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Dynamics of Cu-Mn spin-glass films

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Using high-resolution superconducting quantum interference device magnetometry the dynamics of a 30-Å Cu-Mn spin-glass film have been investigated in the time range $10^{-5} < t < 10^4$ sec. It is found that the temperature dependence of the relaxation times obeys a generalized Arrhenius law, $\ln\tau \propto T^{-2.6}$, in accord with predictions for *two-dimensional* systems from Monte Carlo simulations and the droplet scaling theory. In comparison, results on a 10^4-Å Cu-Mn film show typical *three-dimensional* dynamics with a finite spin-glass temperature. To our knowledge this is the first experimental indication of a crossover behavior from three to two dimensions.

Today, there is a general consensus that the lower critical dimension, d_l, of Ising spin glasses is between two and three. Detailed experimental studies of the dynamic susceptibility of a model three-dimensional Ising spin glass[1] and results from extensive Monte Carlo (MC) simulations on a short-range Ising spin-glass model[2] exhibit a similar functional form for the time-dependent susceptibility. Results of dynamic scaling calculations imply a divergence of the relaxation times at a finite temperature in accord with conventional critical slowing down. MC simulations on two-dimensional Ising spin-glass models[3,4] exhibit a distinctly different behavior for the time-dependent susceptibility and the relaxation times diverge at $T=0$ K according to a generalized Arrhenius law. Recent experimental results on 2D Ising spin-glass materials[5,6] have also been interpreted in the context of a transition at $T=0$ K. The phenomenological droplet scaling theory for spin glasses, recently elaborated by Fisher and Huse,[7] predicts a behavior of spin-glass dynamics in agreement with experimental results and MC simulations especially regarding the influence of dimensionality on the dynamics.

A qualitative and quantitative comparison of the consequences of dimensionality on the spin-glass dynamics between different spin-glass materials is prevented by differences in the exchange interaction and the anisotropies, which impose material-unique details in the dynamics. MC simulations on identical spin-glass systems but of different dimensions yield the possibility of singling out the influence of dimensionality on the dynamics. Progress in material technology has made it possible to produce thin films a few monolayers thick and to adequately characterize the quality of the materials. This possibility has been adopted by Kenning and co-workers[8] to gain further insight into the dimensionality problem of spin glasses. They have recently studied Cu-Mn spin-glass films of varying thickness. Finite-size effects were observed as a gradual shift towards lower temperature of the quasistatic maximum of the zero-field-cooled susceptibility with decreasing film thickness.

In this paper, we report measurements of the *dynamics* on two Cu(13.5 at.% Mn) spin-glass films with thicknesses 30 and 10^4 Å. For the 30-Å film the dynamics was investigated in the time range $10^{-5} < t < 10^4$ sec, with the result that the temperature dependence of the relaxation times follows a generalized Arrhenius form, in agreement with MC simulations on 2D Ising spin-glass models.[3,4] The 10^4-Å film exhibits properties typical for bulk Cu-Mn spin glasses.[9] Pronounced differences between the two spin-glass films were also found in the nonlinear field effects and cooling rate dependence of the field-cooled susceptibility as well as the behavior of the aging phenomenon. These results are all in favor of a *dimensionality crossover* from three-dimensional to two-dimensional behavior on decreasing the film thickness from 10^4 to 30 Å.

The Cu(13.5 at.% Mn) films were produced in a UHV dc-sputtering system. The 30-Å sample was fabricated in the form of a multilayer sample consisting of 100 layers of 30 Å Cu(13.5 at.% Mn)/300 Å Cu. The interlayers are sufficiently thick (300 Å) to make the interaction between adjacent Cu-Mn layers negligible. The 10^4-Å film consists of a single layer of Cu(13.5 at.% Mn). The characterization of the films is described elsewhere.[8] The volume of spin-glass material in each of the two films is approximately 0.05 mm^3.

