

## Finite-size effects in thin Ag-Mn spin-glass layers

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Measurements of the quasistatic spin-freezing temperature  $T_f$ , as a function of spin-glass layer thickness,  $W_{\text{Ag-Mn}}$ , are presented for multilayer samples of the spin-glass  $\text{Ag}_{1-x}\text{Mn}_x$  ( $x=0.04$  and  $0.09$ ) alternated with interlayers of Ag or Cu having thickness  $W_{\text{il}}=30$  nm, a value large enough to magnetically decouple the spin-glass layers. The values of  $T_f$  are independent of whether the interlayer metal is Ag or Cu, and thus insensitive to grain sizes and structural details of spin-glass-interlayer interfaces. To within experimental uncertainty, for a given value of  $W_{\text{SG}}$ , the ratio  $T_f/T_f^b$  ( $T_f^b$  is the bulk transition temperature for a given  $x$ ) for  $\text{Ag}_{1-x}\text{Mn}_x$  is independent of  $x$  and the same as was previously found for Cu-Mn. The data for thin ( $W_{\text{SG}} \leq 10$  nm) spin-glass layers are compatible with the predictions of the Fisher-Huse droplet-excitation model. Under some circumstances, large-angle x-ray scattering is shown to directly measure the layer thickness of the thinner of the two constituents of a multilayer sample.

### I. INTRODUCTION

Modern techniques for fabricating monatomic layers and multilayer systems make it possible to study finite-size effects and three-dimensional (3D) to 2D crossover behavior in magnetic systems. A fair amount is known about finite-size effects in systems with long-range order such as ferromagnets;<sup>1</sup> rather less is known about disordered systems such as spin glasses (SG's). We and others recently showed<sup>2-5</sup> that as the thickness  $W_{\text{SG}}$  of layers of the SG  $\text{Cu}_{1-x}\text{Mn}_x$  ( $0.04 \leq x \leq 0.14$ ) is reduced, the quasistatic spin-freezing temperature  $T_f$  [defined as the location of the peak in the zero-field-cooled (ZFC) linear magnetic susceptibility for a measuring time per data point of several minutes] decreases systematically, approaching 0 K as  $W_{\text{SG}}$  approaches 0 nm. In the present paper we show that a second spin glass, Ag-Mn, displays qualitatively and quantitatively similar behavior to Cu-Mn.

In this section, we limit ourselves to reviewing previous finite-size-effect studies of metallic SG's and the models with which we will compare our data. The reader interested in a more complete background and discussion of related issues, as well as a justification for finite-size studies of  $T_f$  in metallic and nonmetallic SG's, is referred to Ref. 4. Aside from a brief mention below of the nonlinear susceptibility, and of how the dependence of  $T_f$  on measuring time and the aging behavior in  $\text{Cu}_{1-x}\text{Mn}_x$  vary with  $W_{\text{SG}}$ , we focus upon changes in  $T_f$  with decreasing  $W_{\text{SG}}$ . It is important to emphasize that  $T_f$  measures only a dynamic property of the SG and that, to the extent that a SG can be said to undergo a true phase transition, it does so only in the limit  $\tau \rightarrow \infty$ . In Sec. II, we describe sample preparation and characterization. In Sec. III we present and analyze our Ag-Mn data and compare them with previous results for Cu-Mn. Section IV contains a summary and conclusions.

In the first published attempt to observe finite-size

effects in metallic SG's, Cowen, Kenning, and Slaughter<sup>6</sup> studied  $T_f$  as a function of  $W_{\text{Cu-Mn}}$  in sputtered multilayer samples (MS's) with  $W_{\text{SG}}=1.5$  nm of  $\text{Cu}_{0.93}\text{Mn}_{0.07}$  alternated with interlayer thicknesses of  $W_{\text{il}}=1.5$  to 9 nm of Cu or with  $W_{\text{SG}}=10$  to 0.5 nm of Cu-Mn alternated with  $W_{\text{il}}=1.2$  nm of Si. They did not find any clear evidence of finite-size effects, and we now know that their values of  $W_{\text{il}}$  were too small to magnetically decouple the SG layers.

The first definitive demonstration that  $T_f$  decreases systematically with decreasing  $W_{\text{Cu-Mn}}$  for magnetically decoupled SG layers was made by Kenning, Slaughter and Cowen<sup>2</sup> on sputtered MS's with  $W_{\text{Cu-Mn}}=2$  to 1000 nm alternated with 7 nm of Si. They reported that all of their data could be fit with the finite-size scaling equation<sup>7</sup>

$$(T_f^b - T_f)/T_f^b = (W_{\text{SG}}/W_c)^{-1/\nu}, \quad (1)$$

where  $T_f^b$  is the bulk transition temperature for a given  $x$ ,  $W_c$  is the SG width at which  $T_f=0$  K, and  $\nu$  is a constant. For  $\text{Cu}_{1-x}\text{Mn}_x/\text{Si}$  MS's with  $0.04 \leq x \leq 0.14$ , Kenning and co-workers found<sup>2,4</sup> that  $W_c \approx 3.5$  nm and  $\nu \approx 1.6 \pm 0.3$ ; this latter value is slightly larger than the value of  $\nu = 1.3 \pm 0.2$  found from nonlinear susceptibility measurements.<sup>8</sup> The ability of Eq. (1) to fit data over very wide ranges of  $W_{\text{SG}}$  is surprising, since Eq. (1) is expected to be valid only near  $T_f^b$ .<sup>7</sup>

Cowen, Kenning, and Bass<sup>3</sup> and Kenning *et al.*<sup>4</sup> subsequently found that the values of  $T_f$  for Cu-Mn/Cu MS's decreased somewhat less rapidly with decreasing  $W_{\text{SG}}$  than those for Cu-Mn/Si MS's. The Cu-Mn/Cu MS data were also compatible with Eq. (1), but with  $W_c \approx 1$  nm and  $\nu \approx 1.2 \pm 0.3$ . Kenning *et al.*<sup>4</sup> provided evidence that the larger value of  $W_c$  for Cu-Mn/Si was due, at least in part, to contamination of the Cu-Mn by Si. Stubi *et al.*<sup>9</sup> later showed that cooling the sample substrates to room temperature or below during sputtering reduced the

value of  $W_c$  derived for Cu-Mn/Si MS's toward that found for Cu-Mn/Cu MS's.

