Structural studies and magnetic properties of Fe/Ag superlattices

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Fe(110)/Ag(111) superlattices with Ag layer thicknesses of 30 or 60 Å and various Fe layer thicknesses have been fabricated by high-rate sputtering and studied by x-ray diffraction magnetometries, and Mössbauer spectroscopy. A slightly enhanced magnetic moment and hyperfine field, and a reduced T_c have been observed in samples with thin Fe layers. The magnetization exhibits a $BT^{3/2}$ dependence with very large values of B.

The rich phenomena of magnetic thin Fe films have been a subject of persistent interest in recent years. Various studies,[1-6] experimental and theoretical, have been carried out to investigate how reduced layer thicknesses, surfaces and interfaces, and different crystalline orientations affect the moments, the magnetic surface anisotropy, etc. Epitaxially grown Fe on Ag has been a favorable medium for such exploration. Most of the studies utilize Fe(110) on Ag(111), conveniently achieved on mica substrates.[3] A different but more interesting orientation of Fe(100) on Ag(100) has been more recently achieved on GaAs substrates.[5,6] Studies using single-layer Fe thin films however suffer from severe small signal problems, which often limit the measurement to *in situ* high-sensitivity magnetometry, magnetic-optical measurement, and ^{57}Fe Mössbauer spectroscopy.[1,3,4]

Metallic superlattices of Fe/Ag offer many advantages. They not only remove the small signal problem, but also permit the utilization of diffraction studies. In addition, they offer the possibilities of exploring interlayer coupling and superlattice effects. In this work we describe the studies of the structural and magnetic properties of Fe(110)/Ag(111) superlattices. A large number of samples have been studied with different Fe layer and Ag layer thicknesses. We shall restrict our discussion to samples with Ag thicknesses of either 30 or 60 Å.

The Fe/Ag superlattice films have been fabricated by using a sputtering system equipped with two dc magnetron guns and a rotating substrate platform. The vacuum prior to sputtering was about 2×10^{-7} Torr. A series of films with Fe layer thicknesses ranging from a few to 60 Å, and Ag layers of 30 to 60 Å, have been made on mica substrates maintained at room temperature. An Ag buffer layer of about 500 Å was deposited first, followed by multilayer deposition, and finally a 200-Å Ag cover layer. Typically there were a total of about 120 bilayers for each sample.

The Fe/Ag superlattices have been studied by $\theta/2\theta$ x-ray diffraction. The d-spacings of Ag(111) and Fe(110) would give rise to diffraction peaks at $2\theta = 38.15°$ and 44.6°, respectively. Because of the superlattice structure, satellite peaks appears at intervals of $\Delta q = n(2\pi/\lambda)$, where n is an integer and λ is the superlattice parameter. Representative diffraction data are shown in Fig. 1. That only peaks near the Ag(111) and Ag(222) reflections are observable confirms that all the samples have Ag(111) orientations. The diffraction peaks near the Fe(110) and Fe(220) reflections however are observable only for relatively thick Fe layers. All samples also have a single peak at 38.15° due to the thick Ag(111) seed layer and overlayer mentioned above.

It is instructive to calculate the diffraction pattern of an ideal superlattice consisting of n_a atomic planes of spacing d_a and n_b atomic planes of spacing d_b in which the bilayer

