

## Indirect magnetic interaction through silver in epitaxial Fe(110)/Ag(111) multilayers

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Evidence for an indirect magnetic interaction through silver has been observed in three different series of high-quality monocrystalline Fe(110)/Ag(111) multilayer films fabricated by molecular-beam epitaxy. These multilayers consisted of 3-, 5-, or 8-monolayer (ML) bilayer components of Fe; each respective series had Ag bilayer components ranging from 4 to 20 ML's. An analysis of the temperature dependence of magnetization (as determined by  $^{57}\text{Fe}$  Mössbauer spectroscopy) indicated that there was a magnetic coupling between neighboring Fe bilayer components through the intervening Ag. This interlayer coupling could be seen through variations in the  $T^{3/2}$  dependence of the hyperfine field in the 5- and 8-ML Fe component multilayers. However, the 3-ML Fe component series exhibited a linear  $T$  to  $T^{3/2}$  dimensional crossover in the hyperfine-field temperature dependence. Specifically, the linear temperature dependence of the 3-ML film with a thick Ag component (the magnetically uncoupled case) represented the behavior of a genuine two-dimensional ferromagnet. However, as the interlayer magnetic coupling increased with decreasing Ag thickness, a temperature dependence (at low temperatures) developed that increasingly resembled that of three-dimensional ferromagnetism, i.e., a  $T^{3/2}$  law.

### I. INTRODUCTION

In recent years the use of molecular-beam epitaxy (MBE) systems for film growth has made it possible to produce high-quality, ultrathin metallic multilayer films.<sup>1,2</sup> This has enabled researchers to search for the presence of magnetic coupling between bilayer components in various multilayer systems.<sup>3-6</sup> As will be shown in this paper, magnetic coupling in a multilayer can also be observed with  $^{57}\text{Fe}$  Mössbauer spectroscopy in the absence of an applied magnetic field. Variations in the temperature dependence of the magnetization in Fe(110)/Ag(111) multilayers are observed to be dependent on the thickness of the Ag bilayer component separating the individual Fe bilayer components of the multilayer and are a direct consequence of the varying strength of the ferromagnetic coupling between the adjacent Fe bilayers through the intervening Ag barriers.

The multilayers studied were of the form  $(\text{Ag}_x\text{Fe}_y)_z$ , where  $x$  refers to the Ag bilayer component thickness in monolayers [1 ML Ag(111)=2.36 Å],  $y$  refers to the Fe bilayer thickness in monolayers [1 ML Fe(110)=2.03 Å], and  $z$  refers to the number of bilayer repetitions. Three series of films were grown and were composed of Fe(110) component thicknesses ( $y$ ) equal to 3, 5, or 8 ML. Within each respective Fe component multilayer series, films with different Ag(111) bilayer component thicknesses ( $x$ ) were made. For the 5- and 8-ML Fe(110) component series, the Ag component thickness ( $x$ ) was either 5, 12, or 20 ML. For the 3-ML Fe component series,  $x=4, 5, 6, 7, 8, 10, 12, 14, 17, \text{ or } 20$  ML. The 5- and 8-ML Fe series were composed of  $z=30$  repetitions, while the 3-ML Fe series was composed of  $z=8$  repetitions.

### II. EXPERIMENT

The multilayers were produced with a PHI model 430B MBE system equipped with *in situ* reflection high-energy electron diffraction (RHEED), a residual gas analyzer, and a quartz-crystal oscillator for film thickness monitoring. In a previous paper, we reported the detailed growth procedures for Fe(110)/Ag(111) multilayers grown by MBE.<sup>7</sup> In this previous paper, it was shown with RHEED that the flatness and epitaxy of Fe(110) films grown on Ag(111) is optimal at a substrate temperature ( $T_s$ ) equal to 180°C. It is seen that high-quality Fe(110) and Ag(111) RHEED patterns persist with little degradation even after the growth of 50 repetitions of each component. In addition, kinks or breaks in Auger electron intensities at approximately monolayer step coverages observed during the growth of both Fe(110) on Ag(111) and Ag(111) on Fe(110) at  $T_s=180^\circ\text{C}$  are also indicative of flat and continuous bilayer component growth under these MBE growth conditions.

The 5- and 8-ML Fe(110) component multilayers were grown on a thick epitaxial Ag(111) base layer (typically 2.5 kÅ) deposited from an effusion cell onto a synthetic Fe-free mica substrate. Because of the unavailability of additional synthetic Fe-free mica substrates, the 3-ML Fe(110) component multilayers were grown on high-quality natural mica substrates. In order to eliminate the interference from the substantial Fe contamination in natural mica during transmission Mössbauer measurements, a method was developed which allowed for the trivial and complete removal of the multilayers from the natural mica substrates.

Before growing the 3-ML Fe(110) multilayers,  $\approx 300$  Å of NaCl was effused from a Knudsen cell onto the natural

mica substrate (quadrupole mass analysis during growth indicated that NaCl effused as molecules), immediately followed by the growth of an  $\approx 5\text{-k}\text{\AA}$  Ag(111) base layer. The monocrystallinity of the Ag(111) base layer grown on the NaCl deposit was verified with RHEED. Furthermore, the RHEED features of these completed Ag(111) base films was indistinguishable from those of similar Ag(111) base layers grown directly on synthetic mica. The 3-ML Fe(110) multilayers were grown on this NaCl-buffered Ag(111) base. After growth the finished film was easily removed from the natural mica substrate with adhesive tape and a few drops of distilled uhp water. As a further check of this growth procedure, it was seen that the Mössbauer characteristics of a test multilayer with 5-ML Fe(110) and 20-ML Ag(111) components grown on Ag(111)/NaCl/natural mica gave the same parameters as a similar Fe/Ag multilayer grown on a Ag(111)/synthetic mica substrate, within experimental uncertainty.