Recently, Gavrin *et al.*<sup>5</sup> reported that Cu-Mn/Al<sub>2</sub>O<sub>3</sub> MS with  $W_{SG}$  extending down to 6 nm showed decreases in  $T_f$  with decreasing  $W_{Cu-Mn}$  comparable to those found for Cu-Mn/Si, and Vranken and co-workers<sup>10,11</sup> reported similar decreases for Au-Fe single films with  $W_{SG}$  down to  $\approx 6$  nm. Fitting their Cu<sub>0.92</sub>Mn<sub>0.08</sub>/Al<sub>2</sub>O<sub>3</sub> MS data to Eq. (1) for  $W_{SG}$  up to 200 nm, Gavrin *et al.* derived a value of  $W_c \approx 2$  nm and  $\nu = 1.6 \pm 0.2$ . Also using Eq. (1), Vranken *et al.* initially reported obtaining  $W_c \approx 10$  nm for Au<sub>0.99</sub>Fe<sub>0.01</sub>,<sup>9</sup> but subsequently found that thin-film data for Au<sub>0.95</sub>Fe<sub>0.05</sub> deviated from Eq. (1) for  $W_{SG} \leq 10$  nm.<sup>10</sup>

Stimulated by the initial data of Kenning, Slaughter, and Cowen<sup>2</sup> Fisher and Huse<sup>12</sup> examined the 3D to 2D crossover behavior expected in Cu-Mn in terms of a droplet excitation model. For  $T_f$  near  $T_f^b$ , assuming that the lower critical dimension<sup>7</sup>  $d_l$  lies between 3 and 2, they derived an equation similar to Eq. (1), except multiplied by a factor logarithmic in the measuring time  $\tau$ ;

$$\frac{T_f^b - T_f(\tau)}{T_f^b} \propto W_{SG}^{-(1/\nu_3)} \left[ \ln \left( \frac{\tau}{W_{SG}^{\nu_3}} \right) \right]^{[\psi_3 + \nu_2 \psi_2 \theta_3 / \nu_3]^{-1}}. \quad (2)$$

Here subscripts denote dimensionality,  $z$  is the standard exponent for dynamic critical scaling,  $\psi$  and  $\theta$  are scaling exponents for activation barriers and free energies, respectively, and  $W_{SG}$  and  $\tau$  are measured in terms of a microscopic length and a microscopic time, respectively.<sup>12</sup> Our thick  $W_{SG}$  data are not accurate enough to distinguish between Eqs. (1) and (2).

For  $W_{SG}$  thin enough and  $\tau$  long enough so that  $T_f/T_f^b \ll 1$  (but with  $W_{SG}$  still larger than the average separation between impurities), Fisher and Huse derived the equation

$$T_f(\tau)/T_f^b \propto \left( \frac{W_{SG}^{\psi_3 + \psi_2 \nu_2 \theta_3}}{\ln \tau} \right)^{(1 + \nu_2 \psi_2)^{-1}}. \quad (3)$$

For fixed  $\tau$ , Eq. (3) can be simplified to

$$T_f/T_f^b \propto W_{SG}^a, \quad (4)$$

where the condition<sup>12</sup>  $d - 1 \geq \psi \geq \theta$  sets an upper bound of a  $\leq \psi_3 \leq 2$ . The values of the parameters in Eq. (3)— $\theta_3$ ,  $\psi_2 \nu_2$ , and especially  $\psi_3$ —are all highly uncertain. However, from simulations and experimental data,<sup>8,13,14</sup> we can estimate approximate bounds on these values,  $0.2 \leq \theta_3 \leq 0.3$ ,  $1 \leq \psi_2 \nu_2 \leq 2.2$ , and  $0.5 \leq \psi_3 \leq 1$ , which lead to an estimate of  $0.3 \leq a \leq 0.65$ . The only previous measurement of  $a$  from size effects is that by Kenning *et al.*,<sup>4</sup> who found  $a \approx 0.5 \pm 0.1$  for Cu-Mn/Cu MS's with  $2 \text{ nm} \leq W_{Cu-Mn} < 20 \text{ nm}$ . As their data extended only to  $T_f/T_f^b \approx 0.3$ , they did not reach  $T_f/T_f^b \ll 1$ .

Studies have also recently been made of effects of SG layer thickness on the high-field nonlinear susceptibility,<sup>5</sup> on how  $T_f$  varies with measuring time  $\tau$ ,<sup>14</sup> and on how the relaxation rate varies with  $W_{SG}$  for both magnetically isolated and partially recoupled SG layers.<sup>15</sup> These studies provide plausible evidence of 3D to 2D crossover be-

havior, with the second<sup>14</sup> explicitly suggesting that there is a finite value of  $W_{SG}$  below which  $T_f$  extrapolates to 0 K in the limit  $\tau \rightarrow \infty$ .

The foci of the present investigation were (a) to determine whether the behavior of  $T_f$  seen in Cu-Mn is also seen in another SG, and (b) to test whether this behavior is sensitive to the interlayer metal. Ag-Mn was chosen as the SG with properties closest to those of Cu-Mn.<sup>16,17</sup> Cu and Ag were chosen as alternative interlayers because (a) they are the host metals of Cu-Mn and Ag-Mn, (b) their electronic structures are almost identical, but (c) their lattice parameters differ enough ( $\approx 13\%$ ) to produce significantly different interface structures and grain sizes in Ag-Mn/Ag and Ag-Mn/Cu MS's.

