Finite-Size Effects in a CuMn Spin-Glass

G. G. Kenning, J. M. Slaughter, and J. A. Cowen
Department of Physics and Center for Fundamental Materials Research, Michigan State University, East Lansing, Michigan 48824
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Using dc sputtering in an UHV system we have prepared multilayer samples of Cu:Mn-Si with Cu:Mn thicknesses, L, between 4 and 500 nm. The temperature of the peak in the dc magnetic susceptibility of these spin-glass samples shifts with L as $(T_{g0}-T_g)/T_{g0} \approx L^{-0.75 \pm 0.06}$ over the whole range of sample thickness. These results are discussed in terms of finite-size scaling and the possible effects of the electron mean free path on the effective exchange interaction.

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Recently, there has been a great deal of theoretical interest in lower critical dimensions[1] and finite-size scaling[2] as applied to spin-glasses. The absence of experimental results to compare with theoretical predictions can be attributed to the difficulty in fabricating and characterizing spin-glass samples that are thin enough so that finite-size effects can be observed. For example, at 40 K in an applied field of 100 G a single 1-cm$\times$1-cm$\times$4-nm layer of Cu:(7%Mn) has a magnetization of approximately 1×10^{-8} emu, which is very close to the noise limit of commercial SQUID susceptometers. In order to produce large enough magnetization signals for quantitative studies, we have fabricated samples with multiple layers of Cu:Mn alternated with Si. As we shall show, the Si decouples the spin-glass layers both electrically and magnetically, which allows us to analyze the experimental results in terms of isolated spin-glass layers. The multilayer structure also allows us to determine the average thickness of the Cu:Mn-Si bilayer using low-angle x-ray diffraction and transmission electron microscopy (TEM). We report here results for a series of Cu:Mn (7 wt.%) samples with CuMn layer thicknesses between 4 and 500 nm which appear to show finite-size effects.

The samples were produced by dc sputtering in an UHV sputtering system that was pumped to $< 10^{-8}$ Torr before the Ar sputtering gas was admitted to the chamber. The Cu:Mn targets were made from 99.9999+% pure Cu and 99.99+% pure Mn, melted together in an rf vacuum furnace. The Si was a commercial target made from 99.9999+% pure Si which had a room-temperature resistivity $\gg 10\ \Omega$ cm. The films were deposited on $1.2\times 1.2\times 0.5$-cm^3 polished single-crystal Si substrates, which were positioned on a rotatable substrate holder 11 cm from the targets. The sputtering rates were determined with a quartz-crystal film thickness monitor, and the nominal layer thicknesses were calculated as the product of the sputtering rates and the time during which the substrates were positioned over each sputtering source. Typical rates were 1.5 nm/sec for the Cu:Mn alloy and 0.3 nm/sec for the Si. Low-angle x-ray studies verified that the average period of the structure was within a few percent of that calculated from the sputtering rates. Two samples of each concentration and thickness were made simultaneously, one for magnetic measurements and the other for sample characterization—including x-ray diffraction, TEM, and electrical conductivity studies. To verify that the concentration of Mn in the sputtered films was the same as that in the target, 1-μm-thick "bulk" Cu:Mn films were produced. Both chemical analysis and the temperature, T_{g0}, of the maximum in the magnetic susceptibility gave essentially identical results for the sputtered films and for material directly removed from the target.

The nominal thickness of the Si layers in all of our samples was 7 nm. Separate experiments with a fixed Cu:Mn layer thickness of 7 nm and Si thicknesses between 3 and 14 nm showed that T_g was independent of Si thickness down to at least 3 nm. It is thus clear that 7 nm of Si decouples the Cu:Mn layers very effectively. We are in the process of investigating how thin the Si layers must be before coupling between the CuMn layers becomes significant.

The quality of the samples was characterized by three techniques. Low-angle x-ray diffraction, using a θ-2θ sweep on a Rigaku model CN2013 diffractometer, provided information about the period of the multilayers. Table I gives the results for several samples with periods between 13 and 27 nm. The lowest-order peaks were hidden in the central maximum, but we regularly observed between five and ten orders up to n = 16. That we were able to observe such high-order maxima indicates that the layering in our samples was reasonably uniform.

