Thermal Magnetic Relaxation in Quasi-Two-Dimensional Fe Films

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Several Fe(110)/Ag(111) superlattices with the Fe component 2 monolayers thick and the Ag component of varying thickness were produced by molecular-beam epitaxy. The Curie temperature was found to be higher than 500 K and a central feature in the Mössbauer spectra is shown to come from thermal magnetic relaxation of Fe islands. The magnetic hyperfine field has a linear temperature dependence and this is explained by a simple model based on the island structure of the Fe.

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The application of molecular-beam epitaxy (MBE) to the growth of thin metallic films has made it possible to grow ultrathin high-quality single crystals and superlattices with monolayer precision. These advances have led to an increased interest in the experimental investigation of two dimensional (2D) or near 2D magnetic systems. A particularly fascinating problem involves the long-range ferromagnetic order of a two-dimensional lattice. Experimentally, investigators have examined the question of the reduction of the Curie temperature, T_C, as a function of the ferromagnetic film thickness, but conflicting results have been obtained.[1-5] It is not unreasonable to speculate that the variations in the observed values of the Curie temperature may be related more to the quality of the films than to any genuine 2D ferromagnetic behavior.

As films get very thin, particularly in the range of 1 monolayer (ML), they may no longer be continuous, but rather consist of islands. Evidence for this island structure is provided by experiments which indicate that the shape anisotropy of the Fe films, $4\pi DM_s$, monotonically decreases as the films get thinner and thinner.[6] Also, a central feature which appears to be due to nonferromagnetic Fe is usually observed in the Mössbauer spectra for Fe films thinner than 4 ML.[7,8] Under the certain conditions, the thermal fluctuations of the magnetic moments of the individual islands could overcome the anisotropy energy barrier, producing a zero average magnetic moment and resulting in superparamagnetic behavior which shows up as a central feature in Mössbauer spectra. The temperature at which this occurs could be mistaken for the Curie temperature when, in fact, what is being measured is more accurately known as the blocking temperature. In this paper we examine ultrathin Fe/Ag superlattices in order to clearly distinguish between the magnetic relaxation effect and those phenomena clearly associated with 2D magnetism.

Three Fe(110)/Ag(111) superlattices were grown in a PHI 430B MBE system with the Fe component 2 ML thick and the Ag component 5, 12, and 20 ML thick, respectively. Both components were repeated 30 times so that the form of the films was $(Ag_xFe_2)_{30}$ with $x=5$, 12, and 20. During the growth, the substrate of synthetic Fe-free mica was kept at a temperature of 180 °C and the growth was monitored by reflection high-energy electron diffraction (RHEED) and a residual gas analyzer (RGA). The sharp, well defined streaks in the RHEED patterns indicate that the films have a very flat surface at each step of the growth. After the films were made, they were also characterized by various other methods such as LEED, Auger-electron spectroscopy (AES), transmission electron microscopy (TEM), etc. The results show that high-quality single-crystal superlattices have been obtained with no evidence of interdiffusion between Fe and Ag. A detailed discussion about the growth condition and the characterization of our films appears elsewhere.[9] dc SQUID measurements were made on these superlattices. The results for the applied field both parallel and perpendicular to the film surface are shown in Fig. 1. There is a strong shape anisotropy between the two directions, with a saturation field in the perpendicular direction of approximately 20 kG. This value is nearly equal to the value of the shape anisotropy $4\pi M_s$ ($\sim$21.5 kG) expected for a flat Fe film. This indicates that if the Fe components are not continuous but do consist of islands, then the island size must be much greater than the Fe thickness and that the islands must be flat.

The superlattices were analyzed by transmission

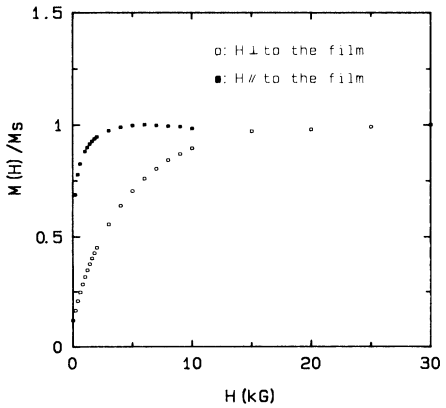

FIG. 1. The M-H curves of $(Ag_{12}Fe_2)_{30}$ at $T=5$ K with H parallel and perpendicular to the film.