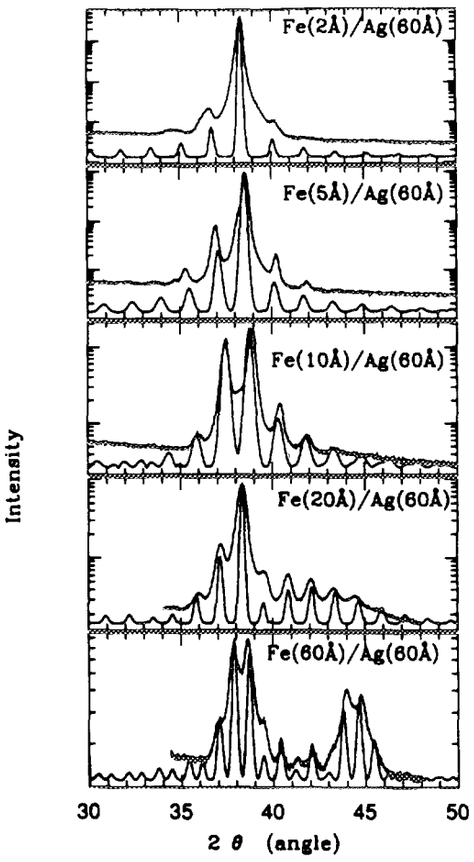

FIG. 1. X-ray diffraction data (upper patterns) of Fe(110)/Ag(111) superlattices with a fixed Ag layer thickness of 60 A. Fe layer thickness ranging from 2 to 60 Å. The theoretical diffraction patterns (lower patterns) assuming an ideal superlattice are also shown.

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thickness $\lambda = n_a d_a + n_b d_b$ is repeated N times. The diffraction intensity has the form of[7]

$$I_{(q)} = \left(\frac{\sin(N\lambda q/2)}{\sin(\lambda q/2)}\right)^2 \left[f_a^2 \left(\frac{\sin(n_a d_a q/2}{\sin(d_a q/2}\right)\right)^2 + f_b^2 \left(\frac{\sin(n_b d_b q/2)}{\sin(d_b q/2)}\right)^2 + 2 f_a f_b \cos\lambda \times \frac{\sin(n_a d_a q/2)\sin(n_b d_b q/2)}{\sin(d_a q/2)\sin(d_b q/2)}. \quad (1)$$

The first factor gives the superlattice peaks at $q = n(2\pi/\lambda)$. The second factor is the envelope function which gives the intensity of the superlattice peaks. In evaluating Eq. (1), the q dependence of the scattering factors f_a and f_b have also been included. The theoretical diffraction patterns of ideal Fe(110)/Ag(111) superlattices are calculated from Eq. (1) as shown in Fig. 1. One notes the following general features: First of all, the f^2 dependence in the envelope function favors the stronger scatterer (i.e., Ag, with f^2 nearly five times that of Fe). Consequently, the intensities of the peaks near the Fe(110) are not noticeable until the Fe layer thickness is comparable to or larger than that of Ag. The diffraction patterns are generally asymmetrical with a large number of superlattice peaks lying *between* $2\pi/d_a$ and $2\pi/d_b$. Finally, the angle of the most intense peak (often called the zeroth order peak) is slightly different from $2\pi/d_a$ and $2\pi/d_b$. A close resemblance between the experimental results and theoretical patterns assuming ideal superlattices is evident. Though it may not be obvious from the diffraction patterns for thin Fe layers, all the Fe layers have a [110] orientation. It should also be mentioned that some superlattices with Fe layer thickness as small as 2 and 5 Å (approximately 1 and 2 monolayers) still exhibit superlattice peaks indicating that the layer structure remains and not a homogeneous alloy. These results indicate sharp interfaces between Fe and Ag.

Magnetization with field parallel to the film plane of the samples with Fe layer thicknesses of 5, 10, and 20 Å and a fixed Ag layer thickness of 30 Å is shown in Fig. 3. All the samples are ferromagnetic. The Fe moment in each case is close to, but appears to be slightly higher than, that of bulk Fe (220 emu/g). The enhancements depend inversely on the layer thickness. Calculations by Ohnish *et al.*[2] have indicated greatly enhanced surface moments of $\mu_{Fe}(100) = 2.98\mu_B$ and $\mu_{Fe}(110) = 2.65\mu_B$, whereas $\mu_{Fe}(\text{bulk}) = 2.2\mu_B$. These very large values of μ_{Fe}, to our knowledge, have not been confirmed experimentally in either (110) or (100) layers.

