Materials Research Society Symposium Proc. Vol. 150. 1989 MRS

THE RELATIONSHIP BETWEEN DEPOSITION CONDITIONS, MICROSTRUCTURE AND PROPERTIES OF RE-TM THIN FILMS

J-W. LEE, S-C.N. CHENG, M.H. KRYDER AND D.E. LAUGHLIN
Magnetic Materials Research Group, Carnegie Mellon University, Pittsburgh, PA 15213 USA

ABSTRACT

Both TbCo and TbFe thin films were dc-magnetron sputtered onto carbon-coated Cu grids as well as glass substrates from a three inch mosaic target using a variety of power levels and argon pressures (P_{Ar}). The perpendicular anisotropy (K_u) displays a maximum in the range of 2.5 to 11.5 mTorr of P_{Ar} and decreased with further increases in P_{Ar}. Films deposited at low P_{Ar} (< 11.5 mTorr) exhibit a smooth and featureless microstructure. Lorentz electron microscopy images reveal the magnetic domains in them as black and white dots which are characteristic of perpendicular magnetization. On the other hand, films deposited at higher P_{Ar} (> 11.5 mTorr) exhibit a rougher surface, a high density of microvoids and in-plane magnetic domains with ripple.

A similar study was also made of the effects of deposition power on the microstructure and domains at P_{Ar} = 5 mTorr. The K_u was found to decrease with increasing power. The microstructure at 30, 40 and 50 watts did not reveal any significant difference except that there was a slight increase in surface roughness with increasing power.

INTRODUCTION

Recently, amorphous rare earth (RE)-transition metal (TM) thin films with perpendicular uniaxial anisotropy (K_u) have been introduced as a rewritable magneto-optical recording medium. In spite of years of study by large numbers of researchers understanding of the origin of K_u remains incomplete, although several models have been proposed including pair ordering,[1] local anisotropy,[2] exchange anisotropy,[3] shape anisotropy,[4] and stress anisotropy.[5] We have previously shown that these films exhibit magnetic properties, which are strongly dependent upon deposition parameters such as argon pressure (P_{Ar})[6] and substrate bias voltage.[7] Our investigations led us to the conclusion that columnar microstructure is not the origin of the perpendicular anisotropy.[6]

Recently we have shown that film-to-substrate stress accounts for up to half the K_u in these films[8] but other mechanisms are also clearly active. We showed that K_u depends upon the P_{Ar} used during deposition and exhibits a peak near 10 mTorr. In this paper we show that the peak in K_u correlates with having a dense and featureless microstructure in the film and that higher P_{Ar} produces a columnar microstructure containing voids. It is also shown that deposition power (P) has somewhat weaker effects on the microstructure and K_u. Increasing P produces a slightly rougher film surface, indicative of more columnar structure, and a decreasing K_u.

EXPERIMENTAL

Both TbCo and TbFe thin films were dc-magnetron sputtered from three inch mosaic targets onto carbon-coated Cu grids for direct observation by transmission electron microscopy (TEM). They were also deposited on glass substrates for cross-sectional TEM observation and torque magnetometer measurements. The sputtering was carried out at the following P_{Ar} : 2, 5, 11.5, 20 and 40 mTorr at P = 40 watts, and P = 30, 40 and 50 watts at P_{Ar} = 5 mTorr. The nominal area composition of the targets were $Tb_{30}Fe_{70}$ and $Tb_{30}Co_{70}$. The nominal thickness of the films was 50 nm. TEM was performed using a Philips EM420T analytical electron microscope. The magnetic domain configurations were observed by Fresnel mode imaging in Lorentz electron microscopy (LEM). K_u was measured by a torque magnetometer in magnetic fields up to 14 KOe.

RESULTS

Perpendicular Anisotropy of TbFe and TbCo Thin Films

The measured K_u of TbFe thin films deposited by DC magnetron sputtering at different P_{Ar} are shown in Fig. 1. It is seen that K_u increases as P_{Ar} is increased from 3 to 12 mTorr, peaks, and then decreases as P_{Ar} is further increased. Beyond 20 mTorr the torque curves indicate both perpendicular and in-plane anisotropy components. Similar data were obtained for TbCo.