For all of the multilayers, growth sources consisted of enriched Fe (35%  $^{57}\text{Fe}$ , 65%  $^{56}\text{Fe}$ ) effused from a Knudsen cell using a high-purity alumina crucible and Ag effused from Knudsen cell using a PBN crucible. The base pressure was typically better than  $5 \times 10^{-10}$  torr, while the pressure during deposition was always less than  $2 \times 10^{-9}$  torr.

Transmission  $^{57}\text{Fe}$  Mössbauer spectroscopy (TMS) was used to analyze all three series of Fe/Ag multilayers at temperatures from 4.2 to 300 K. Unlike surface-sensitive

conversion electron Mössbauer spectroscopy (CEMS), TMS samples all of the multilayer's Fe bilayer components with equal sensitivity. The results of these measurements will be described in the following sections.

### III. 5- AND 8-ML Fe(110) COMPONENT MULTILAYERS

Figure 1 shows the TMS spectra of all the 5- and 8-ML Fe(110) component multilayers measured at room temperature, while Fig. 2 shows the same measurements at 4.2 K. There are several characteristics which can be immediately noted in all of these spectra. First, the sextet nature of the spectra indicates that all of the multilayers are magnetically ordered (with in-plane magnetization) at all temperatures. Second, there is no evidence of a magnetically relaxed singlet spectral component, even at room temperature.

It is apparent from Figs. 1 and 2 that the sextet spectra have sharp linewidths, indicating a narrow distribution of different  $^{57}\text{Fe}$  hyperfine-field sites within each of these films. In fact, there is virtually no difference in hyperfine-field values or spectral linewidths between the  $(\text{Ag}_x\text{Fe}_y)_{30}$  multilayers and the Fe(110)/Ag(111) heterostructures consisting of fewer bilayer repetitions previously reported in Ref. 7, further indicating that each of the 30 Fe bilayer components is virtually identical in structure and flatness. Significant bilayer roughness or irregularity between the Fe bilayers would result in a wider

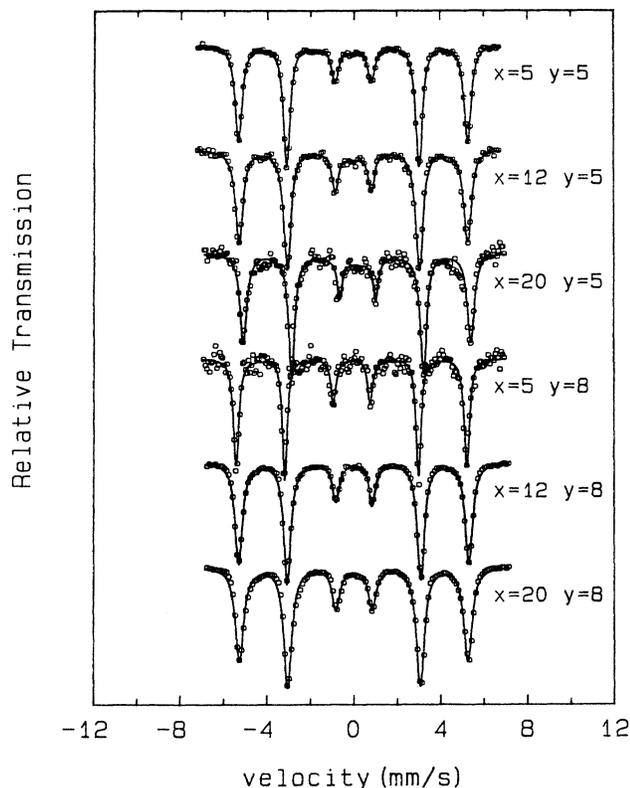


FIG. 1. Mössbauer spectra of the  $(\text{Ag}_x\text{Fe}_5)_{30}$  and  $(\text{Ag}_x\text{Fe}_8)_{30}$  multilayers measured at room temperature.

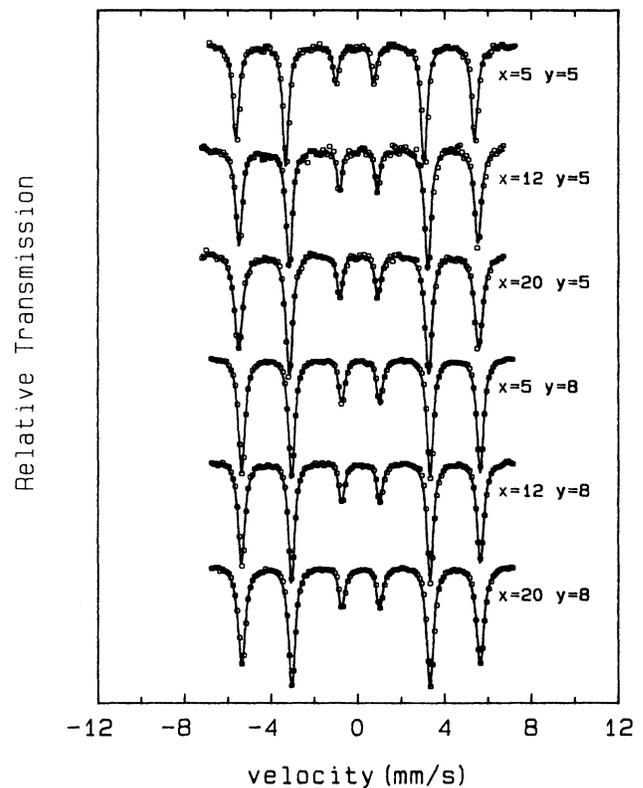


FIG. 2. Mössbauer spectra of the  $(\text{Ag}_x\text{Fe}_5)_{30}$  and  $(\text{Ag}_x\text{Fe}_8)_{30}$  multilayers measured at 4.2 K.