Using a high-resolution SQUID magnetometer the dynamic susceptibility of the Cu-Mn films was investigated by zero-field-cooled (ZFC) and ac susceptibility measurements. The ZFC measurements were performed by cooling the sample in zero field ($< 10^{-3}$ G) from a reference temperature (T_{ref}) well above the maximum of the field-cooled (FC) susceptibility curve, to the measurement temperature (T_m). After a wait time t_w a weak magnetic field ($H=10$ G) was applied and the time evolution of the magnetization $M(t)$ was recorded in the time interval $3\times10^{-1} < t < 10^4$ sec. The sample was then heated to T_{ref}, where a reference value of the ZFC magnetization was obtained. In an applied field of 10 G both films give a magnetic moment of approximately 10^{-9}–10^{-10} Am^2, which is 3 to 2 orders of magnitude larger than the noise limit of our magnetometer. The in-phase $\chi'(\omega,T)$ and the out-of-phase $\chi''(\omega,T)$ components of the ac susceptibility were measured in the frequency range $5\times10^{-1}<\omega/2\pi<1.7\times10^{4}$ Hz using a method described elsewhere.[10] A small temperature-dependent signal originating from the ac-coil system was superimposed to the in-phase signal making a complete scaling analysis of $\chi'(\omega,T)$ difficult. However, the location in temperature of the maximum of $\chi'(\omega,T)$, which coincides with the inflection point of $\chi''(\omega,T)$ could be determined quite accurately.

In the regime of linear response,[11] both the ZFC and ac susceptibilities reflect the time dependence of the zero-field susceptibility $\chi(t)$ and can thus be directly compared with the time dependence of the dynamic spin-spin correlation function $q(t)$ studied in MC simulations[2-4]. $\chi(t)$ is obtained from the experimental ZFC and ac susceptibilities through[1,12]

$$\chi(t)=(1/H)M(t)\approx\chi'(\omega),\ \text{with } t=\omega^{-1}. \tag{1}$$

Figure 1(a) shows $\chi(t)$ versus temperature for the 30-Å film at different observation times t. The FC susceptibility, $(1/H)M_{\mathrm{FC}}$, measured at two different cooling rates (0.9 and 0.02 K/min) is also included in the figure. The location of the cusp (at T_f) of $\chi(t)$ markedly shifts towards higher temperatures and subsequently broadens with decreasing observation time. In addition there is a pronounced cooling rate dependence of the FC susceptibility, with the knee shifting towards lower temperatures with decreasing cooling rate. In comparison a corresponding plot for the 10^4-Å film is shown in Fig. 1(b). The FC susceptibility, recorded at the cooling rate 0.02 K/min, exhibits a maximum at 66.8 K and the time effects of $\chi(t)$ are only about 20% of those of the 30-Å film. A further significant difference between the two films is observed in the nonlinear field effect on the FC susceptibility. It is found that an increase of the field from 3 to 25 G suppresses the FC susceptibility of the 10^4-Å film by 2%, whereas no nonlinear field effect ($\leq 0.1\%$) is found for the 30-Å film at the same field change. This observation implies that the critical point for the 30-Å film is remote from the location of the maximum of the FC susceptibility.

