Microstructural investigations of barium ferrite longitudinal thin-film media

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Crystalline barium ferrite films with in-plane magnetic anisotropy have been obtained by postdeposition annealing of amorphous sputtered films. The magnetic properties, the crystallographic texture, and the microstructure are very sensitive to the sputtering and postdeposition annealing conditions. It was found that both H_c and the grain size decrease with the addition of Co and Ti ions. However, an excessive amount of dopants leads to a loss in the in-plane anisotropy. Barium ferrite films produced by this technique contain physical voids or channels separating the grains which help to decrease the exchange coupling between grains.

I. INTRODUCTION

Barium ferrite has been demonstrated, in the particulate form, to have excellent magnetic and recording characteristics for flexible media such as tape or flexible disks.[1–4] Recently, there has been interest in using thin-film barium ferrite for rigid disk media. The mechanical hardness and inert nature of barium ferrite ensure a long media lifetime and may enable a low flying height with no overcoat. Barium ferrite films sputtered under normal deposition conditions are amorphous in nature. Hence, crystalline barium ferrite films have to be obtained by either applying *in situ* substrate heating during deposition or through a postdeposition annealing process. By using the first technique, previous investigators have examined its usefulness as perpendicular rigid disk media.[5,6] Our recent works have explored the possibilities of utilizing it as thin-film vertical and longitudinal media by employing the latter approach.[7,8] In this study, we examined barium ferrite thin films with suitable magnetic properties and microstructures for high areal density recording. As in the case of flexible media, the in-plane coercivity H_c is adjusted by the incorporation of Co and Ti. The microstructure is controlled by the chemical composition, the sputtering, and the annealing conditions. The interplay between these entities is reported in this article.

II. EXPERIMENTAL PROCEDURE

Amorphous multilayer barium ferrite films were rf-diode sputtered onto thermally oxidized Si substrates in a Leybold Heraus Z-400 sputtering system. Two targets were used: $BaFe_{12}O_{19}$ and $Ba_{1.03}Co_{0.85}Ti_{1.17}Fe_{9.86}O_{19}$. Multilayer films with alternating layers sputtered from these two targets were made and the amount of Co and Ti in the films was adjusted by controlling the thicknesses of the respective layers. The total film thickness was between 130 and 200 nm. A forward sputtering power of 50 W was applied. A total sputtering gas (Ar/O_2) pressure of 5 mTorr with an O_2 partial pressure of 0.6 mTorr was used. The deposited films were first annealed in air at 600 °C to allow for composition homogenization within the film and this was followed by an annealing at 820 °C to induce crystallization. The annealed films were examined in plan view and cross section by transmission electron microscopy.

TABLE I. Chemical composition and magnetic characteristics of films A, B, and C.

Film	A	B	C
Chem. comp.	$BaFe_{12}O_{19}$	$Ba_{0.9}Co_{0.36}Ti_{0.5}Fe_{11}O_{19}$	$Ba_{0.78}Co_{0.85}Ti_{1.17}Fe_{9.86}O_{19}$
H_c (Oe)	4066	2201	758
M_s (emu)/cm^3)	340	340	170
S	0.63	0.62	0.39
S^*	0.9	0.88	0.45

III. RESULTS AND DISCUSSION

The magnetic properties and the chemical composition of films A, B, and C are given in Table I. H_c of film A is similar to that found in particulate media[1,2] but the saturation magnetization M_s is slightly lower than the bulk value of 380 emu/cm^3. In film B, H_c falls with the addition of Co and Ti, which is most likely due to a decrease in the crystalline anisotropy.[9,10] The excessive amount of dopants in film C causes a sharp drop in M_s and also leads to a realignment of the magnetic easy axis from the crystallographic c axis into the hexagonal basal plane.[10] Hence, the easy axes are no longer in plane but are randomly distributed and this causes a drop in coercivity and S^*. It should be noted that the barium content in both film B and film C is less than in stoichiometric barium ferrite.

