This seems to hint either at proximity to mean-field behavior<sup>22</sup> ( $\gamma=1$ ) 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<sup>21</sup> ( $\sigma=2.7$  for  $t=1.3$  nm) in our sample.

The temperature variation of the ZFC dc magnetization under various magnetic fields,  $0.05 \leq \mu_0 H \leq 0.7$  mT, is shown in Fig. 2(a). The curves are qualitatively similar to each other. The peak positions shift slightly downwards in temperature at increasing magnetic field. From the  $M$  vs  $T$  data isothermal cross sections  $M$  vs  $H$  are obtained both below and above the peak positions and fitted to polynomials according to Eq. (1).  $\chi_1$ ,  $\chi_3$ , and  $\chi_5$  vs  $H$  data, thus, obtained are plotted against temperature in Figs. 2(b), 2(c), and 2(d), respectively. In Fig. 2(b) the ZFC magnetization in a field  $\mu_0 H = 0.05$  mT [Fig. 2(a)] is compared with  $\chi_1(T)$ . It is seen that the peak positions of both curves coincide with

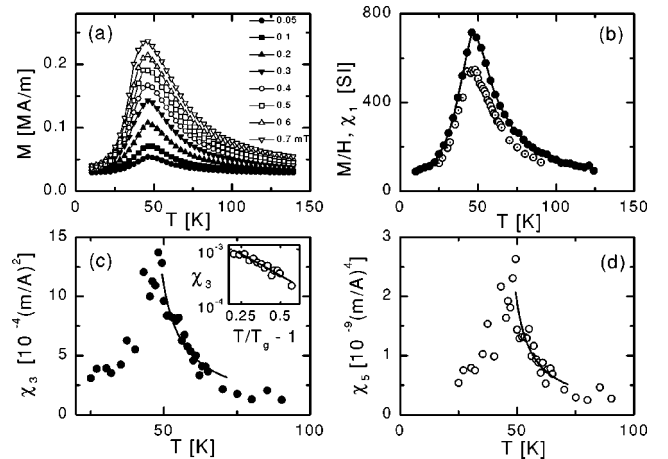


FIG. 2. (a) Temperature variations of the ZFC magnetization in various fields as indicated. (b)  $M/H$  at  $\mu_0 H = 0.05$  mT [Fig. 2(a); solid circles] and  $\chi_1$  [extracted from Fig. 2(a); dot centered circles] vs  $T$ . (c)  $\chi_3$  and (d)  $\chi_5$  vs  $T$  extracted from Fig. 2(a) and best fitted to power laws (solid lines; see text). The inset to (c) shows a double logarithmic plot of  $\chi_3$  vs  $T/T_g - 1$ , best fitted to a power law within 48–70 K.

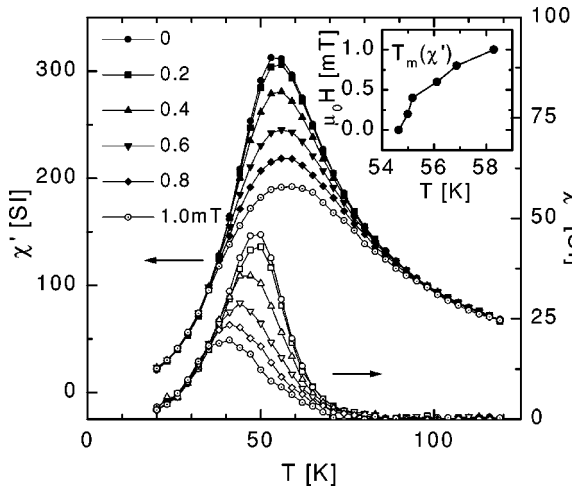


FIG. 3.  $\chi'$  and  $\chi''$  vs  $T$  at constant frequency  $f=1$  Hz and ac amplitude  $\mu_0 H_{ac}=0.05$  mT in various dc fields as indicated. The inset shows the shift of the peaks of  $\chi'$  vs  $T$ .

each other, while the height and acuteness of the cusp appears slightly reduced in the  $\chi_1$  data. In Fig. 2(c) the nonlinear susceptibility  $\chi_3$  tends to diverge close to  $T_g$ . A power law, best fitted in the temperature range from 48 to 70 K (inset of Fig. 3) yields  $\gamma=1.43\pm 0.10$  and  $\chi_3^0=(14\pm 1)\times 10^{-5}$  (m/A) $^2$  keeping  $T_g$  fixed at 43.6 K. These values are in fairly good agreement with those obtained from the above static criticality test.

