Two magnetic-ordering temperatures in Fe/Al multilayered films

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We have prepared Fe/Al multilayers with individual layer thickness ranging from 5 to 200 Å. For magnetic measurements, each sample was cooled in zero magnetic field from room temperature to 5 K. Then a magnetic field of 100 G was applied, and the magnetization was measured as the temperature was raised from 5 to 300 K. The magnetization initially rose as the temperature increased to a certain temperature T_p, beyond which the magnetization decreased with further increase in the temperature. The peak at T_p was found in all of our samples of different layer thicknesses. We have also measured the Curie temperature T_C and found it considerably higher than T_p. We show that T_p represents a transition from one magnetic state to another, and it is suggested that it is probably due to an antiferromagnetic coupling between the Fe layers via the Al layers.

INTRODUCTION

Fe-based soft magnetic films with high saturation magnetization and low coercivity are most suitable head core materials for high-density magnetic recording.[1] Multilayered films, particularly those consisting of pure Fe films alternating with nonmagnetic layers, have been expected to have excellent soft magnetic properties. Recently, the Fe/Al multilayered film system has been shown to be a good candidate for magnetic recording.[1] The Fe/Al system has been treated as an ordinary ferromagnet[2] with one transition temperature at the Curie temperature. In this study we have observed a deviation in the magnetic properties of Fe/Al from the normal ferromagnetic behavior. The results are presented and discussed in the paper.

EXPERIMENTS, RESULTS, AND DISCUSSION

We have made Fe/Al multilayered samples with different bilayer thickness λ using a sputtering unit (λ equals the thickness of the Fe layer plus the thickness of the Al layer within each period). The system was pumped down to a base pressure of $\sim 5\times10^{-10}$ Torr. Before being admitted to the chamber, ultra-high-purity Ar gas was further purified by passing it first through a cold trap at 100 K to freeze out water vapor and then through a gas purifier to remove impurities such as O_2 and N^2 by reaction with a hot Ti-based alloy. A computer-controlled substrate holder moved the substrate quickly between the Fe and the Al sources. Each substrate was a $1.2\times1.2\times0.05\text{cm}^3$ polished single crystal of Si. A quartz-crystal-film monitor was used to determine the thickness of the layer.

For every sample, the Fe layers have the same thickness as the Al layers and the total thickness of the sample was 5000 Å. We chose to have the same layer thickness for Fe and Al in each sample in order to compare this work with our earlier study on Cu/Ni multilayered films.[3] In that study, the Cu and Ni had the same thicknesses in every sample. Both the Cu/Ni and the Fe/Al multilayers were deposited on substrates at about room temperature.

The quality of the Fe/Al multilayered films was characterized by x-ray diffraction. The presence of the satellite peaks at low Bragg angle is indicative of the multilayered structure (Fig. 1). The broadness of the satellites probably is due to the alloying at the interfaces since Al is soluble in Fe. Despite the alloying effect, a satellite was observed for $\lambda = 10$ Å sample (5 Å Fe/ 5 Å Al).

The magnetization measurements were made using a SQUID magnetometer (Quantum Design Model MPMS) with the dc magnetic field parallel to the sample surface. We measured the changes in the magnetization of the

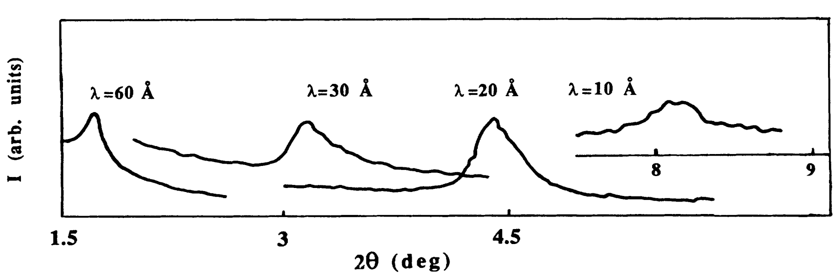

FIG. 1. The first peak of the low-angle x-ray diffraction of some of our Fe/Al multilayers.