Some data from this work have already been published elsewhere.<sup>9,18</sup> Complementary “recoupling” experiments on the return to 3D from quasi-2D in both Cu-Mn and Ag-Mn—based MS's when  $W_{SG}$  is held constant at 4 nm and  $W_{il}$  is reduced from 120 to 1 nm are being published separately.<sup>19</sup>

We previously showed for Cu-Mn/Cu MS's (Ref. 4) that we could rule out, as being of only minor importance, the potentially complicating effects of (a) reduction in the number of nearest Mn neighbors at the surfaces of finite-thickness layers, and (b) “spreading” of Mn out of the Cu-Mn layers and into the Cu during sputtering. The arguments given there apply equally well to the current work on Ag-Mn, and we do not consider these phenomena further. We also showed<sup>4</sup> that the finite-correlation-length spin-density-wave picture of Werner and co-workers<sup>20</sup> did not provide additional insight into our Cu-Mn/Cu data, and the same conclusion follows for the current data. Due to experimental uncertainties in the normalizing values of  $T_f^b$  for our Ag-Mn—based data, we limit our comparison of thick  $W_{SG}$  data with Eq. (1) to showing that the new data cluster around the fit previously obtained for Cu-Mn—based MS's.<sup>4</sup> For thin  $W_{SG}$  data, in contrast, we independently fit our new data with Eq. (4).

## II. SAMPLE PREPARATION AND CHARACTERIZATION

### A. Sample preparation and $T_f^b$

The MS samples were prepared by dc sputtering onto Si(001) substrates cooled to just below room temperature as described elsewhere.<sup>3,4,21</sup> To test whether the values of  $T_f$  are independent of the specific nature of the interlayer metal, both Ag-Mn/Ag and Ag-Mn/Cu MS's were made. Two nominally identical MS's were sputtered simultaneously onto substrates located symmetrically about the axes of the sputtering targets.

For Cu-Mn samples, we have always found the values of  $T_f$  measured on sputtered 500–1000-nm films to be essentially the same as those for shavings taken from the sputtering target, indicating that the Mn content of the films is the same as in the target. To ensure consistency, we took the  $T_f^b$  for a given Mn concentration to be the value for the shavings.<sup>4</sup>

For Ag-Mn, in contrast, the values of  $T_f$  for sputtered

400–1000-nm-thick films were systematically about 40% lower than those for shavings. The Mn content was found to be uniform throughout the Ag-Mn targets, and to be only little changed after substantial sputtering from the target. We thus concluded that the loss of Mn must be occurring during the sputtering process itself. Varying the sputtering rate from 0.6 to 1.8 nm/sec produced no obvious change in Mn content of the sputtered films, provided that the sputtering pressure was maintained constant. However, increasing the sputtering pressure, while holding the other parameters constant, substantially decreased both the sputtering rate and the Mn content, as shown in Fig. 1. The lighter Mn atoms are apparently scattered more strongly by the Ar atoms of the sputtering gas than are the heavy Ag atoms.

In view of this Mn loss during sputtering, we have chosen the values of  $T_f^b$  for the Ag-Mn alloys to be averages of those for 400–1000-nm-thick films. Due to a combination of the angular dependence of the amount of Mn arriving at the substrate noted above, with different locations of different substrates relative to the sputtering gun axis during sputtering, we observed variations around these chosen values. These variations lead to an uncertainty in  $T_f^b$  of about  $\pm 5\%$  for the  $\text{Ag}_{0.91}\text{Mn}_{0.09}/\text{Cu}$  MS's and slightly more for the  $\text{Ag}_{0.96}\text{Mn}_{0.04}/\text{Ag}$  and  $\text{Ag}_{0.96}\text{Mn}_{0.04}/\text{Cu}$  MS's. From the reproducibility and internal consistency of the data for our Ag-Mn MS's (see Table IV below), we are confident that the Mn loss was the same for all of our sputtered samples to within the percentage uncertainties listed for  $T_f^b$ . The specified Mn concentrations of 0.04 and 0.09 were estimated from the known curves of  $T_f^b$  vs  $x$  for Ag-Mn.<sup>16</sup>

### B. X rays and film-thickness determination

Ag-Mn and Ag have nearly the same electron density, and the lattice parameter of Ag does not change appreciably as Mn is added.<sup>22</sup> Multilayer satellites in Ag-

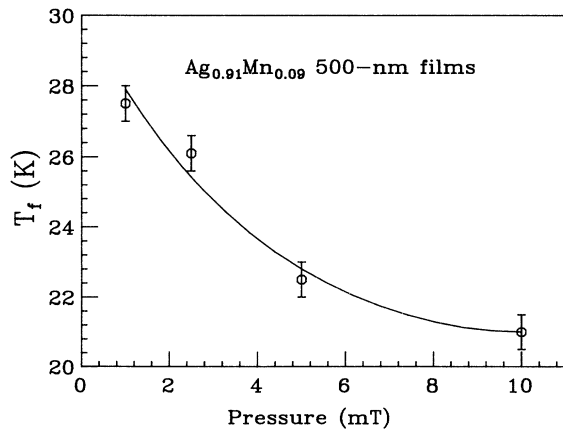


FIG. 1.  $T_f$  vs sputtering pressure  $P_s$  for 500-nm-thick Ag-Mn films prepared with target voltage and current held fixed. The decrease of  $T_f$  with increasing  $P_s$  indicates a decreasing fraction of Mn in the sputtered films. The curve is simply a guide to the eye.

Mn/Ag MS's are thus too weak to be seen. The widths of x-ray lines can, however, yield useful information about correlation lengths (which we take to be the average grain sizes) normal to the MS layers. We presume that grain sizes within the layers are comparable to those normal to the layers, but we have not measured the former.

For Ag-Mn/Cu MS's, in contrast, x rays permit the determination of both the bilayer thickness  $d$  and average grain sizes normal to the layers. We will show that, in some cases, x rays can also directly yield individual layer thicknesses.

#### 1. Average grain sizes normal to the layers

The first two columns of Tables I–III contain the intended values of  $W_{\text{SG}}$  and  $d$ , respectively, for  $\text{Ag}_{0.96}\text{Mn}_{0.04}/\text{Ag}$ ,  $\text{Ag}_{0.96}\text{Mn}_{0.04}/\text{Cu}$ , and  $\text{Ag}_{0.91}\text{Mn}_{0.09}/\text{Cu}$  MS's. The last columns contain the average grain sizes  $\xi$  for each of the samples, determined from the standard Scherrer formula as described previously.<sup>4</sup>

Since Ag-Mn and Ag have nearly the same lattice parameters,<sup>22</sup> we expect epitaxial growth at the interfaces, and indeed Table I shows that the grain sizes for Ag-Mn/Ag MS's are  $\approx 50$  nm, larger than the bilayer thicknesses  $d$  of all but the samples with the largest  $d$ , and nearly independent of  $W_{\text{SG}}$ . Similar behavior was also previously found in Cu-Mn/Cu MS's.<sup>4</sup>

The lattice parameters of Ag and Cu, in contrast, differ by about 13%.<sup>22</sup> Here, we expect grain sizes to be bounded by the individual layer thicknesses. The last two columns of Tables II and III show that this expectation is generally valid.