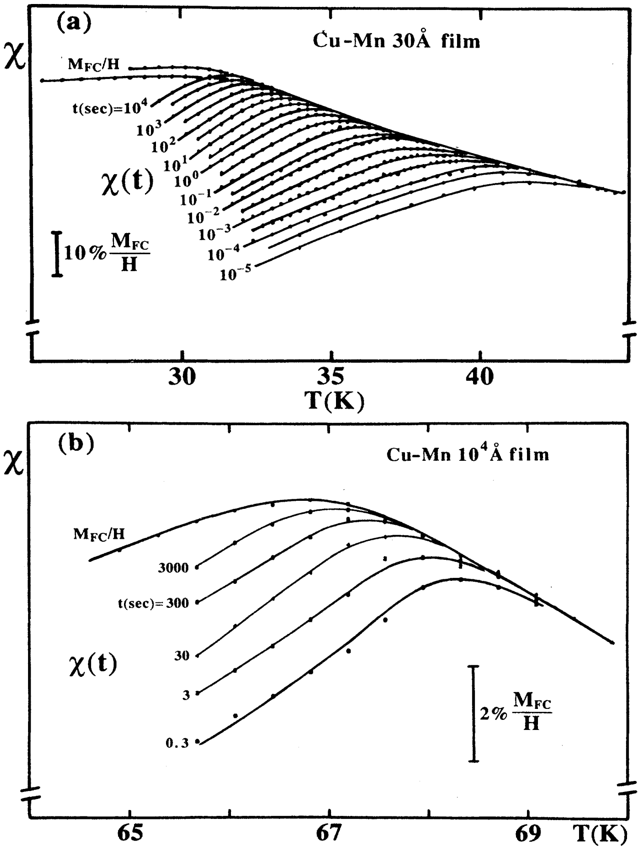

FIG. 1. Temperature dependence of the dynamic susceptibility for two Cu(13.5 at.% Mn) spin-glass films at different observation times. The dynamic susceptibility was obtained from ZFC susceptibility data in the time range 3×10^{-1}–10^{4} sec and from ac susceptibility data in the range 10^{-5}–3×10^{-1} sec. (a) 30-Å film. The FC susceptibility (M_{FC}/H), at two different cooling rates (0.9 K/min, lower curve and 0.02 K/min, upper curve), are also included. (b) 10^4-Å film. The FC susceptibility (M_{FC}/H) at the cooling rate 0.02 K/min is also included.

The main message of this paper is the observation of a dramatic change of the behavior of the dynamic susceptibility on one and the same spin-glass material on decreasing the thickness from 10^4 to 30 Å. An illustrative and model-independent evaluation of the frequency dependence of the freezing temperature T_f is obtained from the quantity $k=(1/T_f)\,dT_f/d\log_{10}t$. For the 10^4-Å film $k=1/200$ at $t\approx1$ s. This value falls into the general range of k values observed in earlier investigations on bulk Cu-Mn (Ref. 13) Ag-Mn (Ref. 14) spin glasses. This and the sharpness of the ac-susceptibility maxima do show that the dc-sputtered "thick" film exhibits typical bulk Cu-Mn spin-glass properties. For the 30-Å film the relative shift in T_f with frequency was found to be 1/40. These results can be compared to results from MC simulations on short-range Ising spin-glass models. In a 2D model[4] $k\approx1/20$ is obtained when extrapolating the time scales of MC simulations to observation times typically used in experiments on real spin glasses. Adopting the same extrapolation procedure on results from a 3D model[2] $k\approx1/60$ is achieved. Thus, there is a qualitative agreement between the present experimental results and MC simulations as to the time dependence of the freezing temperature on dimensionality.

The results from various experimental studies on the spin-glass dynamics on different systems have not led to a unique model for the physical processes behind the dynamic critical behavior. In conventional critical slowing down the relaxation times diverge, when approaching the critical point, as

$$\tau_{\max}=\tau_0\left((T_f-T_g)/T_g\right)^{-z\nu}, \tag{2}$$

where z is the dynamic exponent, ν is the four-spin correlation-length exponent, and τ_0 is a microscopic relaxation time. By defining the cusp temperature of the time-dependent susceptibility curves as the freezing temperature T_f and the corresponding observation time t (or $1/\omega$) as $\tau_{\max}$, ZFC data for the 10^4-Å film analyzed according to Eq. (2) give $z\nu = 9 \pm 1$, and $T_g = 66 \pm 0.2$ K, using $\log_{10}\tau_0 = -13 \pm 1$. The value of $z\nu$ is in good agreement with results obtained from experiments on some different 3D spin-glass materials.[1,15] The value of T_g is close to the temperature of the FC-susceptibility maximum (66.8 K) which is the common observation in conventional dynamic scaling. Using data extracted from both the real part of the ac susceptibility and the time-dependent ZFC susceptibility of the 30-Å film the following parameters are obtained: $\log_{10}\tau_0 = -10$, $z\nu = 19$, and $T_g = 26$ K. Despite a reasonable quality of the fit, the extracted values of the parameters are quite unphysical. This implies that the dynamics of the 30-Å film is poorly described within the framework of conventional critical slowing down.