$\lambda=60$ Å sample in various fixed magnetic fields as the temperature was raised from 5 to 250 K (see Fig. 2). The sample was initially cooled from room temperature to 5 K in zero applied magnetic field and the data were taken with increasing temperatures to 250 K followed by data taken with decreasing temperatures. Every curve exhibits a peak which shifts to a lower temperature with increasing the applied field. When the sample was subsequently cooled in the same field, the magnetization did not retrace the initial curve in all temperature ranges specially below the temperature T_p at which the magnetization has peaked. This irreversible behavior in the magnetization and the peak in the magnetization are not properties of a typical ferromagnetic material.

The thermoremanent magnetization, TRM, of the same sample was obtained by cooling the sample in 35 000 G field from room temperature to 5 K and then switching the field off to get the remnant. The variation of this TRM with temperature is shown in Fig. 3. The TRM decreased with increasing temperature and it had an inflection point at about 70 K. This temperature is approximately equal to the temperature T_p at which the magnetization curve with 100 G applied field (the lowest field used for this sample) has peaked. The fact that the TRM curve changes course at T_p indicates that T_p represents a phase transition from one magnetic state to another. In our recent study on Cu/Ni multilayered films,[3] we have found magnetization versus temperature curves which are similar to those of Fig. 2. The TRM for Cu/Ni also decreased as the temperature increased and actually vanished at the temperature at which the magnetization has peaked. It seems that, in both Cu/Ni and Fe/Al, a magnetic transition occurs at the temperature T_p corresponding to the peak in the magnetization.

To see whether we have a paramagnetic state above T_p in the Fe/Al $\lambda = 60$ Å sample, we measured the magnetic hysteresis loop between 50 and -50 G applied field at a temperature of 300 K, see Fig. 4. Clearly, from the figure, the state is not a paramagnetic one but rather a ferromagneticlike state. To determine the Curie temperature T_c above which the sample will enter a paramagnetic state, we measured the magnetization versus temperature at 1000 G applied field as shown in Fig. 5. We considered the inflection point in the curve of Fig. 5 to represent roughly the Curie temperature T_c which was equal to ~ 540 K for this sample. The Curie temperature is considerably larger than the temperature of the peak T_p (by 470 K).

The magnetization versus temperature curves at an applied field of 100 G for the rest of our samples with different λ thicknesses were measured. All the samples (λ = 10 to 400 Å) exhibited a peak in the magnetization as well as irreversibilities. Some of these curves are shown in Fig. 6. We were surprised to see a peak in the magnetization and irreversibilities in the λ = 400 Å sample since we expected a ferromagnetic behavior for this sample. At these relatively thick layers of Fe (200 Å) separat-