The nonlinear susceptibility  $\chi_5$  in Fig. 2(d) also shows a sharp peak near to the expected transition temperature. One should notice that due to the above truncation of the series expansion, Eq. (1),  $\chi_5$  contains all higher-order terms,  $\chi_7$ ,  $\chi_9$ , etc. Hence, fitting to a power law may not yield the appropriate exponent. Indeed, the resulting exponent  $2\gamma + \beta=1.24\pm 0.14$  appears too small, yielding an unreasonable value  $\beta=-1.6$ , if  $\gamma=1.4$ . Obviously, one should consider higher-order terms, up to at least order  $H^7$ , when intending to make the  $\chi_5$  analysis.

Figure 3 shows the temperature variations of the real  $\chi'$  and the imaginary part  $\chi''$  of the measured ac susceptibility for a frequency  $f=1$  Hz at an ac field with amplitude  $\mu_0 H_{ac}=0.05$  mT superimposed to various dc fields,  $0\leq\mu_0 H\leq 1$  mT. The measurements are performed after ZFC from the superparamagnetic regime, $^5$  e.g.,  $T=150$  K to  $T=20$  K  $< T_g$  and then raising the dc fields to the levels indicated in Fig. 3. The predominant features of these results are as follows.

(1) The large suppression of both  $\chi'$  and  $\chi''$  near the freezing temperature  $T_m$  as defined by the peak of  $\chi'(T)$ . It is noticed that even very small fields cause considerable broadening of both  $\chi'$  and  $\chi''$  vs  $T$ . This observation is in remarkable qualitative agreement with experimental results on other systems such as AuFe (Ref. 23) and  $\text{Fe}_{0.5}\text{Mn}_{0.5}\text{TiO}_3$  (Ref. 20) and with Monte Carlo simulations. $^{24}$

(2) The gentle upward shift of the  $\chi'$  peak at  $T_m(H)$  as  $\mu_0 H$  increases as shown in the inset to Fig. 3. Similar upward shifts of the  $\chi'$  peak have also previously been observed in conventional metallic spin glasses, $^{23,25}$  where they

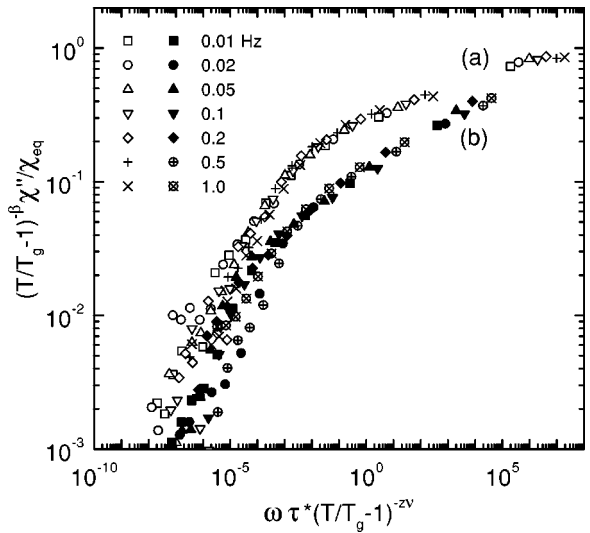


FIG. 4. Dynamic scaling plots  $\chi''\varepsilon^{-\beta}/\chi_{eq}$  vs  $\omega\tau_0\exp(E/k_B T)\varepsilon^{-z\nu}$  for  $\mu_0 H=0$  (a) and 1.0 mT (b) best fitted by the parameter sets (a)  $T_g=44$  K,  $z\nu=8.0$ ,  $\beta=1.0$ ,  $\chi_{eq}=4382$  K/( $T-46.3$  K),  $\tau_0=10^{-7}$  s,  $E/k_B=36$  K and (b)  $T_g=41$  K,  $z\nu=8.0$ ,  $\beta=1.0$ ,  $\chi_{eq}=6397$  K/( $T-34.8$  K),  $\tau_0=10^{-7}$  s,  $E/k_B=32$  K, respectively.