#### 2. Superlattice periodicity

For the Ag-Mn/Cu MS's, columns 3, 4, and 5 in Tables II and III contain the values of  $d$  determined, respectively, from the total sample thickness (measured

TABLE I. Intended SG layer thicknesses  $W_{\text{SG}}$ ; intended bilayer thicknesses  $d$ ; Dektak-measured bilayer thicknesses  $d_{\text{Dek}}$ ; and x-ray measured crystallite sizes  $\xi_T$ , for the  $\text{Ag}_{0.96}\text{Mn}_{0.04}/\text{Ag}$  MS's. The units are nm and the uncertainties are several percent.

$W_{\text{SG}}$	(Ag-4 at. % Mn)/Ag		$\xi_T$
	$d$	$d_{\text{Dek}}$	
100	130	118	50
50	80		48
25	55		48
		56	
12.5	42.5	40	57
		43	
7	37	35	52
5	35	34	64
		37	
3.5	33.5	33	54
2.0	32	31	52
		33.5	

TABLE II. Intended SG layer thicknesses  $W_{SG}$ ; intended bilayer thicknesses  $d$ ; Dektak, SAXD, and LAXD measured bilayer thicknesses  $d_{Dek}$ ,  $d_{SAXD}$ , and  $d_{LAXD}$  and SG layer thicknesses  $W_{SG}^M$ ; and x-ray measured crystallite sizes  $\xi_{Ag-Mn}$  and  $\xi_{Cu}$ , for the separate constituents of the  $Ag_{0.96}Mn_{0.04}/Cu$  MS's. The units are nm and the uncertainties are several percent.

$W_{SG}$	$d$	$Ag_{0.96}Mn_{0.04}/Cu$			$W_{SG}^M$	$\xi_{Ag-Mn}$	$\xi_{Cu}$
		$d_{Dek}$	$d_{SAXD}$	$d_{LAXD}$			
100	130					27	24
		142.5					
50	80	77.5				31	26
		52					
25	55	54			24.5	18	22
		40		44.5	10	10	19
12.5	42.5			40.5	12	13	31
		37.5					
7	37	38		34.5	6.7	7.7	37
		36					
5	35	32.5	44	32.5		4.6	19
		33					
3.5	33.5						
		33		31		2.6	42
2	32	30	38.5	30			32
		31					

with a Dektak IIA profilometer) divided by the number of layers deposited, from small-angle x-ray diffraction (SAXD) and from large-angle x-ray diffraction (LAXD). For the Ag-Mn/Ag MS's, column 3 of Table I contains similar Dektak-determined values. Because of the large value of  $W_{il}$  (i.e., 30 nm) needed to magnetically decouple the SG layers, the SAXD satellites occur at rather small angles, and the LAXD satellites appear superimposed upon the diffraction peaks for Ag-Mn and Cu as illustrated in Figs. 2 and 3. Both the small- and large-angle satellites were thus generally few and weak, leading to uncertainties in  $d_{SAXD}$  and  $d_{LAXD}$  of several percent.

In some circumstances, the satellite structure was dominated not by the oscillations due to the bilayer thickness

$d$ , but rather by the "diffraction patterns" from the thinner of the two layers of the MS (in our case,  $W_{SG}$ ). Figures 4 and 5 illustrate data that directly provide values for  $W_{SG}$  (which we designate by  $W_{SG}^M$ ). Although such "diffraction" induced structure has been seen in SAXD data,<sup>5</sup> and is formally contained in the appropriate x-ray equations, the fact that this structure directly yields  $W_{SG}$  does not seem to be widely appreciated.

Figure 4 compares data (solid curve) for a  $Ag_{0.96}Mn_{0.04}/Cu$  (7-nm/30-nm) MS with a calculation (dashed curve) for a single bilayer having a square-wave concentration profile (i.e.,  $N_{SG}$  layers of the SG with lattice parameter  $a_{SG}$ , and  $N_{IL}$  layers of the interlayer with lattice parameter  $a_{IL}$ ). In the solid curve, the bilayer os-

TABLE III. Intended SG layer thicknesses  $W_{SG}$ ; intended bilayer thicknesses  $d$ ; Dektak, SAXD, and LAXD measured bilayer thicknesses  $d_{Dek}$ ,  $d_{SAXD}$ , and  $d_{LAXD}$  and SG layer thicknesses  $W_{SG}^M$ ; and x-ray measured crystallite sizes  $\xi_{Ag-Mn}$  and  $\xi_{Cu}$ , for the separate constituents of the  $Ag_{0.91}Mn_{0.09}/Cu$  MS's. The units are nm and the uncertainties are several percent.

$W_{SG}$	$d$	$Ag_{0.91}Mn_{0.09}/Cu$			$W_{SG}^M$	$\xi_{Ag-Mn}$	$\xi_{Cu}$
		$d_{Dek}$	$d_{SAXD}$	$d_{LAXD}$			
500	530	571				49	26
		228				41	11
200	230					32	25
		134				30	26
50	80	80					
		54.9			26.2	22	27
25	55						
12.5	42.5	41.3		40.2	11.3	12	23
		34.5		38.7		6.9	25
7	37	36		33.2		5.1	24
		36					
5	35		42.2			4.0	
		36.2					
4	34	32.9		37.6		3.6	32
		32.9					
3.5	33.5						
		31.8	34.5	30.1			36
2	32	31.4		30.8			20
		31.4					
1	31						

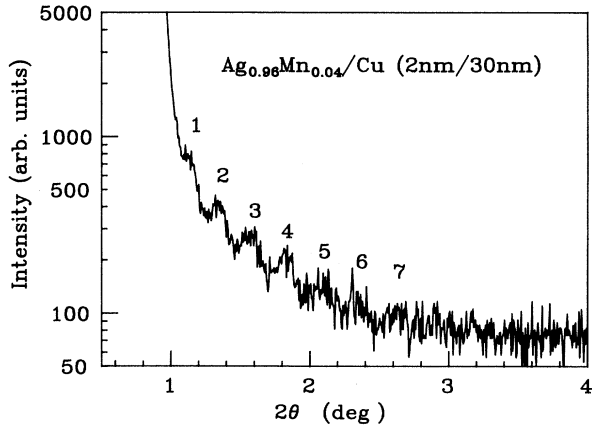


FIG. 2. The SAXD pattern for a  $\text{Ag}_{0.91}\text{Mn}_{0.09}/\text{Cu}$  (2-nm/30-nm) MS. The separation between the SAXD satellites determines  $d$ .