On the other hand, K_u decreases with increasing P. For example in TbFe films the K_u decreases from 2.7 x 10^6 erg/cm^3 to 2.4 x 10^6 erg/cm^3 as P increases from 40 to 50 watts with P_{Ar} = 5 mTorr. Similarly in the TbCo films K_u decreases from 1.3 x 10^6 erg/cm^3 to 1 x 10^6 erg/cm^3 as P increases from 30 to 40 watts with P_{Ar} = 5 mTorr. Generally the K_u is higher in the TbFe films than in the TbCo films. This is believed to be due to the differences in the magnetostriction of the two alloys.

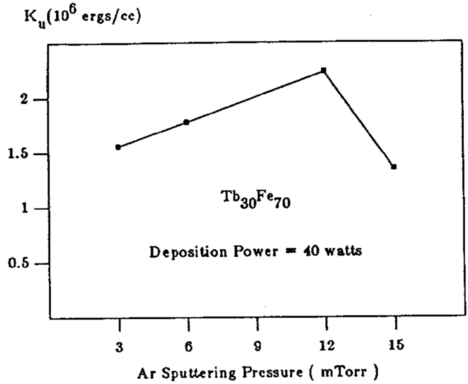

Fig. 1. K_u of TbFe as a function of P_{Ar} .

Microstructure of TbFe and TbCo Thin Films as a Function of P_{Ar}

Figure 1 shows that K_u in TbFe thin films deposited on glass substrates displays a maximum near 12 mTorr and decreases with further increases in P_{Ar}. Here, structural changes are correlated with the changes in K_u. With P_{Ar} = 5 mTorr, the film structure is smooth and featureless, as shown in Fig. 2a. At P_{Ar} = 11.5 mTorr (Fig. 2b), the film morphology is similar to that shown in Fig. 2a. On the other hand, films deposited with P_{Ar} greater than 11.5 mTorr (Figs. 2c and 2d) exhibit a high density of microvoids which we have previously found to be associated with columnar structure.[6] It is noted that the density of microvoids and surface roughness increase with increasing P_{Ar}.

Selected area diffraction (SAD) and microdiffraction patterns of the TbFe films are shown in Fig. 3. At low P_{Ar} (5 mTorr), the SAD pattern shows one broad halo and a weak second halo (Fig. 3a) while at high P_{Ar} (40 mTorr) there is one broad halo and two weak halos (Fig. 3b). The microdiffraction patterns obtained at high P_{Ar} (20 and 40 mTorr) reveal that the first halo contains spots arising from microcrystals. This is consistent with our previous result in TbFeCo thin films[6] where it was shown that microcrystals coexist with an amorphous phase. Increasing P_{Ar} appears to increase the number of microcrystals and/or promote grain growth.

In LEM images at low P_{Ar} (5 mTorr), the magnetic domains appear as black and white dots and stripes which are typical of films with strong perpendicular anisotropy (Fig. 4a). Increasing P_{Ar} (11.5 mTorr) alters the domain structure slightly (Fig. 4b) and still indicates perpendicular anisotropy. At P_{Ar} = 20 mTorr (Fig. 4c), the domain configuration exhibits magnetization ripple as well as black and white dots and stripes. This indicates that the films have a mixture of perpendicular and in-plane magnetization. A further increase in P_{Ar} to 40 mTorr reveals no domain contrast (Fig. 4d). This is due to the reduction in interaction energy between neighboring magnetic portions separated by microvoids[6] and possibly oxidation.

Similarly, microstructure and magnetic domain configurations have been investigated in TbCo films. Low P_{Ar} (2 and 8.6 mTorr) results in films with microstructures that are smooth and featureless (Figs. 5a and b, respectively) while high P_{Ar} (40 mTorr) produces films with microvoids (Fig..5c). This is consistent with our previous results in TbFeCo[6] and TbFe thin films (see Fig. 2). Thus, as far as RE-TM thin films are concerned, high P_{Ar} always produces microvoids.