Another scaling form is associated with thermally activated processes. In particular there is a general belief[16] that the dynamics for two-dimensional spin-glass systems should be governed by such processes. In a theoretical approach based on the droplet scaling theory[7] the following form for the relaxation times at a zero-temperature transition is derived:

$$\ln(\tau_{\max}/\tau_0) \propto T_f^{-(1+\psi\nu)} , \tag{3}$$

where ψ is a barrier exponent. A fit of our data (which covers the wide time interval $10^{-5} < t < 10^4$ sec) to Eq. (3) is shown in Fig. 2 and gives $\psi\nu = 1.6 \pm 0.2$, using $\log_{10}(\tau_0) = -13 \pm 1$. The value of $1 + \psi\nu$ is in reasonable agreement with results from MC simulations which give a corresponding value of approximately 2.[3,4] Assuming a finite critical temperature the relaxation times obey

$$\ln(\tau_{\max}/\tau_0) \propto (T_f - T_g)^{-\psi\nu} T_f^{-1} . \tag{4}$$

Adopting this relation a good scaling is obtained for $0 < T_g < 10$ K (giving $1.6 > \psi\nu > 1.1$) with a best fit at $T_g = 4$ K.

These results indicate a zero-temperature transition. At least, the significant feature for the 30-Å film is the very low critical temperature in relation to the temperature of the FC-susceptibility cusp (30 K). In contrast, activated dynamic scaling on a short-range 3D spin glass[1] gives a critical temperature only 5% lower than the temperature of the FC-susceptibility cusp. A similar result is also found on bulk Cu-Mn systems analyzed according to a Vogel-Fulcher behavior ($\psi\nu = 1$) of the relaxation times.[13] It is found that T_g is approximately 7% lower than the freezing temperature at 10 Hz, independent of Mn concentration within the range 1–10%. Assuming a Vogel-Fulcher behavior our data on the 10^4-Å film gives $T_g = 62.5$ K (i.e., 6% lower than the FC-susceptibility maximum). An analysis with $\psi\nu$ as a free parameter yields $\psi\nu = 0.8$ and $T_g = 63.5$ K. These distinct differences regarding the dynamics between the 30-Å film and various bulk samples indicate a *dimensionality crossover* when decreasing the film thickness. One of the significant problems in interpreting these results as evidence of crossover behavior involves the question of whether they could arise from physical changes which occur as the film thickness is reduced. Two such changes are a modification in the short-range chemical order of the Mn impurities or attenuation of the RKKY interaction due to mean-free-path effects. Although we cannot unequivocally rule out the presence of either of these effects, there are no detailed predictions of the dynamic behavior which would result, while the predictions of the droplet scaling model[7] seem to fit the observed behavior very well.

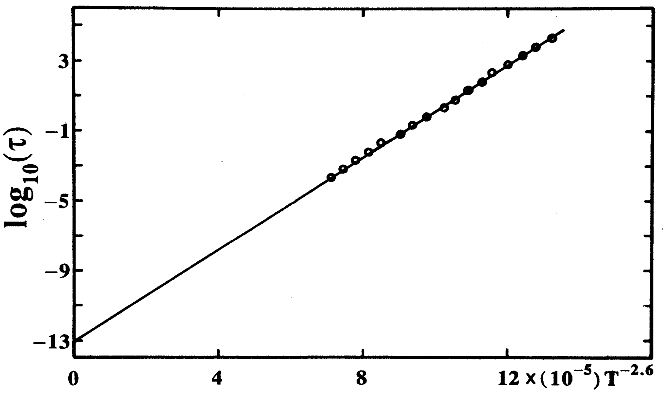

FIG. 2. A fit of the relaxation times on the 30-Å Cu(13.5 at.% Mn) film according to a generalized Arrhenius law, $\log_{10}(\tau)$ vs $T^{-2.6}$

