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Magnetic and structural properties of Co/Cr multilayers with in-plane anisotropy

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Previously we studied the effect of Cr spacer layer on in-plane coercivity in Co/Cr multilayered films. It was found that the high coercivity is related to the easy axis in-plane orientation and the character of the composition modulation. In this paper, the dependence of the saturation magnetization as a function of Co layer thickness is used to study the interface structure. The layer structure is confirmed by small angle x-ray diffractometry for typical films. The magnetic interaction among the grains is studied by examining deviation from the behavior predicted by the Wohlfarth model for noninteracting single-domain particles.

INTRODUCTION

Magnetic multilayered films prepared by sputtering usually have diffuse interfaces and polycrystalline or amorphous individual layers. The extent of diffusion is different for different preparation conditions. This diffusion, along with other aspects, has a strong effect on the magnetic properties. For example a decrease in magnetization of the magnetic layers as they become increasingly thin is commonly observed.¹ It is known that the CoCr films consist of columnar grains² and the nature of the columnar grains depends on the concentration and the deposition conditions. For multilayers, a complete control of the grain size in the film normal direction and fairly good control in the other two directions can be obtained by adjusting the deposition parameters. For both CoCr film and Co/Cr multilayers previous work^{2,3} has shown that the magnetic properties have characteristics of weakly interacting particles. In this work we studied the structural and magnetic aspects for Co/Cr multilayered films and found that the interface effect can be very large under certain deposition conditions. The understanding of these relationships may serve as a guideline to prepare magnetic films with specified properties for applications.

FILM PREPARATION AND MEASUREMENTS

Co/Cr multilayered films were prepared in a multiple-gun dc sputtering system. Films were deposited on both copper and glass substrates which were mounted on a temperature-controlled resistive heater. Substrate temperature (T_s) values between 20 and 500 °C were investigated and it was found that coercivity (H_c) was maximized for $T_s = 400$ °C. The base pressure was below 5×10^{-7} Torr and the argon pressure during sputtering was 10 mTorr. The thickness of the Cr underlayer was kept at 4000 Å while the total Co thickness was kept at 600 Å. Individual layer thicknesses of Co/Cr multilayers were controlled by programming the time that the substrate was stationary above the corresponding target. The crystal structure, texture, and the layer structure were studied by x-ray diffractometry on a Rigaku DMAXB system. Vibrating sample magnetometry was used to measure the magnetic proper-

ties at room temperature, with a maximum field of 16 kOe. A SQUID magnetometer was used to measure the high-field (50 kOe) and/or low-temperature properties.

RESULTS AND DISCUSSION

Structural properties

Typical small-angle x-ray diffraction (XRD) data are shown in Fig. 1 for films with different bilayer thicknesses. Only a weak first-order peak is observed. This confirms the layered structure and indicates rather diffuse Co/Cr interfaces and a sinusoidal type of composition distribution. Large-angle XRD data in Fig. 2 show the Cr(110) and Cr(200) peaks which are mainly from the thick Cr underlayer and two or three peaks from Co layers. Comparing with the standard powder XRD pattern, we know that the Cr underlayer is bcc and oriented with (110) plane in the film plane basically, and the Co layer is hcp and oriented with c axis parallel to the film plane but tipped out a little. It is noticed that the Co peaks move towards lower angle with decreasing Co layer thickness, indicating larger lattice parameter. This change is not likely due to the Cr substitutions in hcp Co sites because Cr has a smaller lattice parameter. It is suggested that the epitaxial growth and defects within each grain cause this lattice parameter increase. The Co(110) and Cr(100) orientations are optimized at $T_s = 400$ °C. The grain size is likely in the single-domain particle range.²⁻⁴

Magnetic properties

Figure 3 shows the hysteresis loops for a typical film with Co 200 Å/Cr 50 Å, 3 bilayers total, 4000 Å Cr underlayer, deposited at $T_s = 400$ °C, and measured at different conditions. From Fig. 3(a) the following are observed. The in-plane coercivity is as high as 1700 Oe. The saturation magnetization (σ_s) of 85 emu/g is much lower than the bulk Co value of 161 emu/g at room temperature. Here σ_s is defined as the magnetic moment per gram of Co. The switching field distribution ($SDF_h = \Delta H/H_c$) is about 0.38, and the squareness (S) is 0.74. This S is larger than 0.637, the value for isolated uniaxial single-domain particles from the Stoner-Wohlfarth Model in two-dimensional case,⁵ but

FIG. 1. Small-angle x-ray diffraction (XRD) patterns for the films with different bilayer thicknesses.