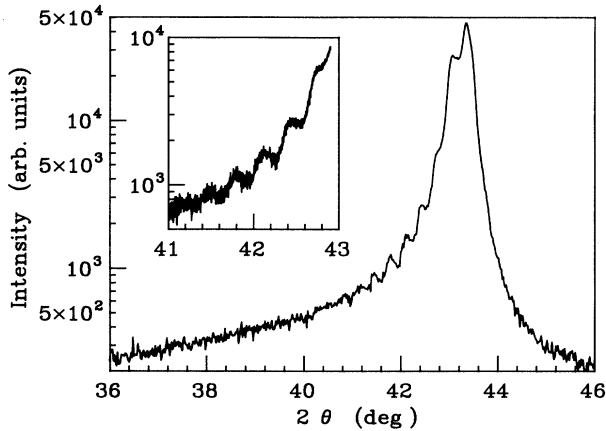


FIG. 3. The LAXD pattern for a  $\text{Ag}_{0.91}\text{Mn}_{0.09}/\text{Cu}$  (1-nm/30-nm) MS. The separation between the LAXD satellites determines  $d$ . The inset is an expanded view of the region from  $41^\circ$ – $43^\circ$  using the same units as the main figure.

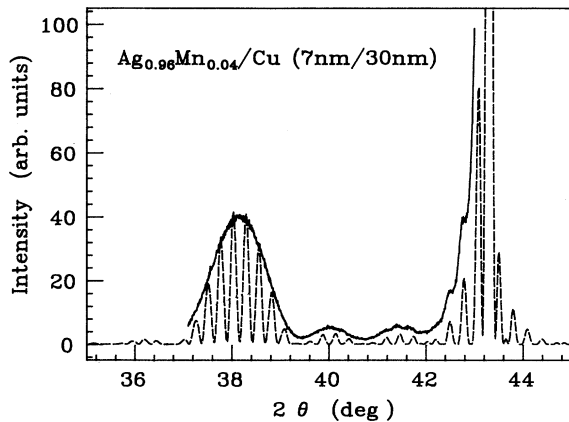


FIG. 4. The LAXD pattern for a  $\text{Ag}_{0.96}\text{Mn}_{0.04}/\text{Cu}$  (7-nm/30-nm) MS compared with the calculated pattern for a rectangular model with (7.1 nm)/(28.6 nm). The short coherence length nearly washed out the  $1/d$  oscillations [from Cowen *et al.* (Ref. 19)].

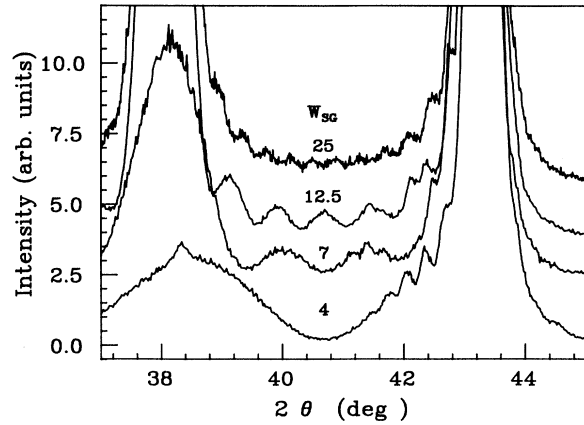


FIG. 5. LAXD patterns for a series of  $\text{Ag}_{0.96}\text{Mn}_{0.04}/\text{Cu}$  ( $W_{\text{SG}}/30\text{-nm}$ ) MS's, showing how the "diffraction" pattern varies as  $W_{\text{SG}}$  increases. The separation between the diffraction peaks determines  $W_{\text{SG}}^M$ .

cillations are nearly washed out, so that the region between the large Ag-Mn and Cu peaks is dominated by the two broad "diffraction" maxima due to  $W_{\text{SG}}$ . Figure 5 shows how the separation between these maxima grows as  $W_{\text{SG}}$  decreases.

### III. EXPERIMENTAL DATA AND ANALYSIS

#### A. $\chi(T)$

Measurements of  $\chi$  vs  $T$  were made on a Quantum Design MPMS superconducting quantum interference device (SQUID) magnetometer. This apparatus is capable of measuring sample magnetic moments over a wide range of temperatures and magnetic fields with a resolution of  $\approx 1 \times 10^{-8}$  emu.

To measure  $\chi$  for our layered samples, the sample plus substrate was cut into 0.5-cm  $\times$  1-cm strips to fit into the measuring coils. The magnetic field was set to zero and the sample was cooled from well above the expected  $T_f$  down to 5 K. The magnetic field was then raised to the measuring value of  $H = 100$  G, and  $\chi = M/H$  measurements were taken at a series of increasing temperatures, with measuring time  $\sim 5$  minutes per data point. The resulting data are called the zero-field-cooled (ZFC) data. With the field held at 100 G, measurements were then taken either at a series of decreasing temperatures back down to 5 K, or the sample was again cooled to 5 K and then warmed again. Such data are called field-cooled (FC) data; little or no differences were found between data taken with the two different FC procedures. Occasional checks showed that  $T_f$  was independent of  $H$  for  $H \leq 100$  G.