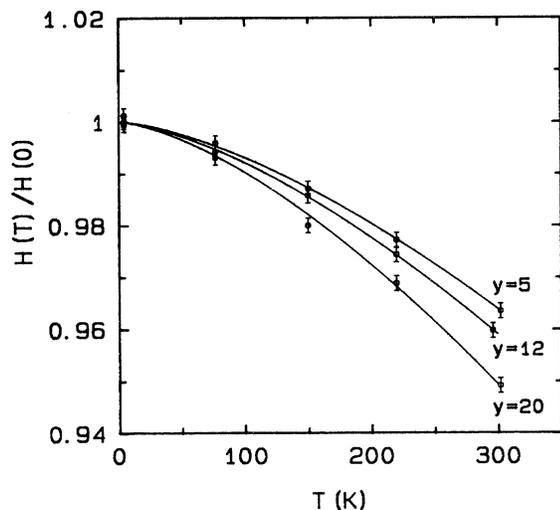


FIG. 3. Normalized hyperfine field  $[H_0/H(T)]$  vs temperature ( $T$ ) plots for the  $(\text{Ag}_x\text{Fe}_8)_{30}$  multilayers.

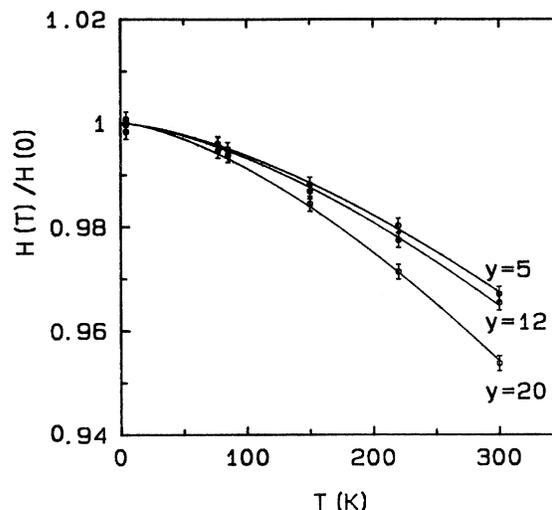


FIG. 4. Normalized hyperfine field  $[H_0/H(T)]$  vs temperature ( $T$ ) plots for the  $(\text{Ag}_x\text{Fe}_5)_{30}$  multilayers.

distribution of different hyperfine-field sites, resulting in greatly broadened spectral linewidths.

Figure 3 shows a plot of  $H$ , the magnetic hyperfine field, versus temperature for the three 8-ML Fe bilayer component multilayers with varying Ag bilayer component thicknesses, while Fig. 4 shows a similar plot for the three 5-ML Fe bilayer component multilayers. All of the curves can be fit to the equation

$$H(T) = H_0(1 - BT^{3/2}), \quad (1)$$

which describes the thermal spin-wave excitations which are present in each of the multilayers. It is worth noting that other groups have also reported  $T^{3/2}$ -like dependences of the hyperfine field in Fe(110)/Ag(111) films at similar Fe component thicknesses.<sup>8,9</sup>

It has been well established that the temperature dependence of the hyperfine field is closely proportional to the local temperature dependence of the magnetization in a film.<sup>8</sup> Therefore, the magnetization also decreases with a  $T^{3/2}$  dependence, as predicted by spin-wave theory.<sup>10,11</sup>

Table I lists the values of  $H_0$  and  $B$  for all the films, where  $H_0$  is the ground-state ( $T=0$  K) hyperfine-field value. Both series of films have values of  $H_0$  enhanced over the bulk value of 340 kG. Since previous investigations have determined that the surface of an Fe film in contact with Ag has a ground-state hyperfine field enhanced by approximately 3% over the bulk value, to approximately 348 kG,<sup>7,12</sup> and since there is a greater relative proportion of an ideal 5-ML Fe bilayer at the hyperfine-field-enhanced Fe/Ag interface sites compared to an ideal 8-ML Fe bilayer, the 5-ML Fe bilayer multilayers are expected to have values of  $H_0$  greater than those of their respective 8-ML Fe bilayer multilayer counterparts (i.e., those with the same Ag spacer layers). This expected behavior is reflected in Table I and is also reflected by the 5-ML slightly greater TMS linewidths as

compared to the 8-ML films with equivalent Ag bilayer thicknesses.

