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Static Scaling on an Interacting Magnetic Nanoparticle System

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The field dependence of the ac susceptibility of a concentrated frozen ferrofluid containing ultrafine Fe-C particles of monodisperse nature has been analyzed using static scaling. For the first time, a divergent behavior of the nonlinear susceptibility of a dipole-dipole interacting system is demonstrated. From the analysis, the critical exponents $\gamma = 4.0 \pm 0.2$ and $\beta = 1.2 \pm 0.1$ were extracted. The results support the existence of a low temperature spin-glass-like phase in interacting magnetic nanoparticle systems. [S0031-9007(98)07527-9]

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The dynamics of systems of magnetic nanoparticles has been a subject of considerable interest during the last decades [1,2] and different, often conflicting, models have been proposed to explain the observations [3,4]. In one model the interparticle interactions are accounted for by a change of the energy barriers of the isolated particles, and this model excludes collective magnetic behavior of the particles [2,3]. Collective phenomena, however, play a key role in another model [4]. Hence, the possible existence of collective behavior and the existence of a low temperature spin-glass-like phase has been the subject of much controversy [2–9]. The main argument for suggesting the existence of a spin-glass phase is that properties such as frustration and randomness characterizing a spin glass are also found in dipole-dipole interacting magnetic particle systems. However, there are some dissimilarities between these systems: For a spin glass the flip time of the individual magnetic moments is of the order of 10^{-13} s and independent of temperature while it can vary from nanoseconds to geological time scales in a magnetic particle system according to $\tau = \tau_0 \exp(KV/k_BT)$, where $\tau_0 \sim 10^{-12}$–10^{-9} s, KV is the anisotropy energy, and k_BT is the thermal energy. Since all particle systems are more or less polydisperse, the subsequent distribution of anisotropy energies inevitably implies a distribution of flip times of the individual magnetic moments. Furthermore, the interaction between magnetic moments in spin glasses is mostly of short-range exchange or long-range RKKY type whereas it is of long-range dipole-dipole type in a magnetic particle system. Despite these differences it has been shown by Djurberg *et al.* [7] that the dynamics of a magnetic nanoparticle sample containing 5 vol % of amorphous $Fe_{1-x}C_x$ ($x \approx 0.22$) particles and an estimated dipole-dipole interaction strength of $E_{d-d}/k_B = 44$ K exhibits a spin-glass-like critical slowing down close to the extracted phase transition temperature, $T_g = 40 \pm 2$ K. These particles have a nearly spherical shape with a median diameter of 4.7 nm, and the volume distribution is well described by a log-normal distribution with $\sigma_V = 0.22$ [10]. In the critical region and for the experimentally accessible time scales this extraordinarily narrow size distribution implies that the characteristic time scale corresponding to the inverse of the transition rate for the largest individual particles is always shorter than the shortest measurable relaxation time caused by collective effects [7]. This is a necessary condition for probing magnetic response governed by a spin-glass-like fixed point [6,7]. For more details about the sample and the sample preparation see Refs. [7,10].

In this Letter the same sample as used by Djurberg *et al.* [7] for the dynamic study is used to perform a static scaling analysis of its magnetic response. From the analysis the transition temperature, T_g, and the critical exponents γ and β are determined. Values of other critical exponents are extracted using standard scaling laws.

The appropriate quantity to study in a static scaling analysis is the order parameter susceptibility, χ, which diverges at the phase transition temperature according to

$$\chi \propto \left(\frac{T}{T_g} - 1\right)^{-\gamma} = \epsilon^{-\gamma}, \qquad T > T_g . \tag{1}$$