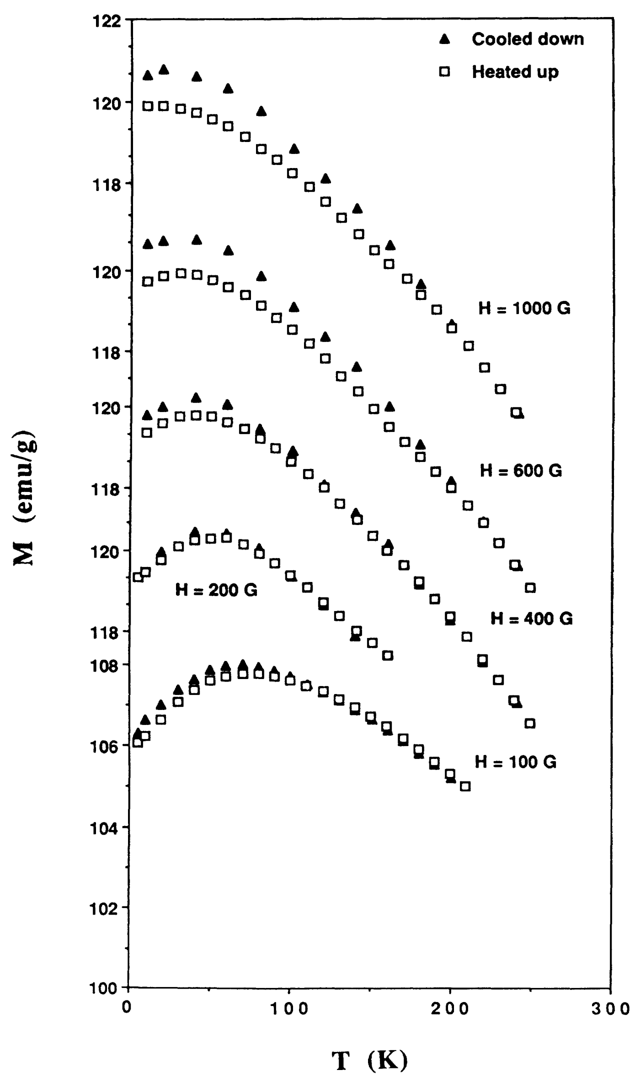

FIG. 2. The magnetization versus temperature of $\lambda=60$ Å sample at different fixed fields.

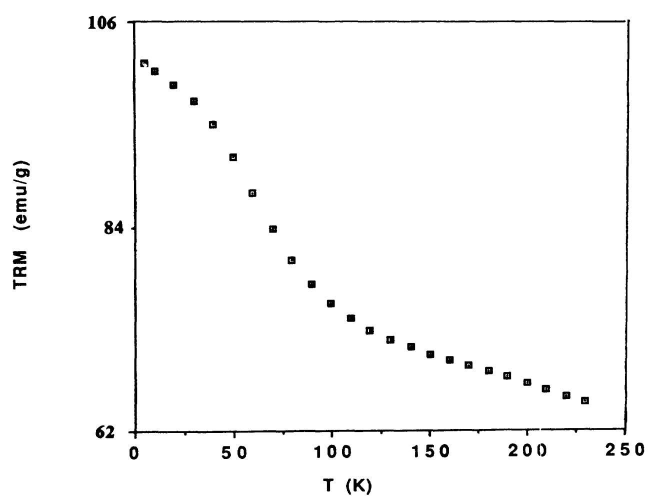

FIG. 3. The thermoremnant magnetization as a function of temperature of the $\lambda=60$ Å sample.

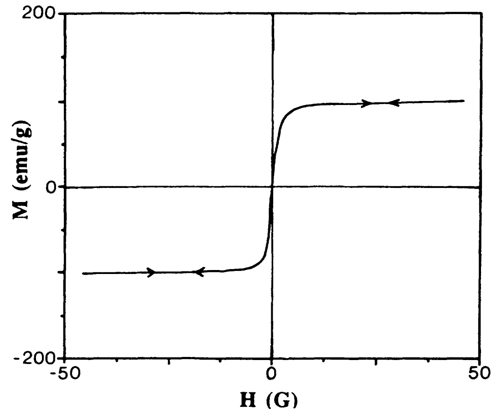

FIG. 4. Hysteresis loop of $\lambda=60$ Å sample at 300 K.

ed by equally thick layers of aluminum, one might expect that there is no coupling of the Fe layers via the 200-Å Al layers and the alloying at the interfaces has little effect on the 200-Å-thick iron layer. If interlayer coupling exists and/or the interface effect is strong, the magnetic properties will be affected and a deviation from a ferromagnetic behavior becomes possible. We think the interfacial effect (due to the alloying of Fe and Al) is negligible in sample with relatively thick layer thickness especially in the $\lambda = 400$ Å sample. Thus the question will be, "Do we have interlayer coupling through the Al layer even when the Al thickness is 200 Å ?" We think the answer to this question is yes, based on the following experiment. We have made a 200-Å single iron film sandwiched between two Al layers. The Al layers serve to create the same interfacial environment for the iron single film as for that in the multilayered film, and to protect the Fe from oxidation. The magnetization versus temperature curve for this 200-Å single film was measured in 100 G field just as in the multilayered films. As shown in Fig. 7, no peak was found in the magnetization. The magnetization at 5 K for this 200-Å single film is 175