were explained in terms of a competition between the non-critical linear susceptibility and the critical nonlinear susceptibility. $^{25}$

Furthermore, the ac susceptibility was measured in external fields,  $\mu_0 H=0$  (Ref. 4) and 1.0 mT (not shown), and frequencies  $0.01\leq f\leq 1$  Hz at an ac amplitude  $\mu_0 H_{ac}=0.05$  mT. The most striking observation is the conservation of dynamical critical scaling properties of  $\chi''$  in nonzero field as shown by comparison of the scaled plots of  $\chi''\varepsilon^{-\beta}/\chi_{eq}$  vs  $\omega\tau^*\varepsilon^{-z\nu}$  in Fig. 4, curves (a) and (b), respectively. Here we proceed similarly as previously $^4$  for the  $\mu_0 H=0$  data with one modification. The relaxational behavior of the superspins is taken into account by incorporating an Arrhenius-Néel-type temperature-dependent relaxation time,  $\tau^*=\tau_0\exp(E/k_B T)$ , $^{26}$  with  $E/k_B=36$  K for  $\mu_0 H=0$  (32 K for 1.0 mT). As prefactors, we have chosen  $\tau_0=10^{-7}$  s as obtained previously for  $\mu_0 H=0$  $^4$  for both sets of data points. Although  $\tau_0$  is a function of an applied field $^{27}$  its value is of no relevance for the quality of the data collapse, since it simply scales the abscissa. While a slight decrease of  $T_g$  from 44 K (a) to 41 K (b) complies with AT-type behavior (see below), the critical exponents,  $z\nu=8.0$  and  $\beta=1.0$ , emerging for both field values are in remarkable agreement with those obtained on frozen ferrofluids in  $\mu_0 H=0$ . $^{26}$  The function  $\chi_{eq}(T)$  is approximated by a respective Curie-Weiss hyperbola,  $\chi'(f=10$  Hz) $=\chi_0/(T-T_0)$ , best fitted within the mean-field range,  $70$  K  $\leq T \leq 200$  K.

The temperature variations of the normalized magnetization  $M^{ZFC}/H$  and  $M^{FC}/H$  under various magnetic fields up to 30 mT are shown in Fig. 5(a). In the inset we show the data for two very weak fields, 0.3 and 0.7 mT. In contrast to most “real” spin glasses, $^{25}$  but in accordance with other superspin-glass systems $^1$   $M^{FC}$  shows a kink rather than a peak at  $T_g$ , although exceptions were found as, e.g.,