For spin glasses it can be shown [11,12] that the order parameter susceptibility can be obtained from measurements of the nonlinear susceptibility, $\chi_{\mathrm{nl}} = \chi_0 - m/H$, where χ_0 is the zero field susceptibility and the magnetization, m, is written as

$$m = \chi_0 H + \chi_2 H^3 + \chi_4 H^5 + \cdots . \tag{2}$$

In this study H is composed of an ac field with angular frequency ω and amplitude h superimposed on a dc field, H_0; i.e., $H(t) = H_0 + h\sin(\omega t)$. For $h \ll H_0$ the lowest order terms of the nonlinear susceptibility become

$$\chi_{\mathrm{nl}} = -3\chi_2 H_0^2 - 5\chi_4 H_0^4 - \cdots . \tag{3}$$

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Suzuki has suggested the following general scaling law for the nonlinear susceptibility of spin glasses:

$$\chi_{\mathrm{nl}} = \epsilon^{\beta} G\left(\frac{H^2}{\epsilon^{\gamma+\beta}}\right), \tag{4}$$

where $G(x)$ is a scaling function [12]. An expansion of $G(x)$ in powers of x yields

$$\chi_{\mathrm{nl}} = a_1 \epsilon^{-\gamma} H^2 + a_2 \epsilon^{-(2\gamma+\beta)} H^4 + \cdots, \tag{5}$$

and from a comparison to Eq. (3) it is seen that $-\chi_2$ diverges like $\epsilon^{-\gamma}$ in the critical region. Geschwind *et al.* have later rewritten Eq. (4) in the form

$$\chi_{\mathrm{nl}} = H^{2\beta/(\gamma+\beta)} \tilde{G}\left(\frac{\epsilon}{H^{2/(\gamma+\beta)}}\right), \tag{6}$$

where $\tilde{G}(x)$ is another scaling function with an argument linear in ϵ [13].

A noncommercial carefully shielded SQUID magnetometer was used for the magnetic measurements [14]. The ac field had the amplitude $h = 4$ A/m while the superimposed dc field was stepwise increased in logarithmic intervals from typically 10 to 1900 A/m using a long solenoid working in persistent mode to ensure a good field stability. The temperature was controlled with an accuracy better than 1 mK.

One property of a magnetic particle system that can render this kind of study more difficult is the nonlinear response of the isolated particles. It is therefore an advantage if the nonlinear susceptibility of the isolated particles is small enough to be negligible compared to the collective nonlinear response. The response of the isolated particles was checked using a dilute sample (15×10^{-3} vol %) where the mean interaction strength between particles is very weak. The magnetization of the dilute sample is plotted vs H_0/T in Fig. 1 for the temperatures $T = 55$ and 80 K and fields up to 6.4 kA/m (80 Oe). The corresponding data for the concentrated sample is plotted for comparison. As expected for a collection of randomly distributed noninteracting particles, the magnetization data corresponding to different temperatures and fields collapse onto one curve when plotted vs the scaling variable H_0/T [15] and, more important, no significant effect of nonlinearity can be observed for H_0/T up to 80 $\mathrm{Am^{-1}\,K^{-1}}$. Since all measurements of the nonlinear susceptibility of the concentrated sample were made below 1.9 kA/m (24 Oe) and for temperatures between 55 and 80 K, the nonlinear susceptibility of the isolated particles can safely be neglected in this study.

Another complication that may arise in dipole-dipole interacting systems regards finding the local mean field acting on the individual magnetic dipole moments for a given sample shape. This problem has at present no generally accepted solution [2,16]. Moreover, even if such a model did exist, the actual sample shape is unknown if it changes when going from the liquid state at room temperature to the solid state at the measuring temperatures. However, provided that certain conditions prevail, the existing models do not exclude the possibility of a local mean field acting on the individual dipoles being equal to the applied field. The effect of the demagnetization field has been checked by measuring the spontaneous magnetic fluctuations in zero field. The imaginary part of the susceptibility, $\chi''(\omega, T)$, can be calculated from the magnetic noise power spectra, $P(\omega, T)$, using the fluctuation dissipation theorem [17]:

$$P(\omega, T) = 2k_B T \frac{\chi''(\omega, T)}{\omega}. \tag{7}$$

From a comparison of the calculated and measured $\chi''(\omega, T)$ curves it is then possible to find the mean local field acting on the individual magnetic dipole moments. A large number of time traces of the magnetic noise were recorded at different temperatures and subsequently Fourier transformed to obtain noise power spectra in the frequency range 10 mHz–1 kHz. An accurate value of the calibration constant used in the calculation of $\chi''(\omega, T)$ was obtained by comparing calculated and measured χ'' at 40 K and at low frequencies where the noise power spectrum is large in magnitude. The temperature dependence of the measured imaginary part of the ac susceptibility is displayed in Fig. 2 together with the corresponding results calculated from noise measurements. The measured $\chi''(\omega, T)$ was plotted without correcting for demagnetizing and Lorentz cavity fields and, since the agreement between the measured and calculated curves is good, the mean local field acting on the individual dipole moments can be set equal to the applied field.