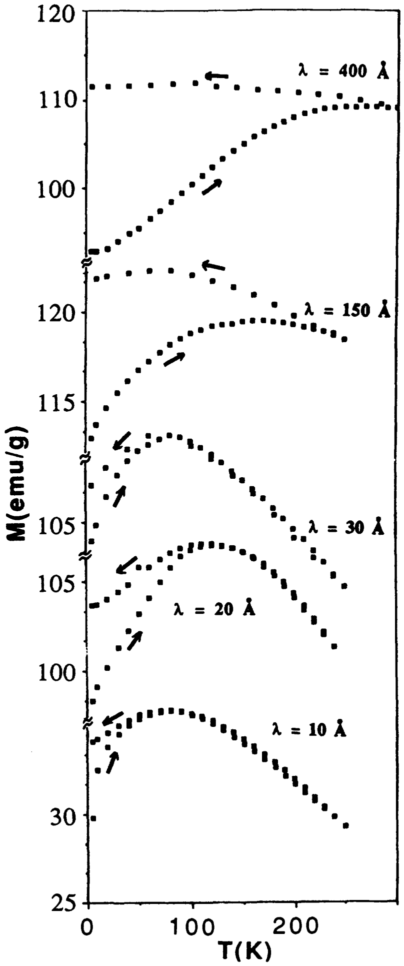

FIG. 6. Magnetization as a function of temperature for some of our Fe/Al films. The arrow to the right represents the heating process and the arrow to the left represents the cooling process.

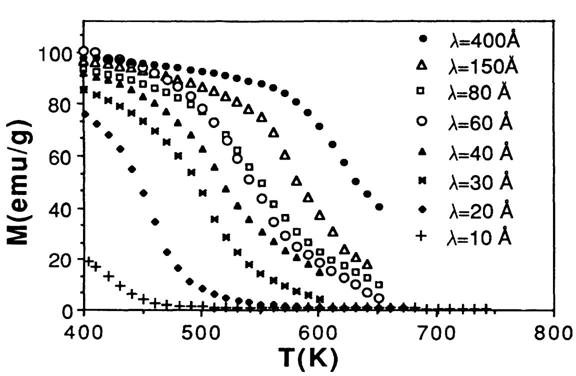

FIG. 5. The magnetization versus temperature at 1000 G field for our Fe/Al multilayered samples of different layer thicknesses.

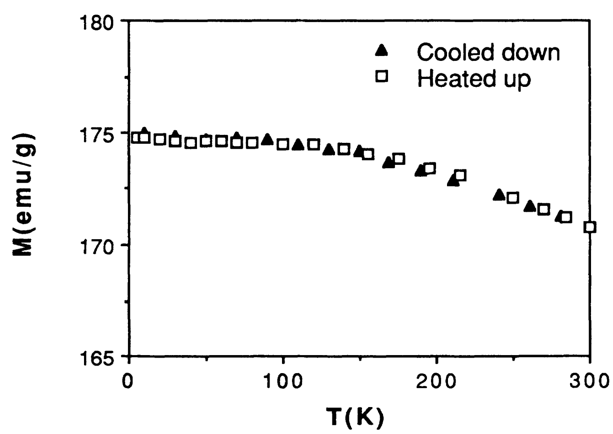

FIG. 7. The magnetization versus temperature for a 200-Å single iron film.

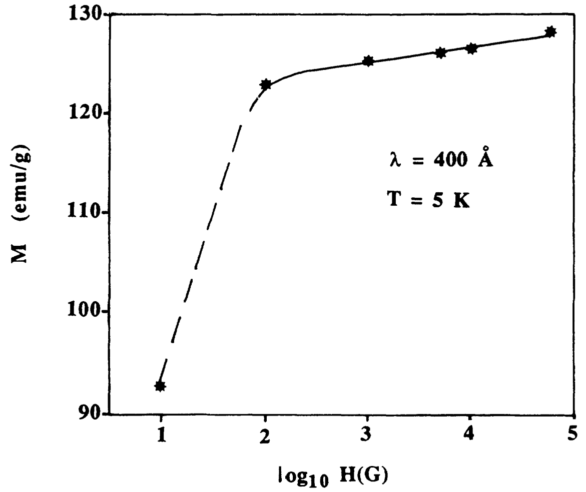

FIG. 8. The magnetization versus the logarithm of the applied magnetic field for the $\lambda=400$ Å sample at 5 K temperature.