The effect of the interface on the magnetic properties of the multilayers is more readily apparent from an examination of the  $T^{3/2}$  relation prefactor ( $B$  value) instead of  $H_0$ . The  $B$  value of bulk Fe, which is a measure of how easy it is to form spin-wave excitations in bulk Fe, is approximately  $5.2 \times 10^{-6} \text{ K}^{-3/2}$  according to a measurement of our own bulklike calibration foil. It has been shown both theoretically and experimentally that spin waves at the interface are softer than in the bulk because of the decreased number of exchange interactions. As a consequence, the  $B$  value at the interface is approximately twice the bulk value.<sup>13-15</sup> Since the  $B$  value may change by a factor of 2 from the bulk to the interface, while  $H_0$  changes by only about 3%, surface magnetic interactions can be more easily and accurately studied by examining the  $B$  value.

For both series of multilayers, the  $B$  value increases as the thickness of the Ag bilayers (separating adjacent Fe bilayer components) increases and in all cases is greater than the bulk value. For the 8-ML Fe multilayers, the  $B$  value is equal to  $6.2 \times 10^{-6} \text{ K}^{-3/2}$  for  $x=5$  ML and in-

TABLE I. Values of the extrapolated ground-state hyperfine field ( $H_0$ ) and  $B$  values for the 5- and 8-ML Fe bilayer component multilayers  $(\text{Ag}_x\text{Fe}_5)_{30}$  and  $(\text{Ag}_x\text{Fe}_8)_{30}$ .

Films	$x$ (ML)	$H_0$ (kG)	$B$ ( $\text{K}^{-3/2}$ )
$(\text{Ag}_x\text{Fe}_8)_{30}$	5	342.5	$6.25 \times 10^{-6}$
	12	342.6	$6.75 \times 10^{-6}$
	20	342.9	$8.81 \times 10^{-6}$
$(\text{Ag}_x\text{Fe}_5)_{30}$	5	342.9	$6.96 \times 10^{-6}$
	12	343.2	$7.88 \times 10^{-6}$
	20	343.4	$9.75 \times 10^{-6}$

creases to  $8.81 \times 10^{-6} \text{ K}^{-3/2}$  for  $x = 20$  ML. This variation in  $B$  value implies that the effects of the surface on the magnetic excitations of the film are more prominent as the Ag bilayer thickness increases. A similar trend is seen in the 5-ML Fe multilayer films. The  $(\text{Ag}_x\text{Fe}_5)_{30}$  multilayer with the thinnest Ag bilayer component ( $x = 5$  ML) has a  $B$  value equal to  $6.9 \times 10^{-6} \text{ K}^{-3/2}$ , while the film with the thickest Ag bilayer component ( $x = 20$  ML) has a  $B$  value equal to  $9.75 \times 10^{-6} \text{ K}^{-3/2}$ .

The larger and more interfacial-like  $B$  values which are observed as the Ag bilayer thickness increases indicates the presence of a magnetic interaction between neighboring Fe bilayer components through the intervening Ag. When the Ag bilayer is thin, the interaction between Fe atoms located at neighboring Fe/Ag bilayer interfaces is stronger, resulting in more bulklike magnetic behavior. As the Ag bilayer becomes thicker, the neighboring Fe interfacial atoms become more isolated, exhibiting magnetic properties more typical of an isolated Fe/Ag interface. Note how the data show that the  $B$  value varies not only with Ag bilayer thickness within a multilayer series, but also with Fe thickness between the two different series. In other words, for any given bilayer thickness ( $x$ ) of Ag,  $(\text{Ag}_x\text{Fe}_5)_{30}$  has a greater  $B$  value than  $(\text{Ag}_x\text{Fe}_8)_{30}$ . Again, this is expected because of the greater relative interfacial population of Fe sites in  $(\text{Ag}_x\text{Fe}_5)_{30}$  compared to  $(\text{Ag}_x\text{Fe}_8)_{30}$ .

According to surface energy arguments,<sup>16</sup> Ag is expected to easily wet Fe and form smooth, continuous bilayers. Moreover, a Ag adatom is typically more mobile than an Fe adatom given the same substrate temperature. Given our slow growth rates for Ag (20 Å/min) on a heated substrate ( $T_s = 180^\circ\text{C}$ ), it is no surprise that RHEED analysis shows that Ag(111) films grown on Fe(110) form very flat surfaces.<sup>7</sup> Furthermore, superconducting quantum interference device (SQUID) magnetometry analysis shows the presence of  $\sim 4\pi M_s$  shape anisotropy for Fe(110)/Ag(111) films with Fe bilayers as thin as 2 ML,<sup>17</sup> behavior which is consistent with the formation of flat Fe bilayers.

Furthermore, it should be remembered that Ag and Fe are very immiscible in bulk, indicating that abrupt, sharp interfaces between Fe and Ag are obtainable. In fact, it is difficult to produce Fe/Ag alloys. Such suspected Fe/Ag alloys are typically produced from ion implantation<sup>18</sup> or rapid quenching processes<sup>19</sup> and are typically characterized by considerable Mössbauer sextet broadening at room temperature or by the presence of Mössbauer singlet/doublet spectral features near zero velocity.<sup>18,19</sup> Experimentally, we find no such evidence of sextet broadening or the presence of singlet and/or doublets in any of the Mössbauer spectra of our 5-ML or 8-ML Fe multilayers.

#### IV. 3-ML Fe(110) COMPONENT MULTILAYERS

Similar to the 5- and 8-ML Fe(110) component multilayers described in the previous section, the Mössbauer spectra of the  $(\text{Ag}_x\text{Fe}_3)_8$  multilayers all exhibited sharp magnetic hyperfine sextet structure with in-plane magnetization throughout the entire temperature range of mea-

surement (4.2–300 K). The room-temperature spectra of  $(\text{Ag}_{20}\text{Fe}_3)_8$  shown in Fig. 5(a) is representative of the spectral appearance of all of the multilayers in this series. In particular, the absence of any additional central features in the spectra is indicative of uniform and continuous film growth.