Figure 6 shows ZFC data for a series of  $\text{Ag}_{0.96}\text{Mn}_{0.04}/\text{Ag}$  MS's along with one example of FC data. These curves are representative of the susceptibility data of all of the Ag-Mn/Cu or Ag-Mn/Ag MS's, particularly in showing that the characteristic features of spin-

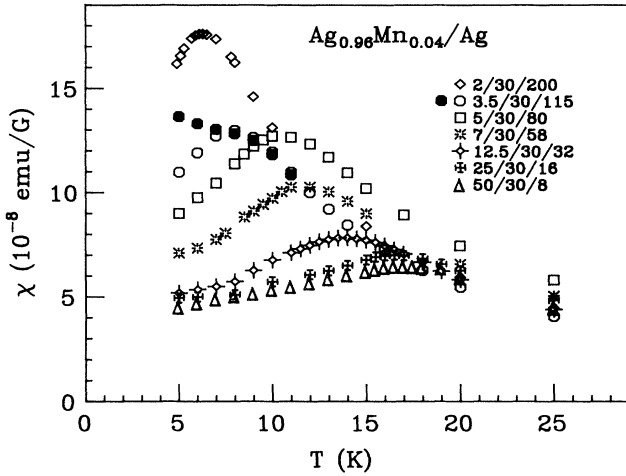


FIG. 6. ZFC  $\chi_{dc}$  versus  $T$  for a series of  $\text{Ag}_{0.96}\text{Mn}_{0.04}/\text{Ag}$  ( $W_{\text{SG}}/30\text{-nm}$ ) MS's, showing how the data continue to display the characteristic form of a SG down to the thinnest values of  $W_{\text{SG}}$  studied, and that  $T_f$  (the location of the maximum in each curve) decreases monotonically with decreasing  $W_{\text{SG}}$ . The filled circles show the FC curve for the 3.5-nm/30-nm sample. The numbers at the right of the graph represent  $W_{\text{SG}}$  (in nm),  $W_{\text{il}} = 30$  nm, and the total number of layers in the MS.

glasses persist right down to the thinnest layers we have studied.

Starting from  $T \approx 5$  K, the ZFC susceptibility increases with increasing temperature up to a maximum, the temperature of which is designated as  $T_f$ . Above  $T_f$ , the ZFC  $\chi$  exhibits Curie-like behavior.

The FC  $\chi$  is also Curie-like above  $T_f$ , but typically slightly larger than the ZFC  $\chi$  at the same temperature. In bulk spin-glass materials, the FC  $\chi$  typically first de-

creases slightly as  $T$  is reduced below  $T_f$ , and thereafter remains nearly constant.<sup>17</sup> In our Ag-Mn-based MS's, and FC  $\chi$  also typically decreases slightly as  $T$  is reduced below  $T_f$ , but then increases as  $T \rightarrow 0$  K.<sup>4</sup> Similar behavior was previously seen in Cu-Mn/Cu MS's.<sup>4</sup>

For the Ag-Mn/Ag MS's, the susceptibilities for  $T > T_f$  all appear to follow generally similar Curie-like behavior, with magnitudes corresponding to approximately the same number of paramagnetic moments. This is what we would expect, since we have attempted to keep the total amount of SG material about the same in all of the samples. Part of the residual differences in  $\chi$  can be attributed to differences in the (nearly temperature independent) susceptibilities of the diamagnetic Si substrates backing the samples, for which no correction has been made. But it is not clear that this is the complete explanation, since remeasurements of Cu-Mn-based MS's<sup>23</sup> after several months have shown that, while  $T_f$  for a given sample remains constant, the magnitudes of both the ZFC and FC  $\chi$ 's, as well as the separations between them, can change.

From Fig. 6, we see that  $T_f$  decreases systematically with decreasing  $W_{\text{SG}}$ . By fitting a smooth curve to the data in the vicinity of each peak, the uncertainty in  $T_f$  could be reduced to  $\pm 0.5$  K.

## B. Magnetic decoupling of SG layers

To determine what interlayer thicknesses  $W_{\text{il}}$  of Cu or Ag were large enough to magnetically decouple the SG layers from each other, Cu-Mn- and Ag-Mn-based MS's with fixed  $W_{\text{SG}} = 4$  nm were prepared with  $W_{\text{il}}$  up to 120 or 60 nm, respectively. For both types of MS,  $T_f$  was found to become independent of  $W_{\text{il}}$ —to within experimental uncertainty—for  $W_{\text{il}} \geq 30$  nm.<sup>4,18,19</sup> Figure 7 illustrates this behavior for the  $\text{Ag}_{0.91}\text{Mn}_{0.09}/\text{Cu}$  MS.

TABLE IV. Values of  $T_f$  for the  $\text{Ag}_{0.96}\text{Mn}_{0.04}/\text{Ag}$ ,  $\text{Ag}_{0.96}\text{Mn}_{0.04}/\text{Cu}$ , and  $\text{Ag}_{0.91}\text{Mn}_{0.09}/\text{Cu}$  MS's with  $500 \geq W_{\text{SG}} \geq 1$  nm, along with the values obtained for target shavings. The uncertainty in the determination of  $T_f$  for a given sample or for the shavings is  $\pm 0.5$  K. The values listed for  $W_{\text{SG}} = 500$  nm are averages over samples made on different runs, and their uncertainties indicate the variations observed.

$W_{\text{SG}}$	$\text{Ag}_{0.96}\text{Mn}_{0.04}/\text{Cu}$		$\text{Ag}_{0.96}\text{Mn}_{0.04}/\text{Ag}$		$\text{Ag}_{0.91}\text{Mn}_{0.09}/\text{Cu}$	
	(a)	(b)	(a)	(b)	(a)	(b)
Shavings			27.5		45.0	
400–1000 films			16.5 $\pm$ 1		26.0 $\pm$ 1	
500					22.7	
200					24.1	
100	17.3	17.7	19.3	18.5	25.7	
50	17.5	16.9	16.3	17.2	22.8	
25	15.9	16.7	16.7	16.6	22.8	
12.5	14.2	14.5	13.5	13.9	17.2	19.0
7	12.2	12.1	11.5	11.8	15.5	17.1
5	10.0	11.1	10.6	10.5	12.6	14.4
4						13.1
3.5	8.35	8.1	7.8	7.9	10.7	12.3
2	5.35	5.5	6.0	6.3	6.8	7.8
1					4.5	5.0