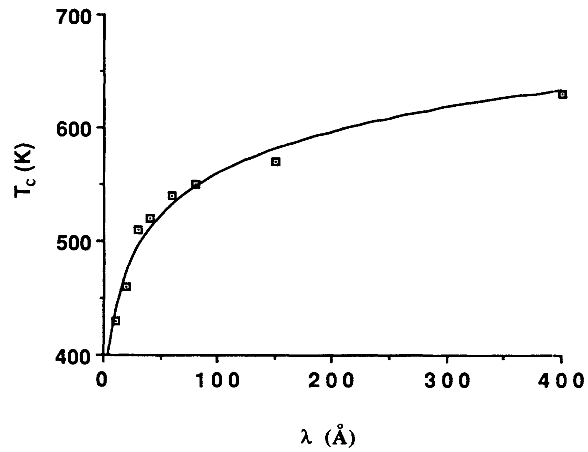

FIG. 10. The Curie temperature is plotted as a function of bilayer thickness λ.

emu/g at 100 G while in the case of the 200-Å Fe/200-Å Al multilayer, the magnetization at 5 K is considerably less even at a high field reaching only 127 emu/g at 55 000 G (about half of the 225 emu/g saturated value for pure iron), see Fig. 8. The reduction in the magnetization values upon multilayering indicates that the interlayer coupling between the ferromagnetic Fe layers is probably antiferromagnetic in nature. It was suggested recently that there could be an antiferromagnetic interaction between ferromagnetic layers via the nonmagnetic layers. However, to our knowledge, most workers could detect interlayer interaction only up to about 60 Å. For example, Cochran, *et al.*[4] concluded, on the basis of ferromagnetic-resonance experiments, Brilliouin light-scattering experiments, and surface magneto-optic Kerr-effect measurements, that there is an antiferrmagnetic coupling between Fe layers through 9 to 12 monolayers of copper. Swaitek *et al.*[5] studied the interlayer coupling between ferromagnetic films separated by nonmagnetic layers by means of light scattering from spin waves and microwave absorption. They determined that there is interaction between the magnetic layers via the nonmagnetic layers. They concluded that this interlayer coupling vanishes when the nonmagnetic layer thickness d_o exceeds 50 Å. But in an earlier study, Massenet *et al.*[6] determined that the interlayer coupling exists even for values of d_o of a few hundred Å.

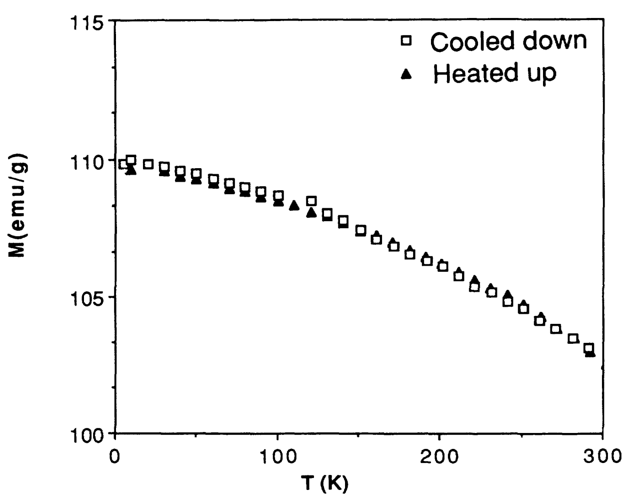

FIG. 9. The magnetization versus temperature of the 30-Å single iron film.

We have measured the magnetization at 100 G applied for another single iron film of 30 Å thickness and also found no peak in the magnetization, Fig. 9. It really seems that the interlayer coupling is causing the peak in the magnetization in multilayered samples.

The Curie temperature T_c was determined for all the samples in a similar way as for the $\lambda=60$ Å sample, see Fig. 5. Unlike T_p, the Curie temperature changes systematically with the layer thickness. It decreases as λ increases as shown in Fig. 10. We list in Table I the values for T_p, T_c, and the difference between the two tempera-

TABLE I. Values of T_p, T_c, and their differences for all samples.