The temperature dependence of the hyperfine field  $H(T)$  for  $(\text{Ag}_{20}\text{Fe}_3)_8$  is tracked by the open circle data depicted in Fig. 6(a). Note that  $H(T)$  for  $(\text{Ag}_{20}\text{Fe}_3)_8$  closely obeys a linear relation, not a  $T^{3/2}$  relation such as was seen earlier for the 5- and 8-ML Fe component multilayers. Such a linear dependence of  $H(T)$  is often associated with two-dimensional (2D) magnetic behavior,<sup>8</sup> resulting from the thickness-dependent quantization condition imposed on an ultrathin film's magnon wave-vector component normal to the film plane ( $k_y$ ), which may be sufficient to suppress spin-wave excitations normal to the film over an appreciable low-temperature regime.<sup>20</sup>

It has been shown<sup>21</sup> that the temperature dependence of the magnetization  $M(T)$  for a 2D Heisenberg ferromagnet with a magnetic surface anisotropy obeys a quasilinear relation

$$\frac{M(T)}{M(0)} = 1 - \left[ \frac{k_B T}{2\pi D} \right] \exp \left[ \frac{-\Delta}{k_B T} \right], \quad (2)$$

where  $D$  is the bulk spin-wave stiffness and  $\Delta$  is the energy gap induced by the surface anisotropy. However, magnetic relaxation in a thin ferromagnetic film composed of flat nonconnected islands can also result in a

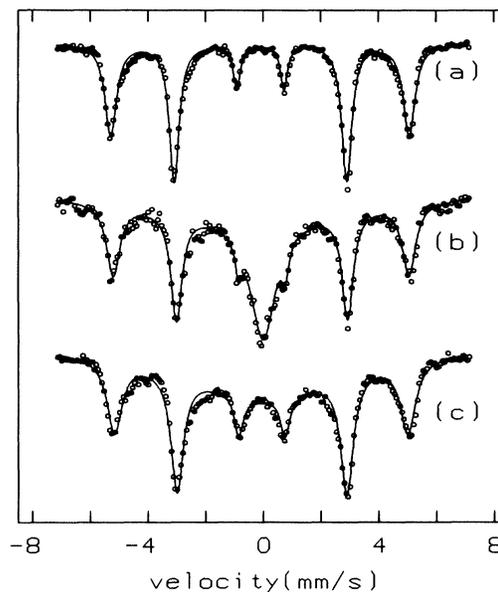


FIG. 5. Mössbauer spectra of (a) the  $(\text{Ag}_{20}\text{Fe}_3)_8$  multilayer measured at  $T = 300$  K with no external applied magnetic field (zero field), which exhibits no magnetic relaxation; (b) the less optimally grown  $(\text{Ag}_{100}\text{Fe}_4)_{10}$  multilayer measured at  $T = 250$  K with no external applied field (zero field); and (c) the  $(\text{Ag}_{100}\text{Fe}_4)_{10}$  multilayer measured at  $T = 250$  K in the presence of  $H_{\text{ext}} = 0.75$  kG applied in the film plane.

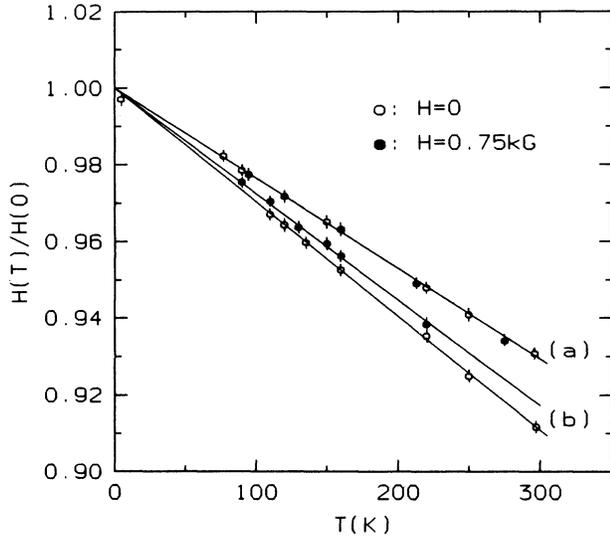


FIG. 6. Hyperfine-field temperature-dependence plots for (a) the  $(\text{Ag}_{20}\text{Fe}_3)_8$  multilayer and (b) the  $(\text{Ag}_{100}\text{Fe}_4)_{10}$  multilayer, which exhibits magnetic relaxation features in its Mössbauer spectrum. For both films the open circles represent zero-field measurements and the solid circles represent the applied-field measurements ( $H_{\text{ext}}=0.75$  kG).

similar linear- $T$  relation for the magnetic hyperfine field  $H(T)$ .<sup>17</sup> Since none of the  $(\text{Ag}_x\text{Fe}_3)_8$  multilayers show additional Mössbauer central feature spectral components (which are always associated with magnetic relaxation<sup>17</sup>), we conclude that the observed linear  $H(T)$  of  $(\text{Ag}_{20}\text{Fe}_3)_8$  results from genuine 2D ferromagnetic behavior. This conclusion is supported by comparative Mössbauer measurements performed with and without the presence of a weak applied magnetic field.

