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The coupling of interlayers in compositionally modulated (Fe–Si)/Si amorphous films

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Abstract. Compositionally modulated (Fe–Si)/Si amorphous films have been obtained by RF sputtering. Varying the thickness d_s of Si layers with a fixed thickness of the Fe–Si layers, it is found that, when d_s is smaller than a critical value, a strong-coupling effect between magnetic layers begins to occur. The results are that the low-hysteresis effect disappears, the saturation magnetization M_s and Curie temperature T_C increase monotonically with decreasing d_s, and perpendicular anisotropy appears. The experiments showed that M_s increases exponentially and T_C increases linearly with decreasing d_s because of the coupling effect.

1. Introduction

Considerable interest has been developing in recent years in compositionally modulated films (CMFs) because they are a useful tool for studying two-dimensional ferromagnetism, magnetism of interfaces, magnetic coupling of interlayers, surface anisotropy, interdiffusion between layers and so on. They provide a new technique in the fabrication of new artificial materials. There is no serious crystal-lattice-matching problem with the amorphous CMFs, so that better modulated structures can be obtained. In addition, the amorphous materials can be easily prepared; their composition can be changed in a wide range, so that their properties can be varied easily (Xiao *et al* 1987). The compositionally modulated (Fe–Si)/Si amorphous films with different thicknesses of Si layers have been prepared successfully by RF sputtering. The interdiffusivities in this material are as low as 10^{-26}–10^{-25} m^2 s^{-1} (Bruson *et al* 1985); so it is a good material for the study because of its good thermostability.

The CMFs which have a set of magnetic films separated from each other by non-magnetic insulating material are a special kind of material. A dead layer, which consists of paramagnetic clusters of iron atoms, in the interface between magnetic and non-magnetic layers was formed because of the diffusion effect of atoms. If the non-magnetic layers are thin enough, magnetic coupling between magnetic layers is induced and strenthened with decreasing thickness of non-magnetic layers. This causes the thickness of dead layers to decrease, and the magnetization of CMFs to increase. Varying the coupling of interlayers is an effective method for adjusting the properties of CMFs.

2. Experimental details

Specimens were prepared with an RF sputtering system which consists of two targets. The targets were discs made of Fe–Si alloy and Si, respectively. The distance between

Table 1. The thickness d_s of Si layers for as-sputtered CMFS.

Sample	1	2	3	4	5	6	7
d_s (Å)	8.8	13.2	26.4	31.7	39.6	53.0	101

the target and the substrate holder during deposition was about 4 cm; both the target and the holder were water cooled. The RF input power was about 180 W. After the chamber had been evacuated to a high vacuum of about 1×10^{-4} Pa, 99.999% pure Ar gas was introduced. During the sputtering process, the Ar charging pressure in the chamber was kept at 0.5 Pa. The substrates were glass slides of 0.2 mm thickness. Single Fe–Si films with soft magnetic properties were obtained with this system. The composition of Fe–Si films determined by electron microprobe analysis was $Fe_{80.5}Si_{19.5}$ This is the lower limit of the composition where the amorphous nature is observed (Shimada and Kojima 1976). X-ray diffraction measurements showed that crystalline peaks were not found for the single Fe–Si films. (Fe–Si)/Si CMFS with different modulation wavelengths can be obtained by sputtering Fe–Si and Si onto the substrate alternately, and controlling the sputtering time according to the deposition rates. The rates were 0.85 Å s^{-1} and 0.88 Å s^{-1} for Fe–Si and Si, respectively. A series of samples with different thicknesses of Si layers were obtained. The thickness of Fe–Si layers was held constant at 17 Å, while the number of modulation periods was 40 for all samples. The thicknesses of Si layers are listed in table 1. X-ray diffraction analysis showed that all samples were amorphous. A strong modulation in composition was shown by small-angle x-ray diffraction for all samples. Four orders of diffraction peaks can be seen mostly. The modulation wavelengths for all samples were checked using the Bragg law. We used high-order diffraction peaks to evaluate the wavelength in order to reduce the angle error. The results were in good agreement with the values designed according to the deposition rate. The error was within 5%.