The imaginary part of the ac susceptibility is shown in the inset of Fig. 2 for the frequencies $\omega/2\pi = 10$ mHz

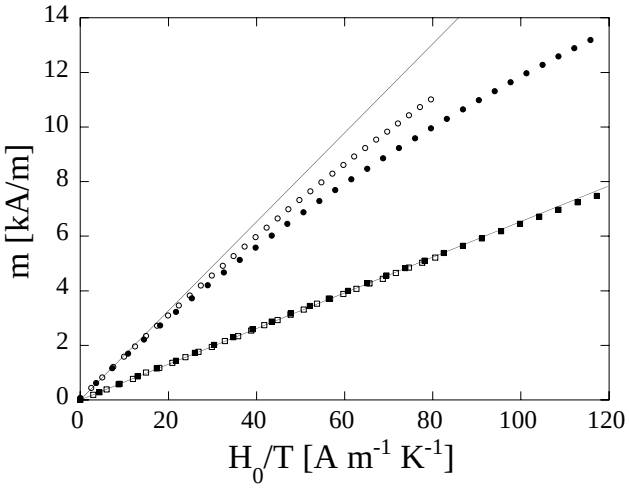

FIG. 1. The magnetization vs H_0/T for the dilute sample (squares) and the concentrated sample (circles) for the temperatures 55 and 80 K (filled and open symbols, respectively) and fields up to 6.4 kA/m (80 Oe). The data on the dilute sample were normalized to the concentration of the concentrated sample. The straight lines are included for visual clarity.

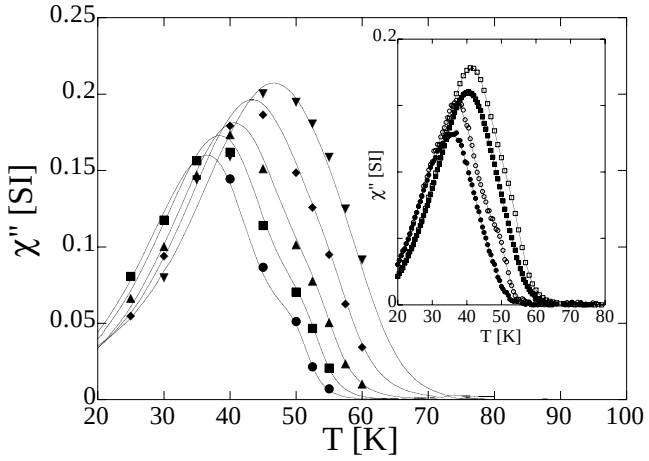

FIG. 2. $\chi''(\omega, T)$ vs temperature for the concentrated sample. The frequencies are from left to right $\omega/2\pi = 17$ mHz, 170 mHz, 1.7 Hz, 17 Hz, and 170 Hz (full lines). The symbols show $\chi''(\omega, T)$ calculated from noise measurements. The inset shows $\chi''(\omega, T)$ vs temperature for $H_0 = 0$ (open symbols) and 400 A/m (5 Oe) (filled symbols) and the frequencies $\omega/2\pi = 10$ mHz (circles) and 1.7 Hz (squares).

and 1.7 Hz and the dc fields $H_0 = 0$ and 400 A/m. When a dc field is applied, the magnitude of the susceptibility is suppressed compared to the zero field susceptibility. Moreover, the temperature corresponding to the onset of a nonzero χ'' decreases with increasing field implying that the temperature at which the magnetic response falls out of equilibrium decreases with increasing field. In a static scaling analysis it is crucial to use nonlinear susceptibility data corresponding to thermodynamic equilibrium, and therefore the frequency $\omega/2\pi = 1.7$ Hz is used in this study to probe the equilibrium magnetic response down to $T = 64$ K, and the frequency $\omega/2\pi = 10$ mHz is used for lower temperatures down to $T = 55$ K.