To see how this behavior could occur, a simple model of the bilayers was constructed. We modeled the 3-ML Fe(110) bilayers in the  $(\text{Ag}_x\text{Fe}_3)_8$  multilayers with a 2D Heisenberg lattice with in-plane magnetization, designating the  $y$  axis as the film normal direction and the  $z$  axis as the direction of the film magnetization (including the possible presence of a weak collinear applied magnetic field). We include the magnetic surface anisotropy energy as  $f_s = K_s u_y^2 - K_{sp} u_z^2$ , where  $\mathbf{u}$  is the unit vector along the magnetization direction. The resulting Hamiltonian is

$$\mathcal{H} = - \sum_{I,I'} J(I,I') \mathbf{S}_I \cdot \mathbf{S}_{I'} + k_{sp} \sum_I S_{Iy}^2 - k_s \sum_I S_{Iz}^2 - h \sum_I S_{Iz}, \quad (3)$$

where  $J(I,I')$  is the exchange interaction between spin  $\mathbf{S}_I$  at site  $I$  and spin  $\mathbf{S}_{I'}$  at site  $I'$ ,  $k_s = K_s a^2 / S^2$ ,  $k_{sp} = K_{sp} a^2 / S^2$ , and  $h = g \mu_B H_{\text{ext}}$  are the normalized terms associated with the surface anisotropy and the applied magnetic field  $H_{\text{ext}}$  ( $a$  is the lattice parameter,  $g$  is the gyromagnetic factor, and  $\mu_B$  is the Bohr magneton). In the low-temperature region, where the Holstein-

Primakoff transformation can be applied, the Hamiltonian can be written in  $\mathbf{k}$  space and diagonalized to yield the following spin-wave energy spectrum:

$$\omega(k) = \Delta + D a^2 k^2, \quad (4)$$

where  $\Delta$ , the energy gap induced by the surface anisotropy and the external magnetic field, is expressed by

$$\Delta = [(2k_{sp}S + h)(2k_{sp}S + 2k_sS + h)]^{1/2}. \quad (5)$$

For an Fe(110) surface, the surface anisotropy constants  $K_s$  and  $K_{sp}$  have been determined to be  $+3.2$  and  $+0.04$  erg/cm<sup>2</sup>, respectively.<sup>22</sup> Then the application of a small magnetic field of  $H_{\text{ext}}=0.75$  kG (corresponding to  $h \approx 2 \times 10^{-17}$  erg) would only cause a negligible change in the energy gap  $\Delta$ . According to Eq. (2), this change in  $\Delta$  should have no discernible effect on the temperature dependence of the magnetization. Consequently, comparative Mössbauer measurements with and without the application of this small applied field should result in indistinguishable hyperfine-field temperature dependences  $H(T)$ . This expected invariant behavior is indeed seen in Fig. 6(a), which shows no discernible change in  $H(T)/H(0)$  for  $(\text{Ag}_{20}\text{Fe}_3)_8$  between the zero-applied-field case (open circle data points) and the applied-field case (solid circle data points).

For comparison, we deliberately grew a  $(\text{Ag}_{100}\text{Fe}_4)_{10}$  multilayer under growth conditions (i.e., at a reduce substrate temperature of  $T_s \approx 70^\circ\text{C}$ ) known to produce less continuous and more island structured Fe(110) bilayers on Ag(111), leading to a linear temperature dependence of the hyperfine field primarily due to superparamagnetic relaxation.<sup>17</sup> Comparative Mössbauer measurements of such an island-structured Fe film, with and without the application of a weak magnetic field, should produce clearly different slope values of  $H(T)$ . To demonstrate this we expand on the simple model presented in Ref. 22 simply by adding an applied field term ( $\mathbf{M} \cdot \mathbf{H}_{\text{ext}}$ ) to the total free energy. The resulting magnetic temperature dependence for an Fe(110)/Ag(111) film composed of Fe islands of thickness  $d$  and basal area  $A$  becomes [see Eq. (4) in Ref. 17]

$$\frac{M(T)}{M(0)} = \frac{\int \cos\phi \exp(-f/k_B T) d\phi}{\int \exp(-f/k_B T) d\phi} \approx 1 - \left[ \frac{k_B}{2KAd(1+d_0/2d + M_0 H_{\text{ext}}/K)} \right] T, \quad (6)$$

where  $K = 4.5 \times 10^5$  erg/cm<sup>3</sup> is the volume anisotropy constant for Fe and  $M_0 = 1.7 \times 10^3$  erg/(cm<sup>3</sup>G) is the bulk magnetization for Fe. For the  $(\text{Ag}_{100}\text{Fe}_4)_{10}$  multilayer,  $d_0/2d \approx 4.4$  and  $M_0 H/K \approx 3$ . Therefore, the application of the small 0.75-kG field should noticeably reduce the linear slope of  $H(T)$ .