The x-ray spectra of these three films along with another film D, which was deposited with rf magnetron sputtering

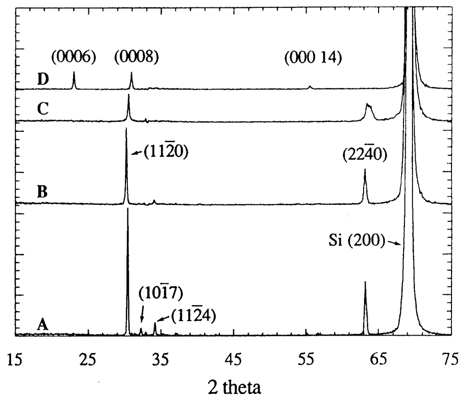

FIG. 1. X-ray diffraction spectra of films A–D.

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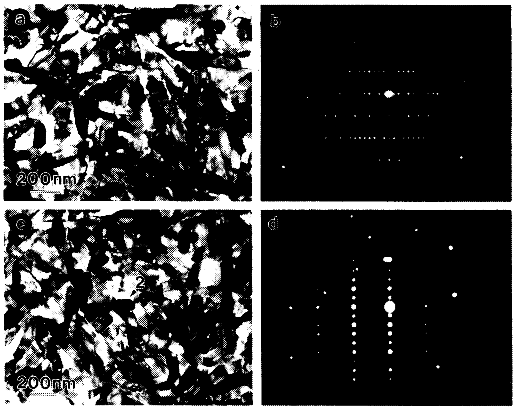

FIG. 2. Pure barium ferrite thin film: (a) and (c) plan-view bright-field (BF) images. (b) $[11\bar{2}0]$ and (c) $[10\bar{1}0]$ microdiffraction patterns of platelet 1 and grain 2 showing the in-plane easy-axis alignment.

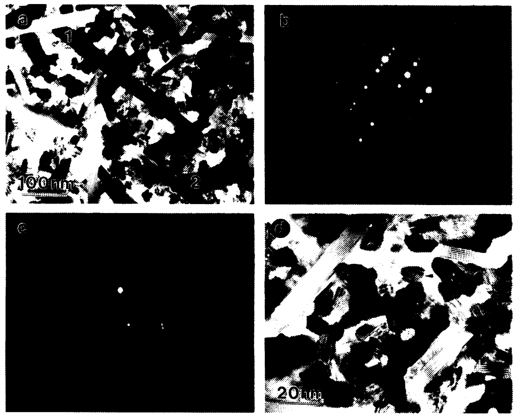

FIG. 4. Co-Ti barium ferrite thin film: (a) and (d) plan-view BF images; (b) $[10\bar{1}0]$ and (c) $[11\bar{2}0]$ microdiffraction patterns of platelets 1 and 2 showing the in-plane easy-axis alignment.

and has a c axis perpendicular to the plane texture,[7] are shown in Fig. 1. A strong c-axis in-plane texture can be found in films A, B, and C. They were annealed at 600 °C for 10 h and then at 820 °C for 3 h. The degree of in-plane texture appears to be strengthened by the addition of Co and Ti, as indicated by the diminishing intensity ratio between $(11\bar{2}0)$:$(10\bar{1}7)$ and $(11\bar{2}0)$:$(11\bar{2}4)$ peaks.

The microstructures of films A, B, and C are shown in Figs. 2–4, respectively. The acicular grains are barium ferrite platelets observed from the edge[7] and they can be found readily in all the c-axis in-plane textured specimens. The majority of such grains in the films studied have their $[11\bar{2}0]$ zone axis perpendicular to the plane of the film which means the c axis is in the film plane. Film A, which lacks doping, consists mainly of such acicular grains and the average plate length is approximately 380 nm with an aspect ratio of about 6.8 [Figs. 2(a) and 2(c)]. Figures 2(b) and 2(d) are microdiffraction patterns of platelet 1 and grain 2. The $[11\bar{2}0]$ and $[10\bar{1}0]$ zone axes reveal the c-axis in-plane alignment of these grains.

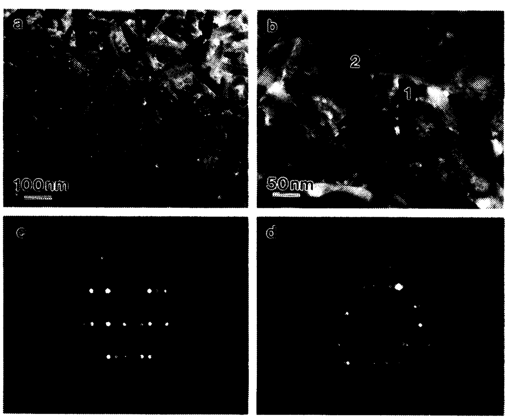

FIG. 3. Co-Ti barium ferrite thin film (multilayer): (a) and (b) plan-view BF images; (c) $[10\bar{1}0]$ and (d) $[11\bar{2}0]$ microdiffraction patterns of platelets 1 and 2 showing the in-plane easy-axis alignment.