λ (Å)	T_p (K)	T_c (K)	T_c-T_p
10	90	430	340
20	120	460	340
30	80	510	430
40	95	520	425
60	70	540	470
80	95	550	455
150	170	570	400
400	270	630	360

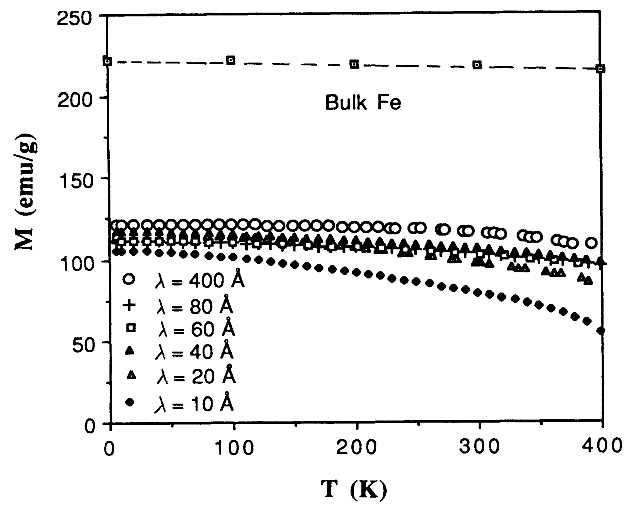

FIG. 11. The magnetization in 5 kG applied field versus temperature of bulk Fe and our Fe/Al multilayered samples.

tures for all of our samples. It is notable that T_c is at least 340 K greater than T_p for any of our samples. The considerable difference between T_c and T_p indicates that T_p does not represent by any means the Curie temperature of the sample. The Curie temperature for the $\lambda=400$ Å sample is 640 K for this sample (as listed in Table I) which is considerably less than that of pure iron (1040 K). Like the magnetization, the Curie temperature of the $\lambda=400$ Å sample is almost half of the correspondence value of the pure iron. The big reduction in T_c from that of pure iron is probably another indication of the antiferromagnetic interaction between the ferromagnetic iron layers through the nonmagnetic Al layers.

At a relatively high applied magnetic field of 5 kG, the peak disappears for all our Fe/Al samples and the magnetization decreases as the temperature increases as shown in Fig. 11. The magnetization curve of bulk iron is also shown in the same figure for comparison. The magnetization M (actually the magnetization divided by the mass of the iron in the multilayered film) is considerably less than that of the bulk iron for all layer thicknesses.

CONCLUDING REMARKS

We have found that Fe/Al multilayered films have two magnetic transition temperatures, T_c and T_p. T_c is the Curie temperature above which the sample becomes a paramagnet and below which it has a ferromagneticlike behavior. The temperature, T_p, that we have identified, is the temperature at which the magnetization peaks and below which irreversibilities set in and the system might enter a different magnetic phase. We are currently investigating the nature of the transition at T_p by studying in details the magnetic properties below and above T_p.

The peak in the magnetization of Fe/Al multilayered films is probably due to the existence of antiferromagnetic interlayer coupling between the iron layers which coexists with the ferromagnetic interaction within each iron layer. This is similar in principle to what happens in spin-glass materials. In these materials, the competition between ferromagnetic and antiferromagnetic interactions causes the spin-glass ordering that is responsible for the peak in the magnetization and irreversibilities.

The effect of the alloying at the interfaces on the magnetization becomes more important as the layer thickness becomes smaller. Thus for samples with small λ, both the interlayer coupling and the alloying at the interfaces are responsible for the magnetic behavior of the sample.

We are planning to investigate the effect of the thickness of the nonmagnetic layer on the interlayer interaction. For a given thickness of the magnetic layer we will prepare a series of samples having different thicknesses of the nonmagnetic layer. Similar series of samples will be prepared for other choices of magnetic layer thickness. Studying the magnetization of these samples will also allow us to determine the critical thickness of the nonmagnetic layer needed to decouple the magnetic layers.

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