

Evidence of Dimensional Crossover of the Spin-Glass Transition in Thin CuMn Multilayers

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We have used high-quality $\text{Cu}_{92}\text{Mn}_8/\text{Al}_2\text{O}_3$ multilayers to investigate the shift in temperature and the change in scaling behavior of the spin-glass transition as a function of the layer thickness. In the thinnest CuMn layers (60 and 88 Å), the spin-glass exponents determined from the nonlinear susceptibility shift to new values, distinctly different from those for the bulk. These exponents are consistent with numerical studies of 2D spin-glass systems, suggesting that a 3D to 2D crossover occurs.

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Recent years have seen a great deal of interest in spin glasses, particularly in the lower critical dimension (d_l).¹ Since the spin-glass transition has not been observed at finite temperature in two dimensions, either in a real material² or in numerical studies,^{3,4} d_l appears to be between 2 and 3. Kenning, Slaughter, and Cowen recently made the interesting observation that the spin-glass transition temperature of CuMn multilayers interleaved with Si, SiO₂, and Cu decreases as the thickness of the spin-glass layer is decreased.^{5,6} They attributed this to finite-size effects, but did not demonstrate the crossover to two dimensions, nor the two-dimensional behavior of the thin layers.

In this Letter, we use the linear susceptibility (χ_l) and the scaling behavior of the nonlinear susceptibility (χ_{nl}) of high-quality CuMn multilayers to determine both the shift exponent λ and the critical exponents which govern the spin-glass transition. We show that the spin-glass exponents shift from values characteristic of bulk CuMn to new values which agree with those obtained in numerical studies of 2D spin-glass systems^{7,8} but differ from those found by Monte Carlo simulations.³

Concerns of interfacial alloying, transmission of the Ruderman-Kittel-Kasuya-Yosida interaction across the inert layer, and contrast in both transmission electron microscopy and x-ray diffraction have led to the choice of Al₂O₃ as a spacer material. Our experience with granular metal solids shows that Al₂O₃ is immiscible to both Cu and Mn. Nanocrystals of each of these metals, as small as 20 Å in diameter, have been prepared in an Al₂O₃ matrix and shown by electron microscopy and x-ray diffraction to retain their integrity and crystal structures. These results indicate a sharp metal-insulator interface: Thus a clean CuMn-Al₂O₃ multilayer may be anticipated. The CuMn alloy used had the composition Cu₉₂Mn₈, nearly the same as those used in some of the experiments reported by Kenning and co-workers. Our multilayer samples were fabricated in a multigun magnetron sputtering chamber with a computer-controlled

shutter system. The samples have a fixed Al₂O₃ layer thickness of 75 Å, and the CuMn layer varies within the thickness range $40 \text{ Å} \leq w \leq 1250 \text{ Å}$. In addition, a single-layer film 4.5 μm thick serves as the bulk specimen. The high quality of these samples is verified by both low-angle x-ray diffraction and cross-sectional transmission electron microscopy (TEM) as illustrated in Fig. 1. The intensity modulation of the superlattice peaks in the x-ray-diffraction pattern is an expected consequence of very good multilayers. All of the films to be discussed here were deposited on (100) Si wafer substrates, and were removed from those substrates for the magnetic measurements.

Magnetic-susceptibility measurements were conducted on commercial SQUID magnetometers. The spin-glass temperature of each sample $T_g(w)$ was identified by the cusp in the zero-field-cooled scan of χ_l vs T (not shown).