The incorporation of Co and Ti ions resulted in smaller grain size. In film B, the average plate length has been reduced to 150 nm with an aspect ratio of nearly 5 (Fig. 3). This decrease in aspect ratio should have a small positive effect on H_c since the c axis is perpendicular to the longer axis of these plates. The finer grain size is advantageous for increasing the signal-to-noise ratio in recording media. In film C, the high quantity of dopants leads to a reduction in the number of acicular grains and gives rise to a much finer grain size (Fig. 4). In addition to the acicular grains, an array of fine equiaxed grains with an average grain size of 16 nm can be seen. Fine defect structures [arrow, Fig. 4(d)] resembling stacking faults being observed edge on can be found within some of the grains. The stacking faults probably arise from the misstacking of the oxygen ions closest-packed planes due to excessive Co and Ti ions and a deficiency of Ba ions. The high Co-Ti content can also account for the loss of platelet shape anisotropy usually associated with barium ferrite. The reduction in grain size could be a result of an enhancement in nucleation sites through the addition of Co and Ti ions. The acicular grains can be completely eliminated by altering the annealing conditions as shown in Fig. 5. This film has the same composition as film C but was annealed at 600 °C for 5 h and then at 800 °C for 3 h. Only fine-equiaxed grains similar to those in film C are present.

From the bright-field (BF) and stereographic images, barium ferrite grains in these films are physically separated and the reasons are twofold. First, the inherent platelet morphology of barium ferrite particles does not favor a high packing density and thus results in physical voids between

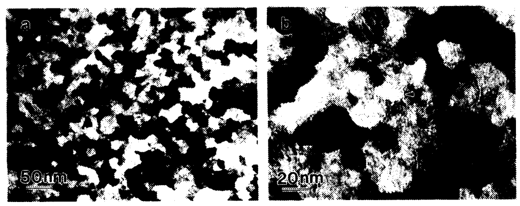

FIG. 5. Co-Ti barium ferrite thin film: (a) and (b) plan-view BF images.

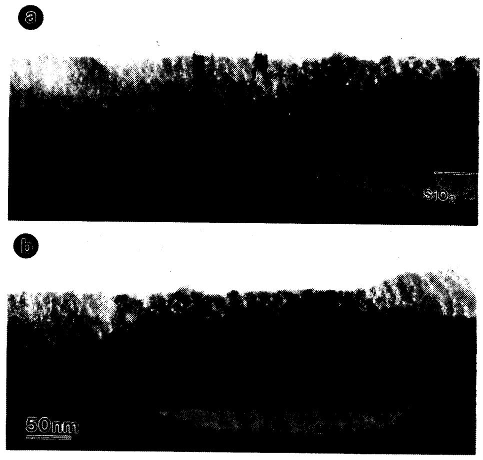

FIG. 6. Co-Ti barium ferrite thin film: (a) and (b) cross-section BF images.

the grains. Second, a volume reduction during the amorphous-to-crystalline phase transformation induces the formation of channels between grains. A cross section of film C is shown in Fig. 6. The rough surface reflects the low packing density, and physical channels can be observed between the grains. Such channels can also be found in Fig. 5(b) (arrow). These voids and channels can account for the slightly low measured M_s value. Moreover, such separation will lead to a reduction in the magnetic coupling between the barium ferrite grains, thus reducing the size of the basic magnetic units and encouraging magnetization reversal by individual particle rotation.

IV. CONCLUSIONS

The crystallographic texture and microstructure of barium ferrite films is very sensitive to the sputtering and postdeposition annealing conditions. The grain morphology and the grain size is controlled by the amount of Co and Ti present in the films and the annealing conditions. Furthermore, H_c can be adjusted by controlling the amount of dopants incorporated into the film. However, excess Co-Ti doping leads to the basal plane becoming a preferred orientation and causes a loss of in-plane anisotropy. Films fabricated with this technique have been found to have physical voids and channels separating the grains which should help decrease the exchange coupling between grains.

ACKNOWLEDGMENTS

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