Spin glasses are subject to an aging process, which is associated with a crossover between *equilibrium dynamics* at short observation times ($\ln t \ll \ln t_w$) and *nonequilibrium dynamics* at long times ($\ln t \gg \ln t_w$) (Ref. 17) (t_w is the wait time at constant temperature before the dynamics are probed). In ZFC measurements the effect of aging is seen as an inflection point in the $M(t)$ vs $\log_{10}(t)$ curve at an observation time of the order of the wait time. Figure 3 shows the ZFC susceptibility versus $\log_{10}(t)$ at wait times $t_w = 10^2$ and 10^4 sec for the two films. The figure illustrates striking differences in the relaxation behavior of the two films. The relaxation rate $(1/H\, \partial M/\partial \ln t)$ is roughly five times larger in the 30-Å film than in the 10^4-Å film whereas the influence of aging is more pronounced for the 10^4-Å film than for the 30-Å film. These types of differences of the relaxation behavior between a 3D and a 2D system are predicted by the droplet scaling theory.[7] However, a quantitative comparison to the theoretical predictions demands extensive experimental investigations of the dynamics in wide time intervals, using different wait times and at different temperatures.

In conclusion, this investigation shows that a 30-Å Cu-Mn film exhibits a dynamical behavior very similar to results from MC simulations[3,4] on 2D spin-glass models and in agreement with the predictions of the droplet scaling theory.[7] Clear indication of a dimensionality crossover from three- to two-dimensional behavior on decreasing the film thickness from 10^4 to 30 Å is found. Time-dependent susceptibility measurements on films of varying thickness represent a unique method to investigate the physical consequences of a dimensionality crossover between 3D and 2D. Within the droplet scaling theory[7] the dynamics at a given *observation time* is directly associated with a corresponding *length scale* in the spin-glass system. By observing the time scales for a crossover between 3D to 2D dynamics a direct measure of the connection between time and length scales in spin glasses is obtained. Such experiments are in progress at our laboratories.

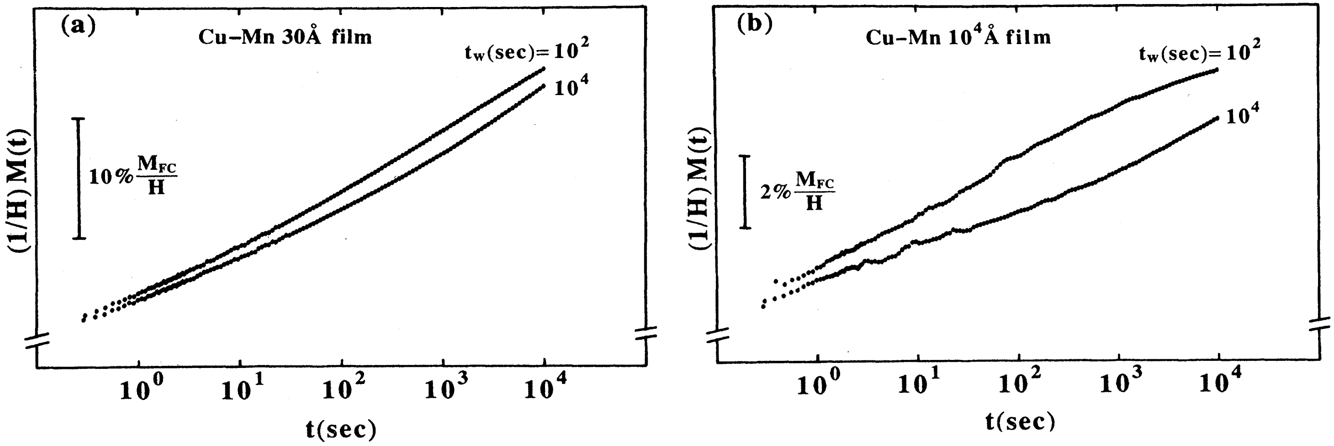

FIG. 3. Time dependence of the ZFC susceptibility at different wait times ($t_w = 10^2$ sec and $t_w = 10^4$ sec) for two Cu(13.5 at.% Mn) spin-glass films. (a) 30-Å film, 10% of the FC susceptibility value indicated, $T = 24.7$ K. (b) 10^4-Å film, 2% of the FC susceptibility value indicated, $T = 61.2$ K.

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