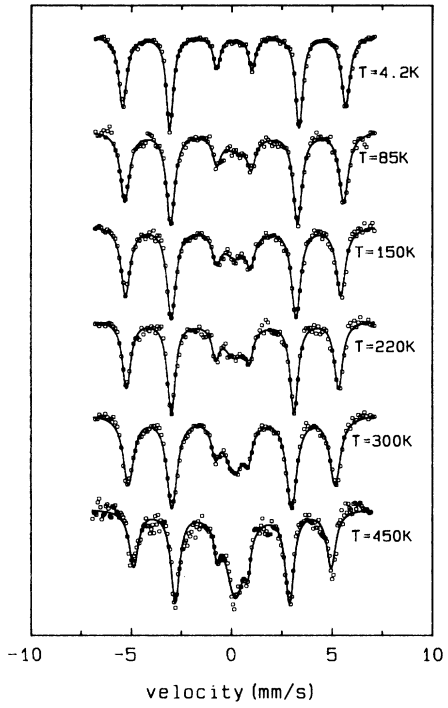

FIG. 2. Mössbauer spectra of $(Ag_5Fe_2)_{30}$ at different temperatures.

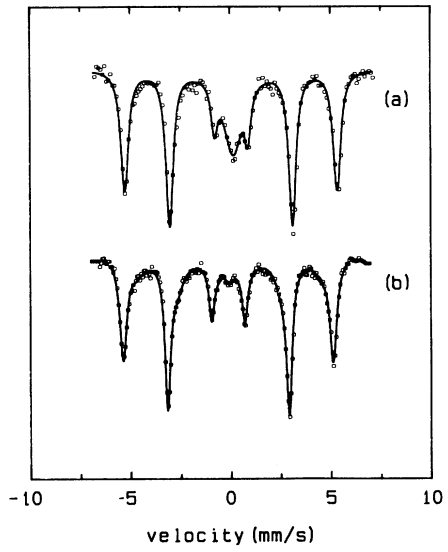

FIG. 3. Mössbauer spectra of $(Ag_{12}Fe_2)_{30}$ at room temperature, (a) without external magnetic field and (b) with an about 5-kG magnetic field parallel to the film.

Mössbauer spectroscopy in order to obtain information from all thirty Fe components. The measurements were made from 4.2 to 450 K. The spectra of $(Ag_5Fe_2)_{30}$ at different temperatures are shown in Fig. 2 and were fitted by one sextet plus a single central component. The spectra of the other two superlattices are similar. The relative intensity of the lines forming the sextet is 3:4:1, indicating that the magnetic moment of each Fe layer is in the plane. Although the intensity of the additional central feature clearly increases as the temperature increases, there is no evidence that the spectrum has totally collapsed even up to 500 K, indicating that the Curie temperature is clearly above this value.

The central feature in all these spectra is a superparamagnetic component resulting from the thermal relaxation of the magnetic moment of small Fe islands and is not connected with a reduced Curie temperature. Figure 3 strikingly proves this. Figures 3(a) and 3(b) both show the Mössbauer spectrum of $(Ag_{12}Fe_2)_{30}$ at room temperature, but the spectrum in Fig. 3(b), which shows a greatly reduced central feature, was taken with a 5-kG external magnetic field parallel to the film. Without the external field the central feature makes up $\sim 28\%$ of the spectrum's intensity. With the magnetic field, the value is reduced to less than 3%. The energy barrier supplied by the external field to each Fe atoms is about $2.2\mu_B H \simeq 10^{-16}$ erg, which is much less than the thermal energy at room temperature, $k_B T \simeq 3\times 10^{-14}$ erg. The external magnetic field is not strong enough to align the Fe moments individually. Therefore, this central feature must not be associated with the ferromagnetic coupling between Fe atoms, but probably represents the thermal relaxation of small Fe islands each of which is already ferromagnetically ordered.

The hyperfine fields of the three superlattices obey a linear temperature dependence (Fig. 4) rather than the usual $T^{3/2}$ dependence found in 3D ferromagnets. This linear behavior has frequently been observed for Fe films approaching 1 ML and it has often been attributed to their 2D nature. However, if this were the case, we would expect that the hyperfine field would decrease more rapidly as the thickness of the Ag component in-

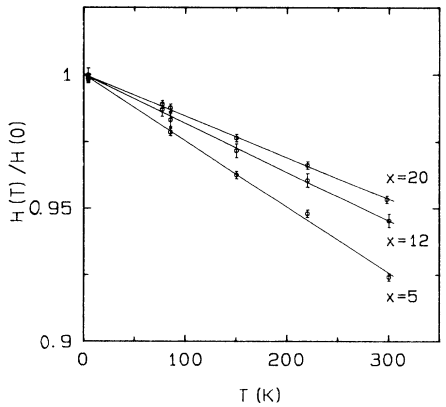

FIG. 4. The temperature dependence of the hyperfine field for $(Ag_xFe_2)_{30}$.

TABLE I. The temperature dependence of the hyperfine field fitted by $H(T)=H_0(1-\alpha T)$.