TEM photographs were also taken of transverse sections of the layers. Dr. John Heckman of the Michigan State University Center for Electron Optics prepared the sections using a microtome and took the photographs. These showed layers of the correct thickness, but with irregular boundaries that contained a great deal of detailed structure. It is not yet clear how much of this structure is intrinsic, and how much is due to fracture

TABLE I. Bilayer thickness determined by low-angle x-ray diffraction compared with that calculated from the product of the sputtering rate and the time. The error in each determination is approximately 3%.

Sample	No. of layers	Orders observed	Period (nm)	
			X-ray	Sput.
23B	26	7 to 17	27.7	27.0
24B	31	6 to 11	16.9	17
28B	67	4 to 8	13.4	13
30B	60	4 to 10	10.1	10.0
32B	84	5 to 11	11.9	12.0
33B	44	9 to 18	21.1	21.0
53B	15	9 to 12	25.6	27.0
55B	43	5 to 11	13.9	14.0
56B	60	4 to 8	12.2	12.0

and deformation produced by the microtome.

To check whether the Cu:Mn might exhibit island formation in the thinnest samples, we measured the electrical resistances of the samples parallel to the layer planes. Our contacts were made by scratching through the layer edges and attaching wires with drops of silver paint in an approximate Van der Pauw geometry.[3] Figure 1(a) is a graph of the resistivities of the samples plotted versus Cu:Mn layer thickness, L. We see that the resistivity remains finite down to the lowest value of L tested. Figure 1(b) shows the temperature dependence of the resistivity for different film thicknesses. For $L > 7.0$ nm, the films show metallic behavior, while for $L < 7.0$ nm the resistivity increases with decreasing temperature.

The dc magnetic susceptibility was measured on a commercial SHE Corp. model VTS SQUID susceptometer. The samples were cooled to 5 K (2 K for $L \leq 5$ nm) in zero field, a measuring field of up to 200 G was applied, and the temperature was cycled up to 60 K and then back down again. This procedure provided both zero-field-cooled and field-cooled data in the same run. Figure 2 summarizes several zero-field-cooled and field-cooled runs for different values of L. T_g is defined as the temperature of the maximum in the zero-field-cooled

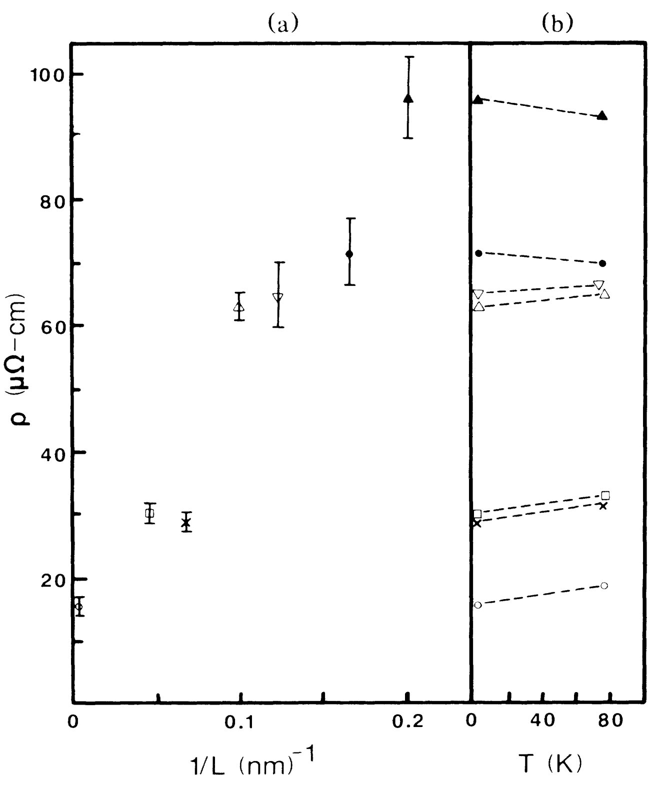

FIG. 1. (a) Resistivity of multilayer films plotted against the reciprocal of the Cu:Mn thickness. (b) The temperature dependence of the resistivities of the corresponding films. 500 nm, open circles; 15 nm, crosses; 20 nm, squares; 10 nm, open triangles; 7 nm, inverted triangles; 6 nm, filled circles; 5 nm, filled triangles; Note that the data were taken at 77 and 4.2 K.