The ease with which the magnetization is aligned, shown in Fig. 2, implies that the Fe moments lie in the plane. This conclusion is confirmed by Mössbauer spectroscopy measurement. The sample with nominally 5-Å Fe layers shows a noticeably different behavior, that is, a larger field is required to align the moments. Whether this is due to the emergence of perpendicular magnetic surface anisotropy as in the Fe(110)/Ag(111) superlattices[7] requires further investigation.

The temperature dependence of the magnetization $M(T)$ has been measured by a SQUID magnetometer. In

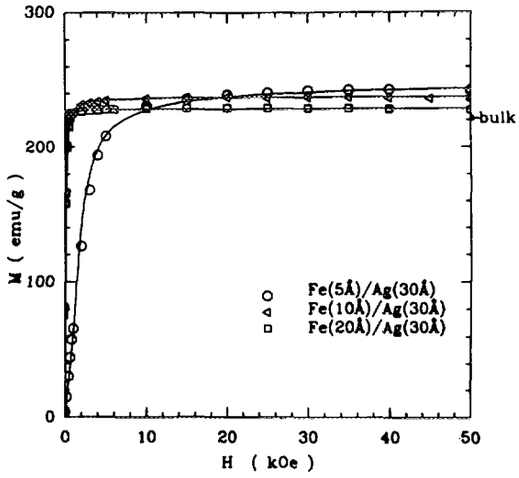

FIG. 2. Magnetization at 5 K for Fe(110)/Ag(111) superlattices with Fe layer of 5, 10, and 20 Å, and a fixed Ag layer (30 Å).

Fig. 3 magnetization in reduced units is plotted as a function of temperature. It is evident that for thinner Fe layers, $M(T)$ decreases more rapidly with T. The samples with Ag layer of 60 Å give essentially the same results. Ag layers of 30 Å is sufficiently thick to decouple the Fe layers. Except for the 5 Å Fe layer, the actual temperature dependence of $M(T)$ is unmistakably $T^{3/2}$. As shown in Fig. 4, the data can be excellently described by $T^{3/2}$. It is well known that at low temperature, the magnetization of a ferromagnet has a temperature dependence of

$$M(T) = M(0)(1 - BT^{3/2} - CT^{5/2}\cdots) \quad (2)$$

due to spin-wave excitations. The Bloch's $T^{3/2}$ law has been observed in many crystalline ferromagnets (e.g., Fe, Ni), amorphous ferromagnets (e.g., a-$Fe_{80}B_{20}$), thin magnetic layers, and surfaces of ferromagnets.[1,8,9] In the case of Fe(20 Å)/Ag(30 Å) shown in Fig. 5, one obtains $B = 17\times10^{-6}$

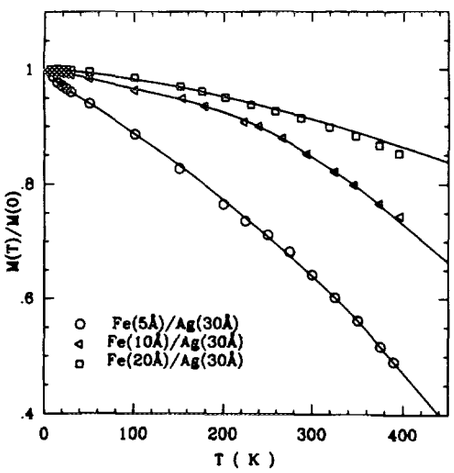

FIG. 3. Magnetization in reduced units vs temperature for Fe(110)/Ag(111) superlattices with Fe layer of 5, 10, and 20 Å and Ag layers of 30 Å.

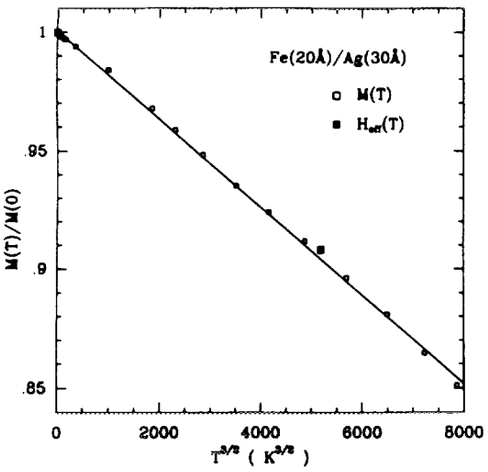

FIG. 4. Reduced magnetization [$M(T)$] and hyperfine field [$H_{\rm eff}(T)$] vs $T^{3/2}$ of a Fe(20 Å)/Ag(30 Å) superlattice.