far below 1.0. In Ref. 3 we pointed out that the main reason for these H_c and σ_s changes compared with pure Co is the significant interatomic diffusion. We will discuss the extent of this effect later. Figure 3(b) gives the remanence loop and the switching field distribution from it, $SFD_r = 0.30$. Figure 3(c) shows the loops in parallel and perpendicular directions measured at room temperature and with the maximum field of 50 kOe. The saturation or knee field $H_k = 20$ kOe in the perpendicular direction. Since in this case

$$H_k = (2K_{\text{eff}}/M_s) + 4\pi M_s \quad (1)$$

this suggests a value of $K_{\text{eff}} = 4.0 \times 10^6$ erg/cc, which is similar to the value of single crystal Co. So the magnetic easy-axis is intrinsically in the film plane and presumably randomly oriented. If measured at low temperature H_k does not change much, as shown in Fig. 3(d). The σ_s decreases by 12% when the measuring temperature is raised from 5 K to room temperature. In the bulk Co case this number is only about 0.9%. To understand this large

FIG. 2. Large-angle x-ray diffraction (XRD) patterns for the films with different Co layer thickness. The films have a total Co layer thickness of 600 Å.

FIG. 3. Hysteresis loops for a film with Co 200 Å/Cr 50 Å, 3 bilayers, on a 4000 Å Cr underlayer. The maximum field used was 50 kOe.

moment loss, σ_s versus temperature (T) for a number of films with different Co layer thicknesses is shown in Fig. 4. The films with thinner Co layers tend to have lower Curie temperature (T_c). For the Co layer thickness of 20 Å, the film is very weakly magnetic (curve 5). For the film deposited at $T_s = 20^\circ\text{C}$ as shown by curve 1, the T_c appears much higher. This suggests that the high substrate temperature T_s is the main reason for diffusion.

Figure 5 gives the Co layer thickness dependence of σ_s and H_c for a group of films with a 50 Å Cr, a 4000 Å Cr underlayer, and $T_s = 400^\circ\text{C}$, measured at room temperature. σ_s increases as Co becomes thicker up to 300 Å. This indicates that the Cr diffusion distance in Co is at least half of this value, i.e., 150 Å. The H_c curve peaks at about 150 Å. The film with 20 Å Co shows almost zero magnetization at room temperature but remains ferromagnetic at low temperatures, suggesting that the Cr concentration is around 25–27 at. % in the Co-rich regions.⁶

FIG. 4. Temperature dependence of the saturation magnetization σ_s for nominal Co for various films with applied field in the film plane. Curve 1 is for film with Co 200 Å/Cr 50 Å. Each curve is normalized to its own saturation magnetization at 5 K.

FIG. 5. Co layer thickness dependence of coercivity (H_c), saturation magnetization (σ_s) for Co with applied field in the film plane. The films nominally have Co X Å/Cr 50 Å individual layer thicknesses and 600 Å total for Co layers, 4000 Å Cr underlayer and were deposited on 400 °C substrates.

To understand the magnetization mechanism Kelly *et al.*⁷ introduced an interaction term ΔM based on the Wohlfarth model of noninteracting grains:

$$M_d(H) = M_r(\infty) - 2M_r(H) + \Delta M(H), \quad (2)$$

where H is the field, M_d is the dc demagnetization remanence and M_r is the isothermal remanent magnetization. Figure 6 shows the ΔM vs H measured at room temperature. For the film with 100 Å Co ΔM is positive, suggesting a ferromagnetic coupling among the grains. For the film with 200 Å Co ΔM is always negative, suggesting a magnetostatic and/or antiferromagnetic dominated interaction. For the film with 300 Å Co ΔM changes sign with field. There is a large difference in the magnitude of the ΔM term for the different films. This might suggest an oscillation of the exchange coupling among the grains for various grain sizes and boundary widths for films with different Co layer thicknesses. A detailed study to confirm this conjecture is under way.

SUMMARY

High coercivity is obtained on Co/Cr multilayered films over a wide range of Co and Cr thickness, as grown

FIG. 6. The magnetic field dependence of the interaction term ΔM from Eq. (2) for three films at room temperature.

with a 4000 Å Cr underlayer at $T_s = 400$ °C. It seems that about 10-Å-thick nominal Co layer at each boundary loses ferromagnetism and there is significant Cr diffusion into Co over distance greater than 100 Å. The interfacial atomic diffusion as well as the proper crystal orientation are responsible for the H_c increase. The high deposition temperature, which is required to achieve in-plane anisotropy, greatly enhances the diffusion and decreases the Curie temperature. The interaction among the grains is a complicated function of the Co layer thickness.

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