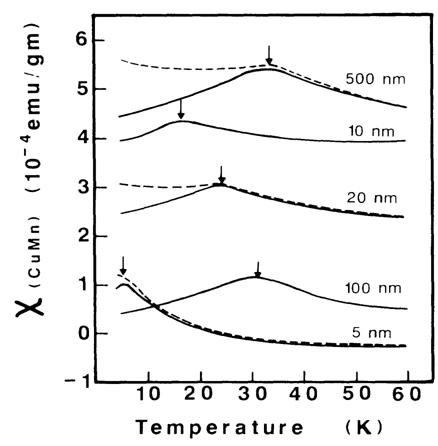

FIG. 2. The temperature dependence of the dc magnetic susceptibility of the multilayer films for different Cu:Mn thicknesses. Solid lines are for zero-field cooling while dashed lines are for field cooling.

susceptibility. The peaks in the susceptibility in Fig. 2 are more rounded than one finds in well annealed and etched bulk samples; this is not surprising, since the density of strains and defects in our samples is likely to be large. It appears that the peaks become somewhat sharper with decreasing L.

The values of T_g for a series of Cu:Mn (7 wt.%) samples prepared in three independent sputtering runs are shown in Fig. 3. T_g begins to deviate from its bulk value of 35 K for L near 100 nm, and apparently approaches zero for L near 4 nm. The major part of the decrease in T_g occurs below 10 nm.

We first consider the behavior at large L. We believe that the downward shift in T_g with decreasing L shown in Fig. 3 is due to the limitation in the correlation length of the spin-glass system as L becomes smaller than the bulk correlation length. Levy and Ogielski[4] have inferred a bulk correlation length of $\simeq 200$ nm from their measurements of the nonlinear susceptibility of Ag:Mn (0.5 wt.%). If we assume that this correlation length is not very concentration or host dependent, their value is quite close to the value of 100 nm where T_g begins to shift downward from its bulk value.

Finite-size scaling theory[2,5] predicts that, in the asymptotic limit of large L, the fractional temperature shifts, $\epsilon = (T_{g0} - T_g)/T_{g0}$, should vary as $\epsilon \approx L^{-\lambda}$, where λ is the shift exponent. Figure 4 shows a plot of ϵ vs L. The data were fitted by means of a nonlinear fitting program. Within the limits determined by the error bars, the data fit

$$\epsilon = L^{-0.75 \pm 0.06}$$

over the whole range $0.06 < \epsilon < 0.82$. If one assumes that $\lambda = 1/\nu$ where ν is the correlation length exponent, our value is in good agreement with $\nu = 1.3 \pm 0.2$ obtained by Levy and Ogielski[4] from their nonlinear susceptibility measurements. It is also in agreement with the values obtained for the 3D Ising model using simulation techniques.[6] While this agreement with the 3D Ising model prediction is intriguing, it is not obvious that the 3D Ising model is appropriate to Cu:Mn. It is also difficult to see why one should observe the same scaling behavior over the whole range of reduced temperature shown in Fig. 4, especially in view of the possible mean-free-path effects which we discuss next.

The increase in resistivity [Fig. 1(a)] with decreasing layer thickness suggests an attenuation of the Ruderman-Kittel-Kasuya-Yosida interaction. We have applied Larsen's[7] quenched uniform model to the layered geometry in order to estimate the reduction in T_g due to mean-free-path effects. Although this is a simple spin-glass model, it should give a reasonable estimate of the shift in T_g.