Film	$10^4\,\alpha$ (K^{-1})	Average size of islands $\langle l\rangle$ (Å)	Relative weight of central peak at 295 K
$(Ag_{20}Fe_2)_{30}$	1.56	158	0.23
$(Ag_{12}Fe_2)_{30}$	1.82	146	0.28
$(Ag_5Fe_2)_{30}$	2.48	125	0.21
$(Ag_{20}Fe_2)_5$	5.69	83	0.63
$(Ag_{20}Fe_2)_5a1$	3.84	100	0.43
$(Ag_{20}Fe_2)_5a2$	2.60	122	0.38
$(Ag_{20}Fe_2)_5a3$	2.08	137	0.39

creased. This trend is expected because as the Ag becomes thicker, the interaction between neighboring Fe film components becomes weaker, making it easier to excite the Fe magnetic moments.[10] In fact, as Fig. 4 and Table I indicate, the actual trend is just the opposite; as the Ag becomes thicker the hyperfine field decreases less rapidly. Therefore, this linear temperature dependence is not associated with a 2D effect. Rather, it is a result of the island structure of the films.

To demonstrate how this linear temperature dependence could result from an island structure, we have developed a simple model relating the slope of the $M(T)$ vs T plot to the island size. The Fe(110) surface has the geometry shown in Fig. 5. The anisotropy energy per unit volume is $f_v=K(u_\xi^2u_\eta^2+u_\eta^2u_\zeta^2+u_\zeta^2u_\xi^2)$, where $K=4.5\times10^5$ ergs/cm^3 and $\mathbf{u}=\mathbf{M}/M_s$ is the unit vector along the direction of the magnetic moment. After a transformation from the $\xi\eta\zeta$ frame to the xyz frame, f_v will have the form of

$$f_v=(K/4)(4u_x^4-3u_y^4-4u_x^2+2u_y^2+4u_x^2u_y^2+1)\,.$$

Adding to this the shape anisotropy $4\pi M_s^2u_z^2$ and the surface anisotropy[11] $f_s=K_su_z^2-K_{sp}u_x^2$, where $K_s=+3.2$ ergs/cm^2 and $K_{sp}=+0.04$ erg/cm^2, we have as the free energy of a Fe(110) island of thickness d and area S

$$f=S[2(K_su_z^2-K_{sp}u_x^2)+4\pi M_s^2u_z^2d+(Kd/4)(4u_x^4-3u_y^4-4u_x^2+2u_y^2+4u_x^2u_y^2+1)]\,. \quad (1)$$

Since both the surface anisotropy and the shape anisotropy force the spins to lie in the plane, we will only consider thermal relaxation in this plane. Therefore, we can let $u_z=0$ and $u_x=\cos\phi$ and $u_y=\sin\phi$. After some algebra, Eq. (1) simplifies to

$$f=S[-(3Kd/4)\cos^4\phi+(Kd-2K_{sp})\cos^2\phi]\,. \quad (2)$$

For thickness d less than $d_0=8K_{sp}/K\simeq71$ Å, the easiest magnetization direction is $[\bar110]$.[11] The average magnetic moment of this island is then

$$\langle u\rangle=\frac{\int_0^{\pi/2}\cos\phi\, e^{-f/k_BT}d\phi}{\int_0^{\pi/2}e^{-f/k_BT}d\phi}\,. \quad (3)$$

If the temperature is not high enough to overcome the energy barrier $(KSd/12)(1+d_0/2d)^2$ resulting from Eq. (2), we can expand $\cos^2\phi$ as $\cos^2\phi\simeq1-\phi^2$. Formula (3) now yields a quasilinear temperature dependence:

$$\langle u\rangle=1-\frac{k_B}{2KSd(1+d_0/2d)}T\,. \quad (4)$$

Using the values of the slopes shown in Fig. 4 (and listed in Table I), an average island size $\langle l\rangle\simeq\langle S\rangle^{1/2}$ for all of the films is calculated and is shown in Table I. Although this average size of 10^{-2} Å is much greater than the films' thickness of 4 Å, the islands sufficiently influence the thermal excitations to produce a quasilinear

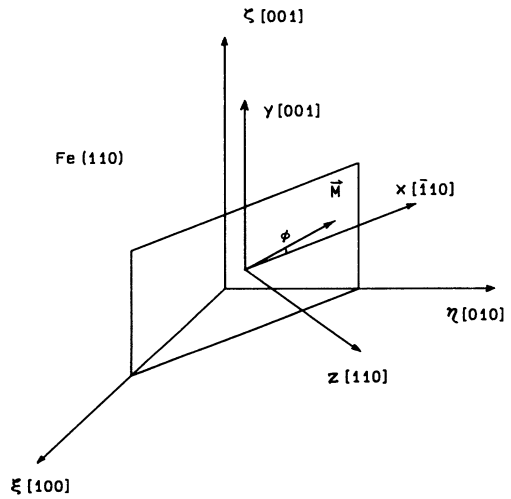

FIG. 5. Configuration of Fe(110) film.