Since the time scale of the experiments is at most 4 orders of magnitude larger than the individual particle relaxation time in the temperature range used for the scaling analysis [7], the temperature range, where the static susceptibility can be observed, is restricted to a region relatively far from T_g and corrections to scaling might therefore be important. The situation is reminiscent to Monte Carlo (MC) simulations of model spin glasses where the time scale of the experiment is typically only 4–7 orders of magnitude larger than the individual spin flip time. Ogielski [18] performed MC simulations of a three-dimensional short-range Ising spin glass on a special purpose computer and found that the temperature dependence of the correlation time followed a critical behavior up to the reduced temperature $\epsilon \approx 0.55$. At higher temperatures a considerable deviation from critical behavior was observed because the correlation length at these high temperatures extends only over a few lattice spacings. In the present study the nonlinear susceptibility was measured up to $T = 80$ K corresponding to a reduced temperature of order unity. A crossover to a noncritical behavior at the highest measuring temperatures may therefore be expected.

The nonlinear susceptibility, χ_{nl}, is presented vs field in a log-log plot in Fig. 3(a) for some temperatures between 55 and 78 K. As expected from Eq. (5) the low-field behavior of χ_{nl} is proportional to H^2 and, hence, $\chi_2(T)$ can be determined. The extracted values of $\chi_2(T)$ are shown vs the reduced temperature in Fig. 3(b) for temperatures between 55 and 80 K. A best power-law fit to Eq. (1) using temperatures below $T = 69$ K ($\epsilon \approx 0.77$) is shown as the full line in Fig. 3(b) and yields $T_g = 39 \pm 1$ K and $\gamma = 4.0 \pm 0.2$. By also including the $\chi_2(T)$ data obtained at higher temperatures a deviation from a straight line is observed which drives T_g to a higher value and subsequently γ to a lower value. Accordingly, a best fit using data in the temperature range 55–80 K yields $T_g = 42 \pm 2$ K and $\gamma = 3.5 \pm 0.4$. The upward deviation of the experimental $\chi_2(T)$ data from the extrapolated power-law fit (dotted line) at higher temperatures in Fig. 3(b) is as expected for a crossover

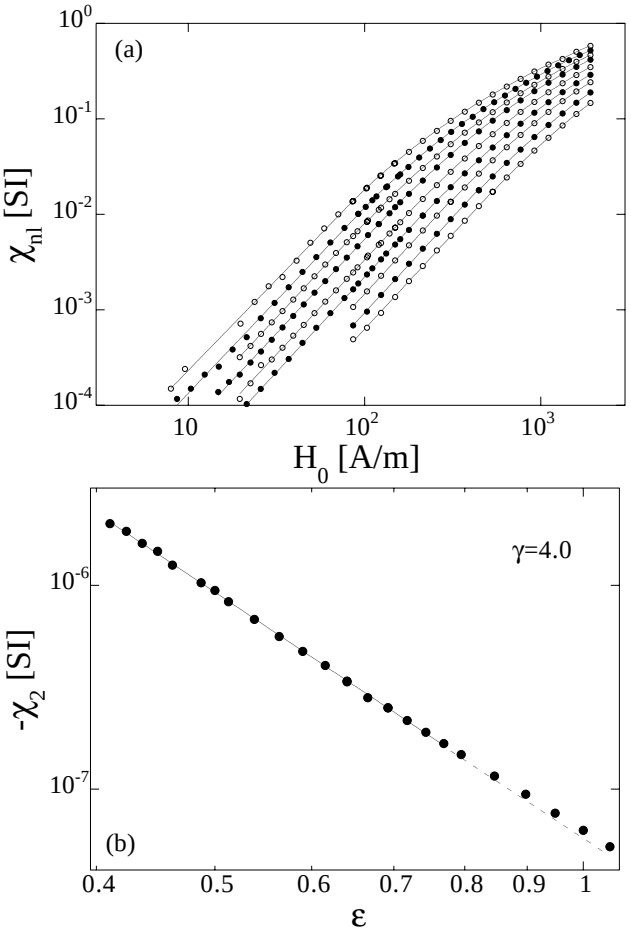

FIG. 3. (a) $\chi_{\mathrm{nl}}(T, H_0)$ vs H_0 for the temperatures 55, 57, 59, 61, 64, 67, 70, 74, and 78 K (from left to right). (b) $-\chi_2(T)$ vs the reduced temperature, $\epsilon = (T/T_g - 1)$, with $T_g = 39$ K.