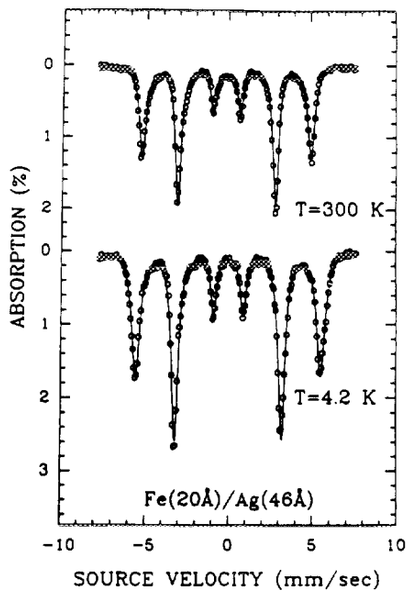

FIG. 5. Mössbauer spectra of Fe(20 Å)/Ag(46 Å) at 4.2 and 300 K.

$K^{-3/2}$ which is much larger than the value of $B = 3.5\times10^{-6}\ K^{-3/2}$ and $B = 7.5\times10^{-6}\ K^{-3/2}$ for bulk Fe and Ni, respectively. Large values of B have also been observed in amorphous ferromagnets and surfaces.

An enhanced B value is the result of a lower T_c and a modification of the spin-wave dispersion relation $\hbar\omega = Dk^2$ due to the thin layer nature of the ferromagnet.[8] To remove effects due to different T_c, it is useful to rewrite Eq. (2) in reduced units,

$$\frac{M(T)}{M(0)} = 1 - B_{3/2}\left(\frac{T}{T_c}\right)^{3/2} - C_{5/2}\left(\frac{T}{T_c}\right)^{5/2} \cdots . \qquad (3)$$

The values of $B_{3/2}$ for both bulk Fe and Ni turn out to be about 0.11. In the case of Fe(20 Å)/Ag(30 Å) we have not yet accurately determined the value of T_c. However, preliminary data using a fast thermal gravimetric method indicates that T_c is approximately 800 K, which is significantly lower than the value 1042 K for bulk Fe. This value of T_c leads a value of $B_{3/2} \approx 0.38$, which is much larger than that of bulk Fe. It may be mentioned that comparably large values of $B_{3/2}$ have previously been observed in amorphous ferromagnets due to the presence of a distribution for exchange interactions.[8]

We have also measured the Mössbauer spectra of some of the samples. An example is shown in Fig. 5 for Fe(20 Å)/Ag(46 Å) corresponding to 10 Fe(110) layers and 20 Ag(111) layers. From the intensity ratio it is immediately obvious that the moments lie in the film plane, confirming the magnetization data. The values of the hyperfine field $H_{\rm eff}$ at 4.2 and 300 K are, respectively, 347 and 320 kOe. The corresponding bulk values are 342 and 330 kOe. Thus, the hyperfine field at 4.2 K is slightly larger than the bulk, whereas at 300 K it is less than bulk. Both results are consistent with magnetization measurements; suggesting that $H_{\rm eff}$ scales with $\mu_{\rm Fe}$. It is also important to note that magnetization and hyperfine field show the *same* temperature dependence, as shown in Fig. 4.

In summary, we have successfully fabricated Fe(110)/Ag(111) superlattices. We have characterized the samples using x-ray diffraction, and studied the magnetic properties by SQUID magnetometry and Mössbauer spectroscopy. We have observed a slightly enhanced magnetic moment and hyperfine field, the $BT^{3/2}$ dependence for the magnetization with a large B coefficient and a reduction of T_c for thin Fe constituent layers.

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