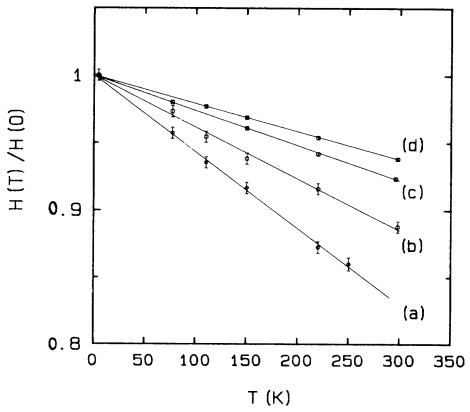

FIG. 6. The temperature dependence of the hyperfine field for $(Ag_{20}Fe_2)_5$ superlattice grown at 75 °C (plot *a*) after the film was made, (*b*) after annealing in Ar at 200 °C for 1 h, (*c*) after annealing in Ar at 250 °C for 1 h, and (*d*) after annealing in vacuum at 350 °C for 1 h.

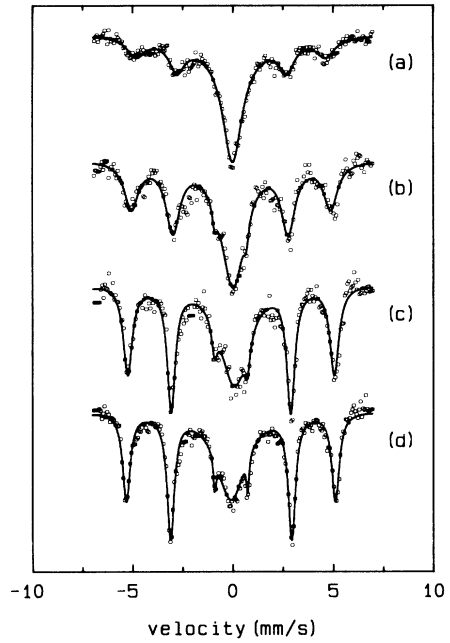

FIG. 7. The room-temperature Mössbauer spectra of $(Ag_{20}Fe_2)_5$ (a) after the film was made, (b) after annealing in Ar at 200 °C for 1 h, (c) after annealing in Ar at 250 °C for 1 h, and (d) after annealing in vacuum at 350 °C for 1 h.

temperature dependence. Obviously there is a range of island sizes, with the smaller islands becoming superparamagnetic at a lower temperature than the larger ones. A simple calculation based on Eq. (2) shows that islands which contribute to the superparamagnetic feature in the Mössbauer spectra at room temperature have a size less than 60 Å.

In order to test this model, another superlattice $(Ag_{20}Fe_2)_5$, was made at 75 °C rather than 180 °C. From our previous work,[11] it is expected that a lower substrate temperature produces a rougher surface, promoting the growth of smaller islands. If our model is correct, this film should exhibit a larger superparamagnetic feature and a hyperfine field with a larger slope than the films grown at higher temperature. Also, these islands might coalesce upon annealing the film at high temperature, decreasing both the superparamagnetic component and the slope of the hyperfine field. After taking the Mössbauer spectra of this film it was annealed in Ar gas at 200 and 250 °C for 1 h and subsequently annealed in vacuum for 1 h at 350 °C [and denoted in Table I as $(Ag_{20}Fe_2)_5a1$, $(Ag_{20}Fe_2)_5a2$, and $(Ag_{20}Fe_2)_5a3$, respectively]. Figures 6 and 7 show the temperature dependence of the hyperfine field and the room-temperature Mössbauer spectra, respectively, after each stage of the anneal. The data clearly support our model. After each stage of the anneal both the slope and the central feature decrease, indicating that the islands have coalesced.

One remaining question concerning our films is why the slope of the hyperfine field is inversely proportional to the Ag thickness in $(Ag_xFe_2)_{30}$. We believe this can be easily understood from the fact that Ag grows more smoothly on Fe than Fe does on Ag. Therefore, a thin layer of Ag on top of an Fe layer will be rougher than a thicker Ag layer. The rough, thinner layer will subsequently produce a rougher Fe layer with a greater number of small islands. As the Ag becomes thicker and smoother the subsequent Fe layer will be smoother with larger islands. This model of growth is completely consistent with the observed slopes of the hyperfine field.

In conclusion, we have found that the island structure of ultrathin Fe(110) films plays an important role in the system's thermal excitations even when the island size is much greater than the Fe film thickness. This must be taken into account when attempting to determine the Curie temperature of a 2D system. A simple model was then proposed to relate the slope of the linear temperature dependence of the magnetic hyperfine field to the averaging size of the islands.

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