SAD patterns of TbCo films deposited at low P_{Ar} (2 mTorr) consist of two broad halos (Fig. 6a) while SAD patterns of TbCo films deposited at high P_{Ar} (40 mTorr) exhibit several broad rings as shown in Fig. 6b. A dark field image formed by using the first halo of the SAD pattern in Fig. 6b of the TbCo film deposited at 40 mTorr is shown in Fig. 5d. It is quite evident that the film has a number of microcrystals within an amorphous matrix. The crystallite size is less than 1.5 nm. Thus, high P_{Ar} promotes crystallization in RE-TM films. On the other hand, if microcrystals exist in films with low P_{Ar}, they are substantially smaller than 1.5 nm.

Lorentz electron microscopy images of the magnetic structure in the TbCo films are generally consistent with those of TbFe. At low P_{Ar} (2 mTorr) black and white dots and stripes which are indicative of perpendicular magnetization are seen in Fig. 6c. However, whereas the TbFe film deposited at 40 mTorr had no observable magnetic structure (Fig. 4d), the TbCo film deposited at 40 mTorr (Fig. 6d) reveals magnetization ripple which is indicative of in-plane magnetization.

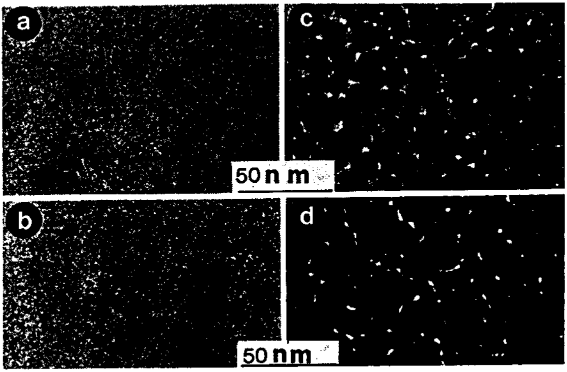

Fig. 2. Bright field TEM images of TbFe thin films as a function of P_{Ar} ; a) 5, b) 11.5, c) 20 and d) 40 mTorr.

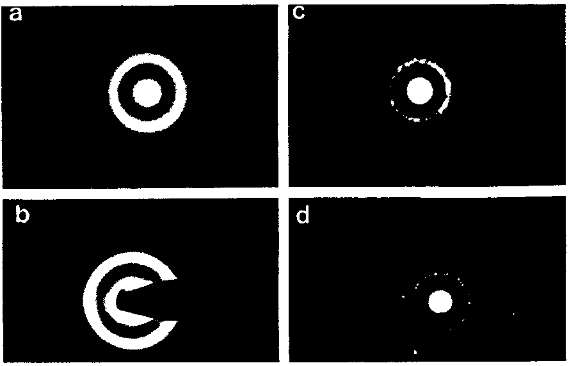

Fig. 3. SAD (a and b) and microdiffraction (c and d) patterns of TbFe films; a) 5, b) 40, c) 20 and d) 40 mTorr.

Microstructure of TbFe and TbCo Thin Films as a Function of Power

Bright field TEM images and representative SAD patterns of TbFe films deposited with P_{Ar} = 5 mTorr and P = 40 and 50 watts are shown in Fig. 7. The films appear to be amorphous and do not reveal any significant difference in morphology. However, there is a slight increase in surface roughness with increasing P (compare Fig. 7a with 7b). Furthermore it should be noted that the SAD pattern at 50 watts (Fig. 7d) has two broad halos while the pattern at 40 watts (Fig. 7c) shows one broad halo with only a weak second halo. The appearance of a more distinct halo at 50 watts suggests that crystallization may be more abundant at 50 watts. LEM images of domain structure reveal black and white dots and stripes which are consistent with perpendicular bubble type domains in both cases.

For TbCo films, there is an obvious difference in surface roughness between films deposited at 30 watts (Fig. 8a) and 50 watts (Fig. 8b) as was also observed in the TbFe thin films. The corresponding microdiffraction patterns reveal that both films (Figs. 8c and 8d) show weak spots within the halo. However, the intensity of spots appears to be stronger at 50 watts. These results suggest once again that the amorphous phase coexists with the microcrystalline phase and that crystallization is more abundant with increasing power. The magnetic domain patterns obtained at 30 watts are shown to be perpendicular stripe type with an average width of less than 1 μm. The domains in the film deposited at 40 watts are larger, but still indictive of perpendicular anisotropy.