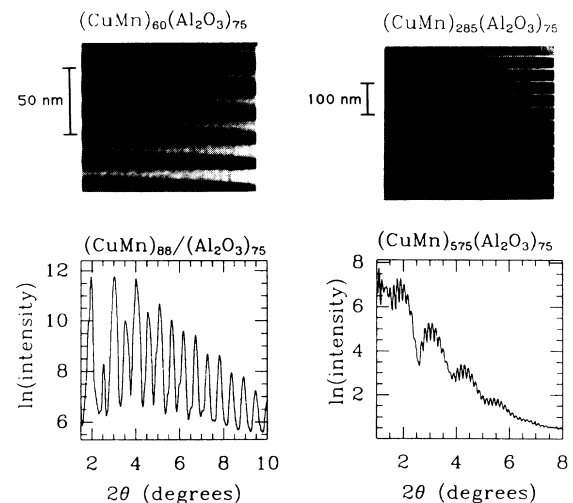


FIG. 1. Representative TEM micrographs and low-angle x-ray-diffraction data of $\text{Cu}_{92}\text{Mn}_8/\text{Al}_2\text{O}_3$ multilayer samples with $w=60, 88, 285,$ and 575 Å . In the micrographs, the dark bands are the CuMn.

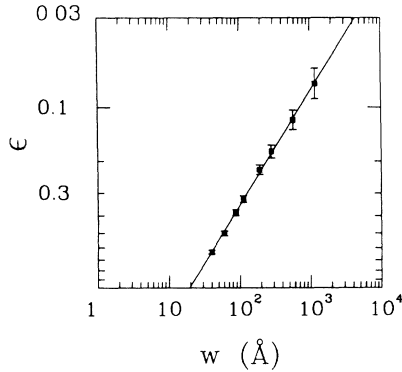


FIG. 2. Reduced temperature $\epsilon = [T_g(\infty) - T_g(w)]/T_g(\infty)$ vs w , the layer thickness of the $\text{Cu}_{92}\text{Mn}_8$. The solid line is the fit by Eq. (1).

In order to illustrate the scaling behavior of T_g , we have plotted the reduced temperature $\epsilon = [T_g(\infty) - T_g(w)]/T_g(\infty)$ versus the layer thickness w on a log-log scale in Fig. 2. We have found that *all* of the data can be described by the simple finite-size scaling relation

$$\frac{T_g(\infty) - T_g(w)}{T_g(\infty)} = \epsilon = Aw^{-\lambda} = \left(\frac{w}{w_0}\right)^{-\lambda}, \quad (1)$$

with $\lambda = 0.64 \pm 0.07$, $A = 6.7 \pm 0.25$, and $w_0 = 19.5 \pm 1.1$ Å. The value of $\lambda = 0.64$ is quite close to the value which was obtained by Kenning, Slaughter, and Cowen⁵ for the thickest films in that study: $\lambda = 0.63 \pm 0.15$ for $w \geq 150$ Å. The solid line represents these calculated results. The excellent fit led us to the unexpected conclusion that a single shift exponent λ is sufficient to describe the data for all thicknesses, and without resorting to corrections such as the logarithmic factor suggested by Fisher and Huse.⁹

In ferromagnetic films, no deviation of T_c can be observed until the films are only a few atomic layers thick. In contrast, it is remarkable to note that significant deviations from the bulk value of $T_g(\infty) = 34$ K may be observed even in the samples with the thickest spin-glass layers ($w = 1250$ Å). Specifically it is the large value of the constant A (or the zero-temperature correlation length w_0) which is responsible for the effect, and which in turn makes this measurement possible. While theoretical calculations focus on scaling behavior and critical exponents, the large value of w_0 has not been addressed. Our result that $T_g \rightarrow 0$ for $w = w_0 \approx 20$ Å was also found by Kenning and co-workers, although their critical thickness is twice as large at 40 Å.

The nonlinear susceptibility is defined as¹⁰

$$\chi_{nl}(H, T) = \chi_l(T) - M(H, T)/H, \quad (2)$$

where $\chi_l(T)$ is the linear susceptibility. We have measured $\chi_l(T)$ by extrapolating to zero field from our measurements at $H \leq 10$ mT. Measurements were conducted in the temperature range from 5 K to approximately

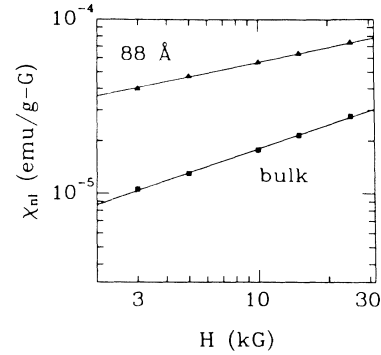


FIG. 3. Nonlinear susceptibility χ_{nl} vs H at the critical isotherms for bulk and for thin layers of CuMn ($w = 88$ Å). The solid lines are fits by Eq. (3).