The Mössbauer spectra of  $(\text{Ag}_{100}\text{Fe}_4)_{10}$  at 250 K, measured with and without the application of the 0.75-kG field, are shown in Figs. 5(b) and 5(c), respectively. The application of the magnetic field eliminates the nonmagnetic central spectral component present in the zero-field spectrum. This behavior is a definite sign of the presence

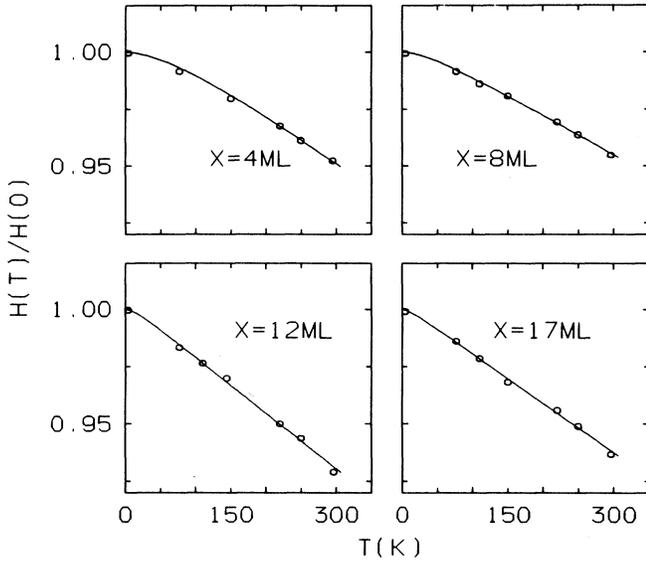


FIG. 7. Hyperfine-field temperature dependence of the  $(\text{Ag}_x\text{Fe}_3)_8$  multilayers, where  $x = 4, 8, 12,$  and  $17$  ML.

of magnetic relaxation of Fe islands in the  $(\text{Ag}_{100}\text{Fe}_4)_{10}$  multilayer.<sup>22</sup> The slope of  $H(T)$  of  $(\text{Ag}_{100}\text{Fe}_4)_{10}$  is also reduced as expected upon the application of the small magnetic field, confirming that the linear temperature dependence is heavily influenced by superparamagnetic relaxation. Therefore, these comparative studies conclusively establish that the linear temperature dependence of the hyperfine field in the  $(\text{Ag}_{20}\text{Fe}_3)_8$  multilayer is not noticeably influenced by superparamagnetic relaxation, implying that the linear  $H(T)$  exhibited by  $(\text{Ag}_{20}\text{Fe}_3)_8$  is the consequence of genuine 2D magnetic behavior.

Next, we consider the magnetic temperature dependence of the  $(\text{Ag}_x\text{Fe}_3)_8$  multilayers with smaller Ag(111) bilayer component thicknesses ( $4 \text{ ML} \leq x \leq 17 \text{ ML}$ ). The  $H(T)$  plots of  $(\text{Ag}_x\text{Fe}_3)_8$  for  $x = 4, 8, 12,$  and  $17$  ML are shown in Fig. 7. It can be clearly seen that as the Ag(111) multilayer component thickness is decreased in

this series,  $H(T)$  assumes an increasingly more bulklike ( $T^{3/2}$ ) appearance. Careful analysis indicated that, strictly,  $H(T)$  can be fit *neither* to a linear relation characteristic of 2D ferromagnetism *nor* to a  $T^{3/2}$  relation characteristic of 3D ferromagnetism. However, this complicated behavior is consistent with the existence of magnetic interaction between neighboring Fe bilayer components through the intervening Ag component. This magnetic coupling, whose strength is expected to be weak and inversely dependent on the Ag(111) thickness, effectively alters the magnetic properties of these  $(\text{Ag}_x\text{Fe}_3)_8$  multilayers whose 3-ML Fe(110) components would otherwise behave as 2D ferromagnets without the coupling.

To better understand how the magnetic coupling between neighboring Fe layers affects the magnetic behavior of these multilayers, we attempted to use a very simple model to describe these films. Suppose that each of the 3-ML Fe(110) bilayer components can be represented by a *single* spin- $\frac{1}{2}$  two-dimensional ferromagnetic plane with  $x, y,$  and  $z$  axes corresponding to the  $[001], [110],$  and  $[\bar{1}10]$  directions, and with an in-plane exchange interaction  $J_0$  and lattice parameter  $a$ . The coupling strength between two adjacent planes (lying in the  $x$ - $z$  plane) separated by a distance  $c$  is described by  $J_1$ . This model is equivalent to a 3D anisotropic ferromagnet whose spin-wave dispersion relation is

$$\omega(k) = 2J_0 \left[ 2 - \cos \left[ \frac{k_x a + \sqrt{2} k_z a}{2} \right] - \cos \left[ \frac{k_x a - \sqrt{2} k_z a}{2} \right] \right] + 2J_1 [1 - \cos(k_y c)]. \quad (7)$$

At temperatures well below the Curie temperature, the planar spin waves that can be excited are only those with long wavelengths defined by  $k_x a \ll 1$  and  $k_z a \ll 1$ . For convenience, we denote  $k_x a, \sqrt{2} k_z a,$  and  $k_y c$  by  $\xi, \eta,$  and  $\zeta,$  respectively. Retaining terms only up to quadratic order in  $\cos[(k_x \pm \sqrt{2} k_z a)/2]$ , the following temperature dependence of the magnetization can be obtained:

$$\frac{M(T)}{M(0)} = 1 - \left[ \frac{1}{2\pi} \right]^3 \int_{-\pi}^{\pi} d\xi \int_{-\pi}^{\pi} d\eta \int_{-\pi}^{\pi} d\zeta \frac{1}{\exp[\omega(k)/k_B T] - 1} \approx 1 - \left[ \frac{k_B T}{2\pi^2 J_0} \right] \int_0^{\pi} \exp \left[ \frac{-2J_1}{k_B T} (1 - \cos \zeta) \right] d\zeta. \quad (8)$$