The magnetic properties of samples were studied using a microprocessor-controlled vibrating-sample magnetometer. The specimens had an area of 5×8 mm^2. The dependences of the specific saturation magnetization M_s of the samples on the temperature T were measured from room temperature to 700 K with the temperature varied at a rate of about 12 K min^{-1}. The applied magnetic field was 8×10^4 A m^{-1} in the plane of film, which was high enough to saturate magnetically for all samples. Figure 1 shows some of the results obtained in the experiments. The numbers on the curves indicate the thicknesses d_s of the Si layers. In order to determine the Curie temperature T_C, we applied the mean-field approximation for the temperature region immediately below the phase transition. A small-argument expansion of the Brillouin function yields (Hendricks *et al* 1971)

$$[M_s(T)]^2 = A(1 - T/T_C) \tag{1}$$

where A is a constant. When the experimental data were presented as $[M_s(T)]^2$ versus T near the Curie temperature, a straight line resulted. From the intercept of this line, T_C can be determined.

3. Results and discussion

The hysteresis loops of all samples were measured at room temperature. Figure 2 shows some typical results. For the samples with $d_s \geqslant 31.7$ Å, the hysteresis loops are similar

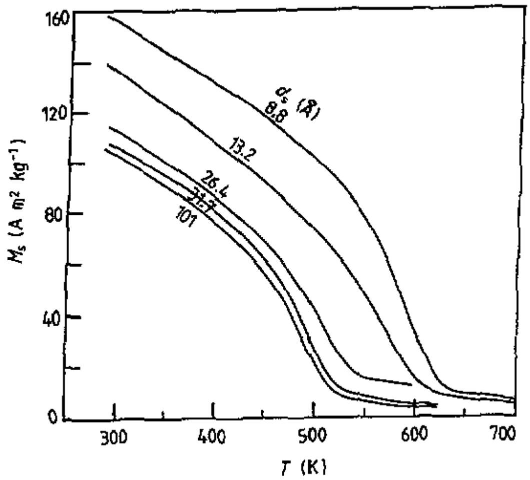

Figure 1. The specific saturation magnetization M_s versus temperature T for different thicknesses d_s of the Si layers.

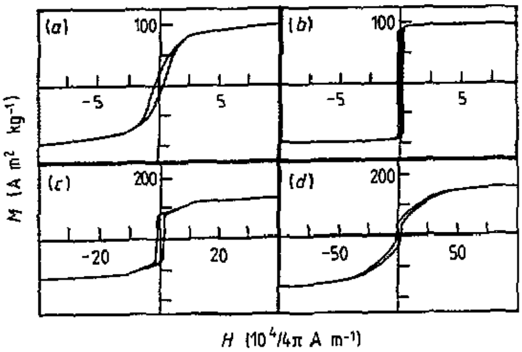

Figure 2. Typical hysteresis loops for CMFs: (*a*) d_s = 31.7 Å; (*b*) d_s = 26.4 Å; (*c*) d_s = 13.2 Å; (*d*) d_s = 8.8 Å.

to figure 2(*a*), which obviously shows a low-hysteresis effect. When the thickness of the Si layers increases, the shape of loop is the same but the maximum magnetizations are slightly reduced. Many experiments have shown that (Shinjo *et al* 1986, Liu and Yang 1988) for a CMF consisting of magnetic and insulating materials, when the thickness of magnetic layers decreases to a certain value and the thickness of non-magnetic layers is thick enough to isolate the coupling between magnetic layers, the effect of low hysteresis or no hysteresis will appear. This is a characteristic of two-dimensional ferromagnetism. When the thickness d_s of Si layers is 26.4 Å, the loops are similar to figure 2(*b*). The low-hysteresis effect disappeared, and the material showed good soft magnetic behaviour. The change in the shape of loops reflects the coupling effect of the magnetic layers, where the main influence is the dipolar field. If the thickness of Si layers decreases from 31.7 to 26.4 Å, the dipolar field between magnetic layers causes the magnetizations in the magnetic layers to cohere. The domain structure of CMFs will also change accordingly (Slonczewski 1966), and the two-dimensional ferromagnetism of samples will disappear.