$2T_g$. The two thin-layer samples were separately measured on different magnetometers at the two institutions with good agreement. We expect χ_{nl} to obey the single-parameter scaling relation¹⁰

$$\chi_{nl}(H, T) \propto H^{2/\delta} f(t/H^{2/\phi}). \quad (3)$$

Here, $t = (T - T_g)/T_g$, δ and ϕ are spin-glass critical exponents which are related to the susceptibility exponent by $\gamma = \phi(1 - 1/\delta)$, and f is a scaling function with the asymptotic properties

$$f(x) = \begin{cases} \text{const}, & x \rightarrow 0, \\ x^{-\gamma}, & x \rightarrow \infty. \end{cases} \quad (4)$$

The critical exponent δ can be determined from the critical isotherm. In Fig. 3, we show a log-log plot of χ_{nl} vs H for the bulk spin glass and for the sample with layer thickness $w = 88$ Å. The slopes give $\delta = 4.4 \pm 0.2$ for the bulk specimen, similar to that obtained previously,¹¹ and very different from $\delta = 7.1 \pm 0.2$ for the thin-layered sample.

Figure 4 shows the full set of $\chi_{nl}(H, T)$ data for the sample with $w = 60$ Å. As expected, the sharp peak usually associated with χ_l does not appear in these data which are taken at high fields ($H \geq 1$ kG). The data were analyzed by seeking those values of the exponents δ and ϕ such that the data collapse onto a single scaling curve as suggested by Eq. (3). Excellent scaling behavior is revealed by the choices of $T_g(w) = 17.5 \pm 0.25$ K, $\delta = 7.0 \pm 0.5$, and $\phi = 6 \pm 1$ as shown in Fig. 5, where we plot $\chi_{nl}/H^{2/\delta}$ vs $t^{\phi/2}/H$. Because of the irreversible and time-dependent nature of the response below T_g we have included only the data from the range $T > T_g$ as indicated in the legend. In the limit of $t^{\phi/2}/H \rightarrow 0$, we indeed observe a constant value, confirming that $\chi_{nl} \propto H^{2/\delta}$. In the limit of large $t^{\phi/2}/H$ (i.e., at small fields), we observe an asymptotic slope of $-1.7 = -2\gamma/\phi$, according to Eq. (4). Combined with the scaling relation $\gamma = \phi(1 - 1/\delta)$, the value of 1.7 leads to $\delta = 6.6$, again consistent with Fig. 3.

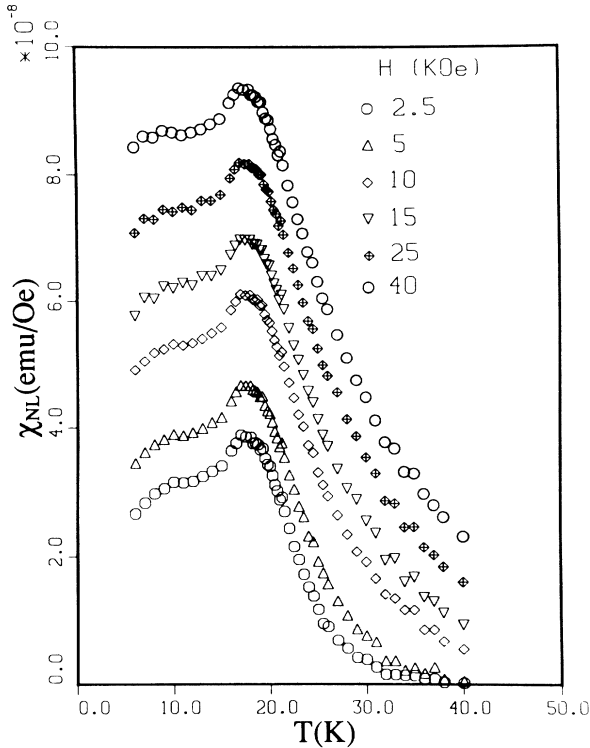


FIG. 4. Nonlinear susceptibility χ_{nl} of CuMn with thickness $w = 60 \text{ \AA}$. The transition occurs at $T_g = 17.5 \text{ K}$ for this sample.