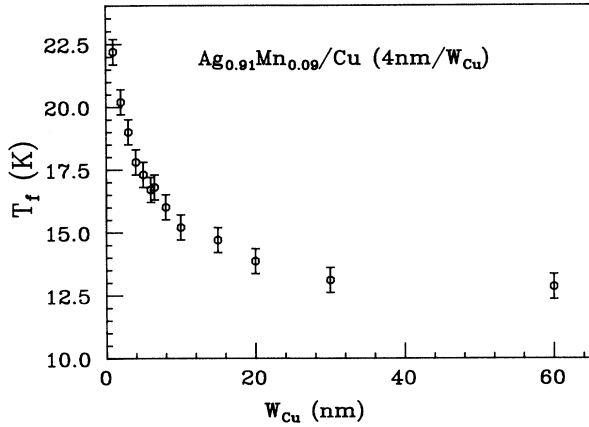


FIG. 7.  $T_f$  versus  $W_{il}$  for a series of  $\text{Ag}_{0.91}\text{Mn}_{0.09}/\text{Cu}$  ( $4\text{-nm}/W_{il}$ ) MS's, showing that  $T_f$  becomes independent of  $W_{il}$  beyond  $W_{il} \approx 30$  nm. The error bars represent uncertainties of  $\pm 0.5$  K.

### C. $T_f$ and $T_f/T_f^b$ for magnetically isolated SG layers

Table IV contains the values of  $T_f$  for all of our Ag-Mn MS's, along with values measured from shavings from each of the two targets. As noted above, the values of  $T_f$  for 400–1000-nm-thick films are chosen as  $T_f^b$ .

For simplicity, we plot only the averaged values of  $T_f$  for MS's prepared together. Figure 8 shows these averaged values in the form  $\log T_f/T_f^b$  vs  $\log W_{\text{Cu-Mn}}$  for the  $\text{Ag}_{0.96}\text{Mn}_{0.04}/\text{Ag}$  (inverted triangles),  $\text{Ag}_{0.96}\text{Mn}_{0.04}/\text{Cu}$  (erect triangles), and  $\text{Ag}_{0.91}\text{Mn}_{0.09}$  (squares) MS's, along with a few values for  $\text{Ag}_{0.96}\text{Mn}_{0.04}$  single films (open crosses) and data for a  $\text{Cu}_{0.93}\text{Mn}_{0.07}/\text{Cu}$  MS's (filled circles) from Ref. 4. The logarithmic scale for  $W_{\text{Cu-Mn}}$  permits display on a single graph of data for  $W_{\text{Cu-Mn}}$  ranging from 1000-nm down to 1 nm. The logarithmic scale for  $T_f/T_f^b$  permits direct visual determination of whether the data for thin samples are compatible with Eq. (4). The data for all four sample sets are the same to within experimental uncertainties, which are usually dominated by the uncertainties in  $T_f^b$ .

The dashed curve in Fig. 8 corresponds to the fit to Eq. (1) previously obtained with Cu-Mn-based MS's.<sup>4</sup> The substantial uncertainties in  $T_f^b$  for the present samples preclude an independent fit to this equation, but we see that the Ag-Mn-based data cluster around the previous fit. The solid line in Fig. 8 corresponds to Eq. (4). The slope of this line,  $a = 0.65 \pm 0.1$ , is slightly larger than the value  $a = 0.5 \pm 0.1$  previously reported for Cu-Mn/Cu MS's<sup>4</sup> but still compatible with the rough estimates of  $a$  given in Sec. I. We noted in Sec. I that the Cu-Mn/Cu MS data extended down to only  $T_f/T_f^b \approx 0.3$ , and thus that only the thinnest SG layers were beginning to satisfy the validity condition of Eq. (4):  $T_f/T_f^b \ll 1$ . The Ag-Mn data of Fig. 8 extend down to  $T_f/T_f^b \approx 0.2$ , further into the expected region of validity of Eq. (4), and this extension accounts for their larger value of  $a$ .

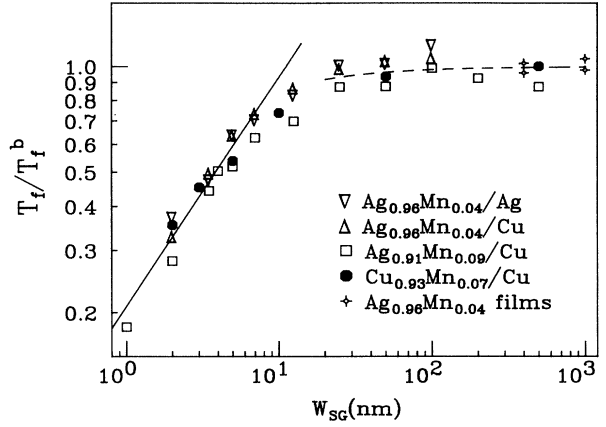


FIG. 8.  $\log T_f/T_f^b$  versus  $\log W_{\text{SG}}$  for a variety of  $W_{\text{SG}}/30\text{-nm}$  MS's and a few thick  $\text{Ag}_{0.96}\text{Mn}_{0.04}$  films, showing that the data for the Ag-Mn-based MS's fall closely around those for the Cu-Mn-based MS's. The plotted values of  $T_f$  for the Ag-Mn MS's are averages of those listed in Table IV. The dashed line indicates the behavior predicted by Eq. (1) with  $\nu = 1.2$ . The solid line indicates behavior compatible with Eq. (4) for  $a = 0.65$ .

## IV. SUMMARY AND CONCLUSIONS

(1) From comparison of values of  $T_f$  for 400–1000-nm-thick sputtered film's with those for shavings from the Ag-Mn targets, we infer that the Mn concentration in the Ag-Mn-based MS's is only about 60% of that in target. We attribute this smaller Mn concentration in the MS's primarily to greater scattering of the lighter Mn atoms by the sputtering gas. The reproducibility of our data for very different samples indicates that this Mn concentration must be nearly the same for all of our MS.

(2) Typical SG characteristics (i.e., a peak in ZFC  $\chi$  and irreversibility below  $T_f$ ) persist to SG layer thicknesses down to at least  $W_{\text{SG}} = 1$  nm.