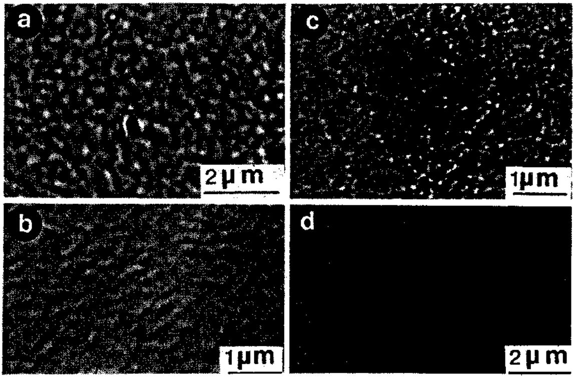

Fig. 4. Lorentz images of TbFe films: a) 5, b) 11.5, c) 20 and d) 40 mTorr.

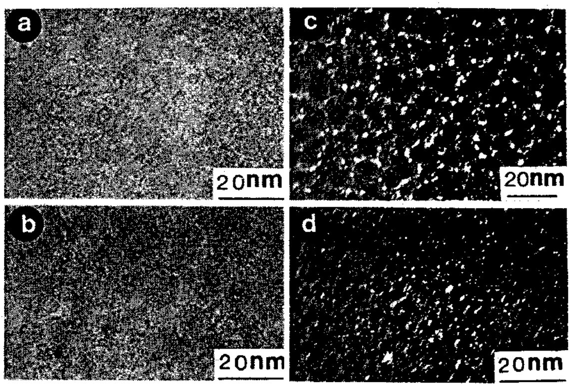

Fig. 5. Bright (a, b and c) and dark (d) field TEM images of TbCo films: a) 2, b) 8.6, c) and d) 40 mTorr.

DISCUSSION AND CONCLUSIONS

The data presented in the previous sections make clear the fact that the sputtering conditions strongly affect the microstructure of rare earth-transition metal alloys and, consequently, their magnetic properties. It was shown that K_u increases with P_{Ar}, reaching a peak near 12 mTorr, and then decreases. Observations of film microstructure reveal that films deposited at low to moderate P_{Ar} (2 to 11.5 mTorr) have smooth and featureless microstructure while films deposited with high P_{Ar} contain microvoids and columnar structure which are associated with a declining perpendicular anisotropy.

Accompanying this change in microstructure and K_u is a change in the magnetic domain structure, as observed by Lorentz electron microscopy. With low P_{Ar} the magnetic domain structure is observed to consist of stripes and bubbles, indicative of strong K_u. As P_{Ar} is increased, the domain structure begins to show evidence of magnetization ripple, which is indicative of in-plane magnetization. In TbFe films at very high P_{Ar} the domain structure vanishes, probably because the film is so porous that it completely oxidizes.

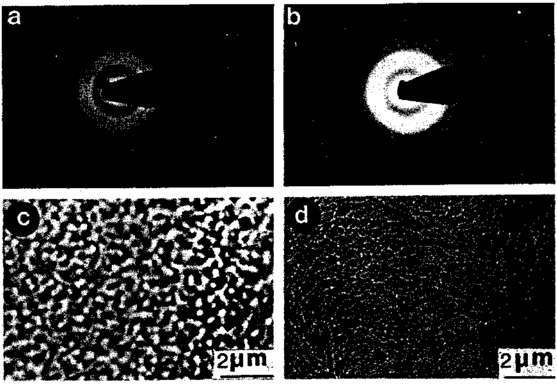

Fig. 6. SAD patterns and Lorentz images of TbCo films: a) and c) 2, and b) and d) 40 mTorr.