Table I summarizes the critical exponents we have obtained from χ_{nl} for thin layers and bulk spin glasses. The quantities with errors are the directly determined values, whereas those without errors are derived from the scaling laws. One well-known prediction of finite-size scaling theory is that the shift exponent should be given by the inverse of the correlation-length exponent, i.e., $\lambda = \nu^{-1}$. We have used this along with our value of δ to obtain values for the remaining exponents for the bulk spin glass, as given in Table I. The agreement with values for a wide variety of bulk spin glasses is quite good. Lévy and Ogielski¹² have determined the values $\beta = 0.9 \pm 0.2$, $\delta = 3.3 \pm 0.2$ for bulk AgMn, and Barbara, Malozemoff, and Imry have found $\gamma = 3.4 \pm 0.4$, $\delta = 4.15 \pm 0.15$.¹¹

The literature on theoretical calculations of the 2D exponents is somewhat fragmentary, thus we have calculated the values in the last column of Table I from the results for γ and ν taken from numerical studies, which appear to be more reliable than the Monte Carlo results. Although such calculations must be regarded with some caution, it is gratifying to note the excellent agreement with the experimental results from the 60- and the 88-Å multilayers. Furthermore, it is important to stress that the large differences between the values determined for the layered samples and those for bulk are suggestive of a dimensional transition, without reference to the theoretical values.

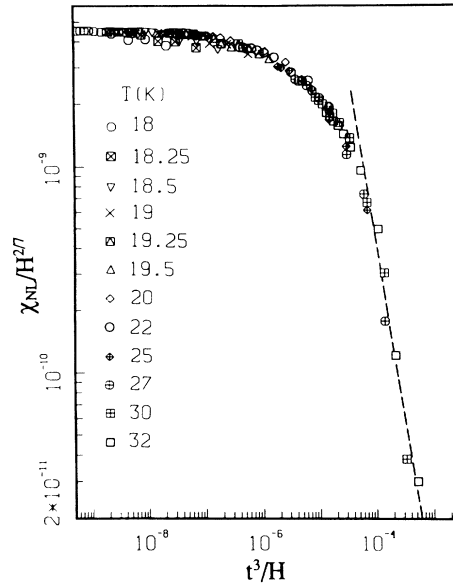


FIG. 5. Scaling plot of $\chi_{nl}/H^{2/\delta}$ as a function of $t^{\delta/2}/H$ for CuMn with thickness $w = 60 \text{ \AA}$, where $t = (T - T_g)/T_g$, $\delta = 7$, $\phi = 6$, and $T_g = 17.5 \text{ K}$.

It has been recognized that the correlation lengths that characterize spin freezing are rather long, making finite-size effects important.¹³ According to this model, as the temperature is reduced to a crossover temperature $T_x > T_g$ the correlation length exceeds the layer thickness w . This in turn causes the sample to behave quasi two dimensionally—with the critical exponents of a two-dimensional system, but with a nonzero freezing temperature $T_g(w)$. Where T_g is significantly reduced, we expect $T_x \approx T_g(\infty)$.¹⁴ Thus, we expect that for $T_g(w) < T < T_g(\infty)$ the exponents governing the various thermodynamic quantities will take on their two-dimensional values. As seen in Table I, our results suggest such a dimensional crossover. The experimental temperature range for the thin-layer sample is clearly well within the 2D regime. It is incorrect to assume, as have Sandlund *et al.*,¹⁵ that 2D effects are only observed in conjunction with a $T = 0 \text{ K}$ transition.

TABLE I. Experimental and theoretical values of the spin-glass critical exponents.