(3) To within experimental uncertainties,  $T_f/T_f^b$  is the same for a given value of  $W_{\text{SG}}$  for both Ag-Mn/Ag and Ag-Mn/Cu MS's over the range of  $W_{\text{SG}}$  studied. The SG properties of these MS are thus not significantly affected by the different grain sizes and interface structures arising from the different lattice parameters of Ag and Cu.

(4) In agreement with the behavior previously found for  $\text{Cu}_{1-x}\text{Mn}_x/\text{Cu}$  MS's, to within experimental uncertainties,  $T_f/T_f^b$  is independent of Mn concentration for  $\text{Ag}_{0.96}\text{Mn}_{0.04}$  and  $\text{Ag}_{0.91}\text{Mn}_{0.09}$  MS's.

(5)  $T_f/T_f^b$  is also the same to within experimental uncertainties for Cu-Mn and Ag-Mn-based MS's. Finite size effects are thus very much the same in these two similar SG's.

(6) The data for both thick and thin SG layers are compatible with the Fisher-Huse model, with the thin sample data yielding an exponent in Eq. (4) of  $a = 0.65 \pm 0.1$ . This value is at the upper end of the range derived from independent estimates of the coefficients in Eq. (3), but well below the rigorous upper bound of 2. However, not

all of the data used to determine this value lie within the range of expected validity of Eq. (4).

(7) We show that, under certain circumstances, LAXD measurements on MS can directly yield the thickness of the thinner layer in an MS.

We conclude that our data for magnetically decoupled Ag-Mn-based MS's behave very similarly to those previously reported for Cu-Mn-based MS's.

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<sup>1</sup>A. Fert (unpublished)

<sup>2</sup>G. G. Kenning, J. M. Slaughter, and J. A. Cowen, *Phys. Rev. Lett.* **59**, 2596 (1987).

<sup>3</sup>J. A. Cowen, G. G. Kenning, and J. Bass, *J. Appl. Phys.* **64**, 5781 (1988).

<sup>4</sup>G. G. Kenning, J. Bass, W. P. Pratt, Jr., D. Leslie-Pelecky, L. Hoines, W. Leach, M. L. Wilson, R. Stubi, and J. A. Cowen, *Phys. Rev. B* **42**, 2393 (1990).

<sup>5</sup>A. Gavrin, J. R. Childress, C. L. Chien, B. Martinez, and M. B. Salamon, *Phys. Rev. Lett.* **64**, 2438 (1990).

<sup>6</sup>J. A. Cowen, G. G. Kenning, and J. M. Slaughter, *J. Appl. Phys.* **61**, 4080 (1987).

<sup>7</sup>M. N. Barber, in *Phase Transitions and Critical Phenomena*, edited by C. Domb and J. L. Lebowitz (Academic, New York, 1983), Vol. 8.

<sup>8</sup>L. P. Lévy and A. T. Ogielski, *Phys. Rev. Lett.* **57**, 3288 (1986).

<sup>9</sup>R. Stubi, J. A. Cowen, D. Leslie-Pelecky, and J. Bass, *Physica B* **165&166**, 459 (1990).

<sup>10</sup>J. Vranken, C. Van Haesendonck, H. Vloeberghs, and Y. Bruynseraede, *Phys. Scr.* **T25**, 348 (1989).

<sup>11</sup>H. Vloeberghs, J. Vranken, C. Van Haesendonck, and Y. Bruynseraede, *Europhys. Lett.* **12**, 557 (1990).

<sup>12</sup>D. S. Fisher and D. A. Huse, *Phys. Rev. B* **36**, 8937 (1987).

<sup>13</sup>A. J. Bray and M. A. Moore, *J. Phys. C* **17**, L463 (1984); W. L. McMillan, *Phys. Rev. B* **30**, 476 (1984); W. Kinzel and K. Binder, *Phys. Rev. B* **29**, 1300 (1984); A. P. Young, *Phys.*

*Rev. Lett.* **50**, 917 (1983); C. Dekker, A. F. M. Arts, H. W. de Wijn, A. J. van Duyneveldt, and J. A. Mydosh, *ibid.* **61**, 1780 (1988); J. Mattson, P. Granberg, P. Nordblad, and L. Lundgren (unpublished).

<sup>14</sup>L. Sandlund, P. Granberg, L. Lundgren, P. Nordblad, P. Svedlindh, J. A. Cowen, and G. G. Kenning, *Phys. Rev. B* **40**, 869 (1989). P. Granberg, P. Nordblad, P. Svedlindh, L. Lundgren, R. Stubi, G. G. Kenning, D. L. Leslie-Pelecky, J. Bass, and J. A. Cowen, *J. Appl. Phys.* **67**, 5252 (1990).

<sup>15</sup>J. Mattsson, P. Granberg, P. Nordblad, L. Lundgren, R. Stubi, D. Leslie-Pelecky, J. Bass, and J. Cowen, *Physica B* **165&166**, 461 (1990).

<sup>16</sup>K. H. Fischer, *Landolt-Börnstein Tables*, New Series, Gruppe III, Vol. 15a, edited by K. H. Hellwege (Springer-Verlag, Berlin, 1982), p. 289.

<sup>17</sup>K. Binder and A. P. Young, *Rev. Mod. Phys.* **58**, 801 (1986).

<sup>18</sup>L. Hoines, R. Stubi, R. Loloee, J. A. Cowen, and J. Bass, *Phys. Rev. Lett.* **66**, 1224 (1991).

<sup>19</sup>R. Stubi, D. L. Leslie-Pelecky, and J. A. Cowen, *J. Appl. Phys.* **67**, 5970 (1990); R. Stubi, J. Bass, and J. A. Cowen, *J. Appl. Phys.* **69**, 5054 (1991); J. A. Cowen, J. Bass, P. Granberg, L. Lundgren, and R. Stubi, *Physica B* **69**, 299 (1991)

<sup>20</sup>S. Werner, *Commun. Solid State Phys.* **15**, 55 (1990).

<sup>21</sup>C. Fierz, R. Stubi, and W. P. Pratt, Jr. (unpublished).

<sup>22</sup>W. B. Pearson, *A Handbook of Lattice Spacings and Structures of Metals and Alloys* (Pergamon, New York, 1958).

<sup>23</sup>D. Leslie-Pelecky (unpublished).