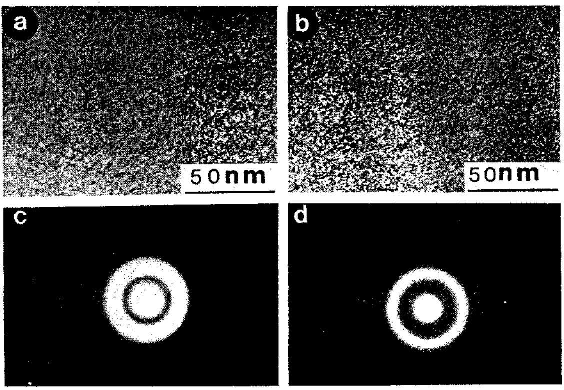

Fig. 7. Bright field TEM images and their representative SAD patterns of TbFe films as a function of P: a) and c) 40, and b) and d) 50 watt

The reason for the change in microstructure with the change in P_{Ar} is believed to be that increasing P_{Ar} induces a reduction in the surface diffusivity of adatoms and causes a more random angle of incidence of sputtered atoms onto the substrate. This in turn leads to self-shadowing and the generation of microvoids.

Studies of the changes in microstructure and anisotropy as a function of P reveal only minor changes in the range of power examined. Increases in P produced minor increases in surface roughness and minor decreases in K_u. Both the values of anisotropy measured and the changes in anisotropy with power of deposition were somewhat larger in TbFe than in TbCo films.

The increases in surface roughness with increasing P are consistent with the zone model of microstructure presented by Thornton [9]. High P leads to Zone 2 microstructure which is columnar and exhibits a rougher surface. Interestingly enough, the data indicate that this type of microstructure causes a decrease in K_u.

It is concluded that there is strong correlation between deposition conditions, microstructure, K_u and magnetic domain structure in DC magnetron sputtered TbFe and TbCo films. Studies indicate that the films which have maximum anisotropy and are preferred for magneto-optic recording applications are deposited with low P_{Ar} and have featureless microstructure.

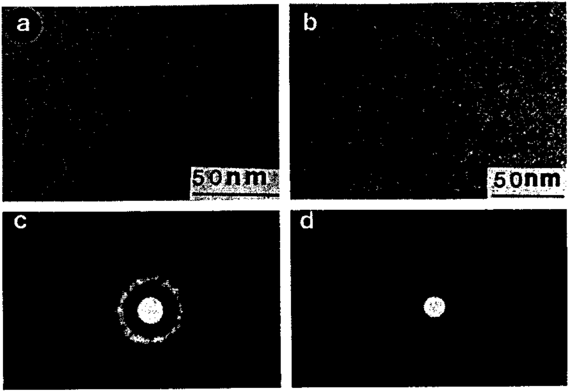

Fig. 8. Bright field TEM images and their representative microdiffraction patterns of TbCo films: a) and c) 30, and b) and d) 50 watt

ACKNOWLEDGEMENTS

This research was funded by the Magnetic Materials Research Group, through the Division of Materials Research, National Science Foundation, under Grant No. DMR-8613386.

1 R.J. Gambino, P. Chaudhari and J.J. Cuomo, *AIP Conf. Proc.* No. 18, 578 (1974).
2 N. Heiman, A. Onton, D.F. Kyser, K. Lee and C.R. Guarniere, *AIP Conf. Proc.* No. 24, 573 (1973).
3 W.H. Meiklejohn, F.E. Luborsky and P.G. Frischmann, *IEEE Trans. Magn.* Mag-23, 2272 (1987).
4 H.J. Leamy and A.G. Dirks, *J. Appl. Phys.* 50, 2871 (1979).
5 H. Takagi, S. Tsunashima and S. Uchiyama, *J. Appl. Phys.* 50, 1642 (1979).
6 J.-W. Lee, H.-P.D. Shieh, M.H. Kryder and D.E. Laughlin, *J. Appl. Phys.* 63, 3624 (1988).
7 H.-P.D. Shieh, M.H. Kryder and J.-W. Lee, *IEEE Trans. Magn.* Mag-24, 2446 (1988).
8 S-C.N. Cheng, M.H. Kryder and M.C.A. Mathur, Paper HQ-03,InterMag 89, Washington, DC, March, 1989, to be published in *IEEE Trans. Mag.*
9 J.A. Thornton, *J. Vac. Sci. Tech.* 11, 666 (1974).