Exponent	60 Å	88 Å	Bulk	2D Ising
δ	7 ± 0.5	7.1 ± 0.2	4.4 ± 0.2	(6.6)
ϕ	6 ± 1	6 ± 1	3.9	(6.2)
β	0.86	0.85	0.9	(0.95)
γ	5.1	5.2	3.0	5.3 ± 0.3^a
ν	$3.4 (d=2)$	$3.4 (d=2)$	1.6 ± 0.2	3.6 ± 0.1^b
η	0.5	0.49	0.11	(0.53)

^aReference 7.

^bReference 8.

In summary, 3D spin-glass ordering appears to be highly susceptible to finite-size effects. As noted by Kenning and co-workers, this may be related to the large regions of coherent spin ordering observed in neutron scattering.¹⁶ We have observed for the first time 2D scaling behavior in thin layers of CuMn spin glasses. The combination of finite-size scaling in Ref. 5 and the 2D behavior reported here adds considerable weight to the current picture of phase transitions in Ising-like spin glasses.

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¹K. Binder and A. P. Young, *Rev. Mod. Phys.* **58**, 801 (1986).

²C. Dekker, A. F. M. Arts, and H. W. de Wijn, *Phys. Rev. B*

38, 11512 (1988).

³R. N. Bhatt and A. P. Young, *Phys. Rev. B* **37**, 5606 (1988).

⁴A. J. Bray and M. A. Moore, *J. Phys. C* **17**, L463 (1984).

⁵G. G. Kenning, J. M. Slaughter, and J. A. Cowen, *Phys. Rev. Lett.* **59**, 2596 (1987).

⁶J. A. Cowen, G. G. Kenning, and J. M. Slaughter, *J. Appl. Phys.* **61**, 4080 (1987); J. A. Cowen, G. G. Kenning, and J. Bass, *J. Appl. Phys.* **64**, 5781 (1988).

⁷W. L. McMillan, *Phys. Rev. B* **29**, 4026 (1984).

⁸R. R. P. Singh and S. Chakravarty, *Phys. Rev. Lett.* **57**, 245 (1986).

⁹D. S. Fisher and D. A. Huse, *Phys. Rev. B* **36**, 8937 (1987).

¹⁰J. Chalupa, *Solid State Commun.* **22**, 315 (1977); M. Suzuki, *Prog. Theor. Phys.* **58**, 1151 (1977).

¹¹B. Barbara, A. P. Malozemoff, and Y. Imry, *Phys. Rev. Lett.* **47**, 1852 (1981).

¹²L. P. Lévy and A. T. Ogielski, *Phys. Rev. Lett.* **57**, 3288 (1986).

¹³D. A. Huse and I. Morgenstern, *Phys. Rev. B* **32**, 3032 (1985).

¹⁴P. Pfeuty and G. Toulouse, *Introduction to the Renormalization Group and to Critical Phenomena* (Wiley, New York, 1977), p. 144.

¹⁵L. Sandlund, P. Granberg, L. Lundgren, P. Norblad, P. Svedlindh, J. A. Cowen, and G. G. Kenning, *Phys. Rev. B* **40**, 869 (1989).

¹⁶J. A. Gotaas, J. J. Rhyne, and S. A. Werner, *J. Appl. Phys.* **57**, 3404 (1985).

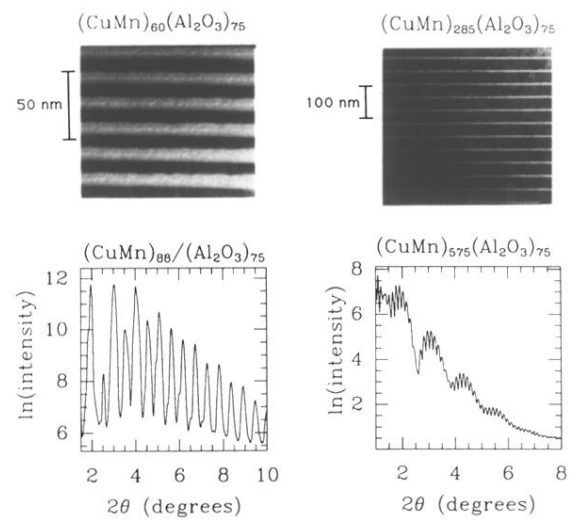


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