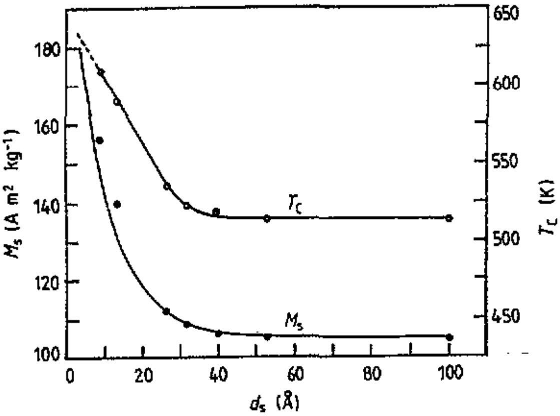

Figure 3. M_s at room temperature and T_C versus the thickness d_s of Si layers. The full curve for M_s is from equation (4).

One can say that, when the thickness of the magnetic layers is 17 Å, the critical thickness of Si layers under which the dipolar interaction between magnetic layers appears is about 30 Å. If the thickness of Si layers decreases further, the samples will show the characteristic of perpendicular anisotropy. Figures 2(*c*) and 2(*d*) show the loops for the samples with $d_s = 13.2$ Å and 8.8 Å, respectively. These are the typical hysteresis loops that films with perpendicular anisotropy exhibit (Prutton 1964, Suran *et al* 1984). The characteristic of perpendicular anisotropy was also verified by magnetic torque measurements in our laboratory. The effect of perpendicular anisotropy of samples increases with decreasing thickness of Si layers, but the shape anisotropy of samples plays a main role in all cases. We do not understand what causes this perpendicular anisotropy yet. It seems different from the surface anisotropy proposed by Néel (1953). It is probably related to the coupling of interlayers and the exchange interaction in the interfaces because it strength is directly related to the thickness of Si layers.

The dependences of M_s at room temperature and T_C on the thickness d_s of Si layers are shown in figure 3. When $d_s > 35$ Å, M_s and T_C for the samples approach constant values. In combination with figure 2(*a*), this indicates that, when $d_s > 35$ Å, the coupling effect between magnetic layers can be neglected, and the magnetic properties of samples are mainly determined by Fe–Si layers. When $d_s < 35$ Å, M_s and T_C increase rapidly with decreasing d_s. This reflects the rapid strengthening of the coupling effect between magnetic layers. The coupling of interlayers includes the dipolar and exchange interactions of magnetization in the magnetic layers through the barrier of Si layers, and the exchange interaction of the magnetic atoms in the interfaces through the Fe atoms which are inevitably introduced into the Si layers by the sputtering process. The results of these interactions cause the internal field to increase and the dead layers to become thinner. So M_s and T_C increase.

There is a zone of interfacial iron atoms between the Fe–Si and Si layers which consists of paramagnetic clusters of iron atoms and weaker magnetic iron atoms. One can use an equivalent dead layer instead of this interfacial zone. The magnetic properties of CMFs are determined by these zones. It is shown by the curves in figure 3 that, if the Si layers are thicker ($d_s > 35$ Å), the thicknesses of the equivalent dead layers are unchanged on varying the thickness of the Si layers. When the Si layers are thin enough, the Fe atoms which diffuse into the Si layers from the Fe–Si layers on both sides of the

Si layers will pair up. The paramagnetic Fe atoms residing in the dead layers magnetically interact through the Fe atoms in the Si layers (Kazama and Fujimori 1983). This reduces the number of paramagnetic Fe atoms at the interfaces. The thickness of the equivalent dead layers is decreased accordingly, and M_s is increased. The Fe fraction which diffuses into the Si layers increases with decreasing thickness of the Si layers. So the exchange interaction increases and M_s increases monotonically.