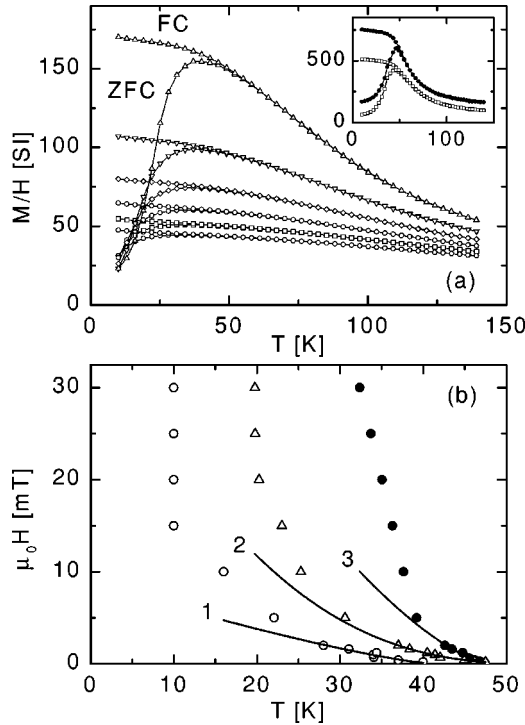


FIG. 5. (a)  $M/H$  vs  $T$  in fields  $\mu_0H=5, 10, 15, 20, 25, 30$  mT (from top to bottom). The inset shows  $M/H$  for  $\mu_0H=0.03$  and  $0.07$  mT (solid and open symbols, respectively). (b) Field dependence of  $T_{AT}(H)$  determined from data shown in Fig. 5(a) as “points of inflexion” (curve 1) and “points of intersection” (curve 2) of  $\Delta M(\mu_0H)$  vs  $T$ , respectively, and as peaks of  $M^{ZFC}/H$  vs  $T$  (curve 3), best fitted to Eq. (2) by solid lines for  $\mu_0H \leq 2$  mT (see text).

$\text{Fe}_{0.5}\text{Mn}_{0.5}\text{TiO}_3$ .<sup>28</sup> At higher magnetic fields the amplitudes of  $M/H$  strongly decrease owing to the nonlinearity of  $M$  vs  $H$ , while the peaks of  $M^{ZFC}/H$  are shifted to temperatures  $T < T_g$ . Furthermore,  $M/H$  increasingly flattens as  $H$  is increased.

The ZFC and FC magnetization curves [Fig. 5(a)] clearly reveal irreversibility, the characteristic feature of a spin-glass system. The peaks of  $M^{ZFC}/H$  occur a little below the onset temperature of the irreversibility, which can be attributed to the aforementioned spurious blocking of larger particles whose blocking temperatures  $T_b$  exceed  $T_g$ . For this reason the difference  $\Delta M = M^{FC} - M^{ZFC}$  appears as a smooth function of  $T$  without sharp kinks due to weak (i.e., transverse) or strong (i.e., longitudinal) irreversibility.<sup>14</sup> In order to evidence the very existence of a de Almeida-Thouless-type phase boundary,

$$\mu_0H(T_{AT}) = A\tilde{\varepsilon}^\alpha, \quad (2)$$

where  $\tilde{\varepsilon} = 1 - T_{AT}(H)/T_{AT}(0)$  and  $T_{AT}(0) \equiv T_g$ ,<sup>16</sup> we have tested three different, but equally plausible modes to determine  $T_{AT}(H)$ . In Fig. 5(b) we present the points of inflexion of  $\Delta M(\mu_0H)$  vs  $T$  (curve 1), the intersections of the steepest tangent of  $\Delta M(\mu_0H)$  vs  $T$  with the  $T$  axis (curve 2), and the peak positions of  $M^{ZFC}(\mu_0H)$  vs  $T$  (curve 3). Figure 6 shows some selected plots of  $d\Delta M/dT$  and  $\Delta M$  vs  $T$  for  $\mu_0H$

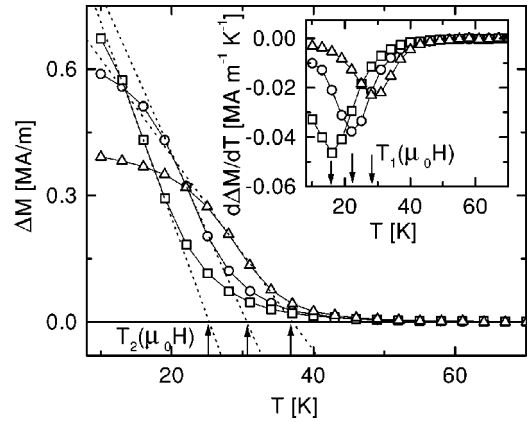


FIG. 6. Plots of  $d\Delta M/dT$  (inset) and  $\Delta M$  vs  $T$  for  $\mu_0H=2$  (triangles), 5 (circles), and 10 mT (squares). The corresponding points of inflexion,  $T_1(\mu_0H)$ , and of intersection,  $T_2(\mu_0H)$ , are indicated by arrows.