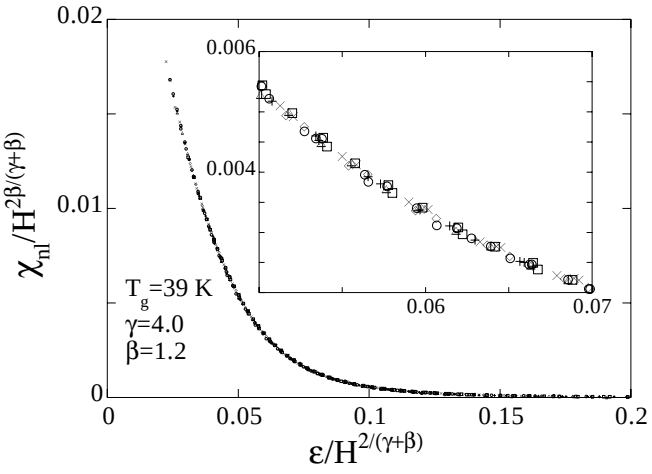

FIG. 4. Scaling plot of the nonlinear ac susceptibility for the temperatures 55, 56, . . . , 69 K, and dc fields between 10 and 1900 A/m. The inset shows a magnified part of the scaling plot.

to noncritical behavior [18]. The present extracted value of T_g is within the range $T_g = 40 \pm 2$ K, previously estimated using a dynamic scaling analysis [7].

In order to also extract the exponent β, an overall scaling of the measured nonlinear susceptibility data has been performed using the equation of state [Eq. (6)]. The data are plotted accordingly in Fig. 4 using the extracted $T_g = 39 \pm 1$ K and $\gamma = 4.0 \pm 0.2$. The best data collapse is obtained with $\beta = 1.2 \pm 0.1$. It is now possible to derive other critical exponents from scaling laws. The exponent for the specific heat is $\alpha = 2 - 2\beta - \gamma = -4.4 \pm 0.4$, and the exponent ν describing the divergence of the correlation length, ξ, as $\xi \propto \epsilon^{-\nu}$ is determined from the hyperscaling law, $\nu = (2 - \alpha)/d = 2.15 \pm 0.15$, where d denotes the spatial dimension. By adopting the transition temperature found in this study, a reanalysis of the data used for the dynamic scaling analysis in Ref. [7] gives $z\nu = 12.5 \pm 1.5$. The exponent, z, relating the correlation time, τ, to the correlation length as $\tau \propto \xi^z$ is then $z = 5.9 \pm 0.4$. This value is similar to estimates from MC simulations on model spin glasses where $z = 5.0 \pm 0.7$ [18,19].

In conclusion, the critical exponents in a dipole-dipole interacting magnetic nanoparticle system have been determined for the first time using static scaling analysis of the nonlinear susceptibility. The inherent properties of a magnetic particle system imply that the scaling analysis must be performed at comparably large reduced temperatures which may yield higher apparent values of the critical exponents [20]. However, according to extensive MC simulations on an Ising spin-glass system, the critical region is found to extend up to reduced temperatures $\epsilon \approx 0.55$ [18]. In the present analysis this is supported by the observation of corrections to scaling at reduced temperatures, $\epsilon \gtrsim 0.77$. From the analysis, the critical temperature $T_g = 39 \pm 1$ K and the critical exponents $\gamma = 4.0 \pm 0.2$ and $\beta = 1.2 \pm 0.1$ were extracted. Corresponding values for short-range Ising and Heisenberg spin glasses fall in the range $\gamma = 4.0 \pm 0.5$ and $\beta = 0.6 \pm 0.1$ [13,19,21–23]. The larger value of β obtained for the magnetic nanoparticle system may indicate a system closer to its lower critical dimension since β is predicted to diverge at its lower critical dimension [24]. It could also be that a system with long-range dipole-dipole interaction is better compared with RKKY spin glasses. Unfortunately, there is a large diversity in the reported values for the critical exponents for metallic spin glasses which makes such a comparison unreliable [21].

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