In the two limiting temperature regimes  $k_B T \gg 2J_1$  and  $k_B T \ll 2J_1$ , the above equation reduces to

$$\frac{M(T)}{M(0)} \approx 1 - \frac{k_B T}{2\pi J_0} \quad \text{when } k_B T \gg 2J_1, \quad (9a)$$

or

$$\frac{M(T)}{M(0)} \approx 1 - \frac{k_B T}{4\pi J_0} \left[ \frac{k_B T}{\pi J_1} \right]^{1/2} \quad \text{when } k_B T \ll 2J_1. \quad (9b)$$

In the low-temperature regime ( $k_B T \ll 2J_1$ ), when the significant contribution comes only from long-wavelength modes in *all* directions, the temperature dependence obeys the  $T^{3/2}$  dependence characteristic of a 3D ferromagnetic system. In the high-temperature regime ( $k_B T \gg 2J_1$ ), the strong thermal excitation of spin waves along the perpendicular ( $y$ ) direction effectively *decouples* the individual 2D ferromagnetic planes. Consequently, these decoupled 2D ferromagnetic planes give rise to a

TABLE II. Results for the in-plane and interlayer exchange interaction strengths  $J_0$  and  $J_1$  obtained from best fits using Eq. (8) for  $(\text{Ag}_x\text{Fe}_3)_8$  heterostructures.  $H_0$  is the hyperfine field at  $T=4.2$  K, and  $T_d$  is the characteristic crossover temperature.

$x$ (ML)	$H_0$ (kOe)	$J_0/k_B$ (K)	$J_1/k_B$ (K)	$T_d$ (K)
4	344.4	710	51	102
5	344.2	750	50	100
6	344.1	720	46	92
7	344.1	770	34	68
8	344.0	890	26	52
10	344.7	640	21	42
12	345.4	660	7	14
14	345.1	660	6	12
17	344.8	740	5	10
20	345.3	670	5	10

linear temperature dependence of the magnetization. Therefore, as  $T$  is increased, the spin-wave excitation of the anisotropic ferromagnetic system undergoes a  $3D \rightarrow 2D$  dimensional crossover. The characteristic temperature  $T_d$  is determined by the interlayer coupling strength according to the relation  $T_d = 2J_1/k_B$ .

Using Eq. (8), excellent fits were obtained for the hyperfine-field temperature-dependence data for all of the  $(\text{Ag}_x\text{Fe}_3)_8$  multilayers. Figure 7 shows that the  $H(T)$  curves obtained from our simple model agrees well with the experimental data from the  $x=4, 8, 12,$  and  $17$  ML Ag(111) component films. Table II compiles the values of  $J_0$  and  $J_1$  obtained from the fits to the data, along with the obtained values of the characteristic crossover temperature  $T_d$ , for all of the  $(\text{Ag}_x\text{Fe}_3)_8$  multilayers. The derived in-plane exchange interaction constants all consistently fall in the range expected for the bulk exchange interaction of Fe. The interlayer coupling strength  $J_1$  monotonically increases with decreasing Ag(111) component thickness ( $x$ ). Compared to the in-plane coupling, however, the interlayer coupling is still considerably weaker. Even for the multilayer with the thinnest Ag(111) component,  $J_1$  is still approximately  $< 0.07J_0$ . Nonetheless, its effect on the magnetic properties of the multilayer is significant. For example,  $J_1$  makes the tem-

perature dependence of the magnetization of the multilayers more bulklike at low temperatures ( $T \ll T_d$ ). Without  $J_1$  the 3-ML Fe(110) components would display a temperature dependence of the magnetization characteristic of a 2D ferromagnetic system over all of the temperature range tested. Since decreasing the Ag component thickness also increases  $T_d$ , the  $H(T)$  becomes more 3D-like over the range of measurement temperatures investigated as  $T_d$  increases.

## V. SUMMARY

$^{57}\text{Fe}$  Mössbauer spectroscopy has been used to study the temperature dependence of the magnetization of three series of Fe(110)/Ag(111) multilayers:  $(\text{Ag}_x\text{Fe}_3)_8$ ,  $(\text{Ag}_x\text{Fe}_5)_{30}$ , and  $(\text{Ag}_x\text{Fe}_8)_{30}$ . Variations of the  $B$ -value prefactors of the observed  $T^{3/2}$  dependence of the magnetization for the 5- and 8-ML Fe(110) component multilayers indicate the presence of magnetic coupling between adjacent Fe bilayer components through the intervening Ag, resulting in more interface/like magnetic behavior as the Ag component thickness increases. The thinner Fe component  $(\text{Ag}_x\text{Fe}_3)_8$  multilayer exhibited a transition from a linear temperature dependence of the hyperfine-field of a 2D ferromagnet when  $x=20$  ML, toward a more bulklike  $T^{3/2}$  dependence when  $x=4$  ML. This crossover behavior is also indicative of the presence of magnetic coupling through Ag.

The sharpness of the Mössbauer spectra as well as the quality of the multilayer RHEED patterns, strongly suggests that the magnetic interaction is indirect in nature. The nature of the interlayer coupling through the non-magnetic Ag bilayer components is not yet clear. Most likely, it is an RKKY-like interaction mediated by the conduction electrons in the Ag. The weakness of the interlayer coupling can be accounted for by the low degree of valence-electron-band overlap between Fe and Ag.

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