According to the above analysis, the magnetization $M_s(d_s)$ of a CMF is given by

$$M_s(d_s) = M_0[1 - d_D(d_s)/d_m] \tag{2}$$

where M_0 is the magnetization of amorphous bulk Fe–Si alloy, d_m is the thickness of the Fe–Si layers (in our case, $d_m = 17$ Å) and $d_p(d_s)$ is the thickness of the equivalent dead layers when the thickness of the Si layers is d_s. One can assume the following relation between $d_D(d_s)$ and d_s:

$$d_D(d_s) = d_D(\infty)\{1 - \exp[-\alpha(d_s - t)]\} \tag{3}$$

where $d_D(\infty)$ is the thickness of the equivalent dead layers when $d_s \rightarrow \infty$, α is a constant and t is the thickness of the Si layers at which the interfacial zone has disappeared (it is approximately the separation between two nearest planes of Fe–Si atoms). Equation (3) indicates that the Fe atoms which are introduced into the Si layers by the sputtering process are exponentially distributed with respect to depth in the Si layers. We obtain from equations (2) and (3)

$$M_s(d_s) = M_s(\infty) + M_D \exp[-\alpha(d_s - t)] \tag{4}$$

where $M_s(\infty) = M_0[1 - d_D(\infty)/d_m]$ is the magnetization of the CMF with $d_s \rightarrow \infty$, $M_D = M_0 - M_s(\infty)$. The result of fitting equation (4) with experimental data is shown in figure 3, where we used $\alpha = 0.1$, $M_0 = 180$ A m^2 kg^{-1} and $M_s(\infty) = 105$ A m^2 kg^{-1} (from experiments), and $t = 3$ Å (d_s and t are in ångströms). The experimental data are shown as full circles, and the theoretical curve from equation (4) as a full solid curve. They are in good agreement except for at two points with minimum d_s at which the experimental M_s are larger than the theoretical values. This is because we did not consider the magnetization coupling effect between magnetic layers in equation (4).

Figure 3 shows that the variation of the Curie temperature T_C is different from that of M_s. When $d_s < 30$ Å, the experimental data for T_C satisfy a linear relation

$$T_C(d_s) = T_C(0) - T_C(d_s) = kd_s \tag{5}$$

where $T_C(0)$ is the Curie temperature of bulk Fe–Si alloy and k is a constant. The difference between T_C and M_s versus d_s reflects the different mechanisms causing their behaviours. The dead layers influence M_s for the CMF, but not T_C because the short-range exchange interaction of interface atoms can only change the fraction of ferromagnetic layers in the CMF but does not increase the intensity of exchange interaction in the ferromagnetic layers, and T_C reflects the disruption of the strongest exchange interaction in the magnetic ions. The main influence on T_C is the magnetic coupling between magnetic layers, which increases with decreasing d_s. The mechanism of magnetic coupling of interlayers has not been understood until now.

The diffusion effect of interlayers while temperature is increasing is another factor which influences T_C. For the samples with thicker Si layers, the atomic interdiffusion of Fe–Si and Si layers on increasing the temperature will cause the number of Fe atoms in the magnetic layers to decrease, the number of Si atoms to increase and the average internal field to be reduced; so the Curie temperature decreases. For the samples with thinner Si layers, the Fe atoms which diffuse into Si layers from both sides of the Si layer during the temperature increase will pair up. This increases the exchange coupling effect of interlayers, and the number of Fe atoms in the magnetic layers does not change much. In this case, the diffusion effect favours ferromagnetism (Kazama and Fujimori 1983), and so T_C tends to increase. The influence of diffusion effects perhaps is not serious because of the low diffusivities of the Fe and Si atoms (Bruson *et al* 1985).

4. Summary

On varying the thickness of Si layers for Fe–Si/Si CMFs, the coupling degree of magnetic layers can be changed, and the properties of films will also change. If the thin Fe–Si layers are fully isolated from each other, the CMF shows a low saturation magnetization, low-hysteresis effect and low Curie temperature. These are the characteristics of two-dimensional ferromagnetism. When the thickness of Si layers is decreased, the coupling of interlayers is strengthened and the dead layers are thinned. The results are that the magnetization and the Curie temperature increase, the low-hysteresis effect disappears, and perpendicular anisotropy appears and becomes stronger.

Acknowledgments

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