The calculation shows that mean-free-path effects suppress the transition temperature by as much as 14 K in our thinnest samples. However, the shift is less than 1 K for resistivities $\leq 30\ \mu\Omega$ cm, and thus for samples thicker than about 15 nm. This is in qualitative agreement with the data of Vier and Schultz.[8] By the addition of nonmagnetic impurities to Ag:Mn (2.6 wt.%) they produce shifts in T_g of approximately 1 K for resistivity changes of 50% from "pure" Ag:Mn.

Simple scaling-law analysis should break down when the electron mean free path becomes sufficiently short. If, using the above argument, we fit our data only for $L \gtrsim 15$ nm we obtain the dashed line in Fig. 4 for which $\lambda = 0.63 \pm 0.15$.

In the small-L regime the resistivity increases vary rapidly with decreasing L, implying a correspondingly

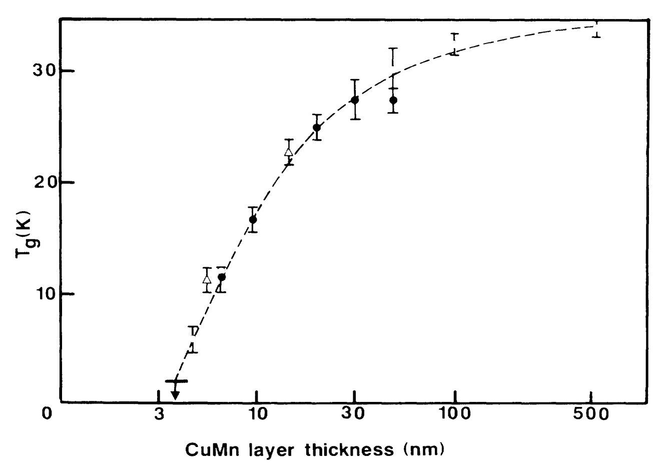

FIG. 3. Temperature of the peak in the magnetic susceptibility vs Cu:Mn thickness. In all cases the Cu:Mn layers were separated by 7 nm of Si. The various symbols pertain to samples from three different sputtering runs and the dashed line is provided to guide the eye.

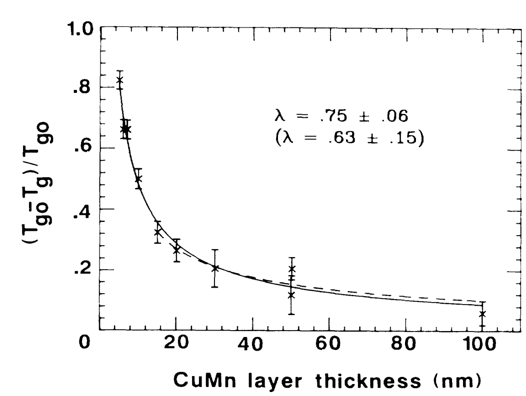

FIG. 4. The reduced temperature $\epsilon = (T_{g0} - T_g)/T_{g0}$ vs the Cu:Mn layer thickness. The solid line is the fit to all eleven data points and gives the exponent $\lambda = 0.75 \pm 0.06$. The dashed line is the fit to the thickest six samples and gives $\lambda = 0.63 \pm 0.15$.

rapid reduction in the electron mean free path. This suggests that the rapid dropoff and vanishing of T_g at small L is enhanced by a modification of the conduction-electron–mediated exchange interaction in the Cu:Mn due to mean-free-path effects.

Independent evidence of a short length scale is obtained from Werner[9] who has derived a coherence length of $\simeq 4$ nm for Cu:Mn samples from measurements of the width of incommensurate satellite peaks that he observes with neutron scattering. While the positions of the peaks depend on Mn concentration, the widths of the peaks do not. This coherence length is suspiciously close to the value of L for which $T_g \rightarrow 0$ K in our samples.

To summarize, we have produced Cu:Mn (7 wt.%) spin-glass films between 4 and 500 nm thick in the form of multilayers with Si interlayers of sufficient thickness to decouple the films both magnetically and electrically. The shift in the reduced temperature of the peak in the susceptibility, ϵ, can be fitted over the whole range of thickness with a single exponent. Resistance measurements on the same samples give evidence of mean-free-path effects in the thin layers. This appears to be the first evidence of finite-size effects in a spin-glass system.

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