$=2, 5,$  and  $10$  mT and the corresponding points of inflexion,  $T_1(\mu_0H)$ , and of intersection,  $T_2(\mu_0H)$ , respectively. Although the curves in Fig. 5(b) are seriously shifted against each other along the  $T$  axis, they have one important similarity in common. They are fairly flat in the low-field range,  $\mu_0H \leq 2$  mT, while they sharply ascend beyond  $\mu_0H \approx 5$  mT. Interestingly, best fits of the low-field data points to Eq. (2) (solid curves) yield very similar exponents,  $\alpha = 1.3 \pm 0.6$  (1),  $2.6 \pm 1.0$  (2), and  $1.5 \pm 0.4$  (3), but different prefactors,  $A = 8.8$  mT (1),  $38.5$  mT (2) and  $52.0$  mT (3), and  $T_{AT}(0) = (41.1 \pm 3.1)$  K (1),  $(54.4 \pm 4.9)$  K (2) and  $(48.0 \pm 0.6)$  K (3). While all exponents come close to the mean-field prediction,  $\alpha = 3/2$ ,<sup>11</sup> comparison with the value  $T_g = (43.6 \pm 1.5)$  K obtained from the nonlinear susceptibility (see above) clearly favors curve 1 to become the best candidate for the AT line. In addition, its prefactor is very close to those predicted for the 3D Heisenberg ( $A = 8.2$  mT) and the 3D Ising model<sup>6</sup> ( $A = 15.0$  mT) when inserting superspin values  $S = 5000 \mu_B$  referring to an average number of atoms per granule,  $N \approx 2700$ , each of which carrying the specific magnetic moment per atom in a  $\text{Co}_{80}\text{Fe}_{20}$  alloy,  $m \approx 1.8 \mu_B$ .

It is proposed that the irreversibility line, curve 1, thus obtained in the range of magnetic fields  $\mu_0H$  up to about 2 mT corresponds to the ubiquitously expected AT line. Its steep rise in fields  $5 \leq \mu_0H \leq 30$  mT [Fig. 5(b)] clearly hints at failure of the AT theory, and thus at a destruction of the spin-glass phase transition. Owing to the restricted range of temperatures,  $T \geq 10$  K, no points of inflexion are available for  $\mu_0H > 10$  mT, where curve 1 merely shows the largest  $\Delta M(\mu_0H)$  values. Since the strong rise of all curves in Fig. 5(b) reminds of the AT-to-GT (Ref. 15) or of the AT-to-CG crossover<sup>13</sup> predicted for the randomly anisotropic 3D Heisenberg spin glass, we tentatively propose that a crossover into a chiral glass regime might characterize the field range exceeding  $\mu_0H \approx 5$  mT also in our randomly anisotropic 3D dipolar system. Here curve 2 (“points of intersection”) might take the role of the crossover line and replace curve 1 (“points of inflexion”), which shifts to very low

temperatures. The bending into a convex phase line,  $A\tilde{\epsilon}^\alpha$  with  $\alpha=0.5$ , expected<sup>13,15</sup> at very high fields,  $\mu_0H \gg 30$  mT, is still to be discovered. Work is in progress to clarify details concerning both the crossover and the saturation behavior of the high-field phase line.

#### IV. CONCLUSION

The existence of low-temperature superspin-glass ordering due to random dipole-dipole interaction in our DMIM system is evidenced by the divergence of the nonlinear susceptibility  $\chi_3$ , which is considered as a symptom of the spin-glass phase transition. The static criticality and the zero-field dynamical scaling independently yield consistent  $T_g$  values. The survival of the spin-glass phase in an external field is still debatable. However, the apparent validity of dynamical

scaling in both  $\mu_0H=0$  and 1.0 mT and the observed AT-line seem to evidence its existence in the low-field regime thus corroborating recent computer simulations<sup>12</sup> and torque measurements<sup>17</sup> on Heisenberg-like spin-glass systems. While no GT-type transverse irreversibility could be detected, a crossover into a chiral glass-like regime seems to occur at higher fields.

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