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# Enhanced coercive and remanence fields for $\text{CoFe}_2\text{O}_4$ and $\text{BaFe}_{12}\text{O}_{19}$ bilayers deposited on (111) MgO

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High quality epitaxial bilayer films of spinel  $\text{CoFe}_2\text{O}_4$  (CoF) and M-type hexagonal ferrite  $\text{BaFe}_{12}\text{O}_{19}$  (BaM) were deposited onto (111) magnesium oxide (MgO) substrate by pulsed laser ablation deposition. X-ray diffraction patterns of both BaM/CoF and CoF/BaM films showed only (0001) BaM, (111) CoF, and (111) MgO peaks. The highest coercive field of  $H_c = 1.4$  kOe was obtained for a BaM/CoF film where the CoF layer was deposited at 400 °C, which was higher than typical  $H_c$  values of  $\sim 0.4$  kOe found for single layer BaM films on (111) MgO. This was less than the  $H_c \sim 3.0$  kOe found for (111) CoF films deposited at 400 °C, probably due to *in situ* annealing effects during the growth of the overlying BaM film at 900 °C. This  $H_c$  enhancement for BaM/CoF bilayers as compared to BaM/MgO films may provide a means to combine large coercive fields with high quality hexaferrite films. © 2002 American Institute of Physics. [DOI: 10.1063/1.1452212]

## I. INTRODUCTION

Thick and highly oriented (0001) films of barium hexaferrite (BaM) were recently deposited by pulsed laser ablation deposition onto (111) magnesium oxide (MgO) substrates.<sup>1</sup> These as-produced films showed coercive fields ( $H_c$ ) of the order of 0.2 to 0.6 kOe. In addition, the hysteresis loop squareness ( $SQ = M_r/M_s$ ) ranged between 0.07 and 0.20, where  $4BM_r$  is the remanent magnetization and  $4BM_s$  is the saturation magnetization. For ferrite devices operating at high frequencies, it is desirable to have ferrites that are self-biased and have high SQ. In addition, narrow ferrimagnetic resonance (FMR) linewidths are required for practical ferrite devices.<sup>2</sup> High SQ usually implies high  $H_c$  in hexaferrites. High  $H_c$  also implies inhomogeneous excitation and, therefore, increased FMR linewidth. Thus, it is important to develop the means to deposit high quality epitaxial hexaferrite films that have both narrow FMR linewidths and high  $H_c$  or SQ for high frequency applications.

The M-type hexagonal ferrite has a crystal structure equal to that of the mineral magnetoplumbite. The magnetoplumbite structure can be viewed as being built up from cubic spinel blocks having [111] orientation (called *S* blocks) and *R* blocks containing the barium ion.<sup>3,4</sup> Cobalt ferrite (CoF) is unique in the spinel family of materials in that it has a positive first order magnetocrystalline anisotropy constant ( $K_1$ ), such that the  $\langle 100 \rangle$  directions are the easy axes for bulk materials. Previous studies on CoF films deposited on (100) and (110) MgO substrates, showed strain effects enhanced the magnetic anisotropy field, such that  $H_c$  was mea-

sured to be between 4.0 and 6.0 kOe.<sup>5-7</sup> In particular, for CoF films growth on (100) MgO substrates it was found that the [100] axis was hard and had relatively high  $H_c$  values for films deposited at 200 °C.<sup>6</sup> In general, the  $\langle 111 \rangle$  axes for bulk CoF are the hard axes, but based on these previous results it may be possible to make the [111] an easy axis if appropriate strains arise from the deposition process. Thus, in this article we are growing BaM/CoF and CoF/BaM bilayers in order to modify the strains. Moreover, we are also attempting to utilize the high  $H_c$  value of the spinel film to magnetically bias the hexaferrite film as a means to increase the hysteresis loop SQ while maintaining a low FMR linewidth profile for the hexaferrite film.

## II. EXPERIMENTAL DETAILS

Phase-pure CoF and BaM targets were used for film deposition onto (111) MgO single crystal substrates using a KrF excimer laser ( $\lambda = 248$  nm). All films were deposited onto  $10 \times 10 \times 0.5$  mm (111) MgO substrates. Bilayer films were deposited for two cases of interest: either BaM was deposited as the top layer (BaM/CoF) or the bottom layer (CoF/BaM) relative to the substrate. In addition, the substrate heater temperature was fixed at either 400 or 800 °C for the CoF buffer layer deposition, but was set at 900 °C for BaM deposition. This choice of CoF deposition temperatures was obtained from previous measurements of CoF films deposited between  $300^\circ\text{C} < T < 900^\circ\text{C}$ , which showed that CoF films deposited at  $T_s \geq 700^\circ\text{C}$  and  $T_s \leq 400^\circ\text{C}$  had more stress than the films at  $T_s = 500$  and  $600^\circ\text{C}$  from their lattice dispersions. The oxygen pressure was 30 mTorr for all

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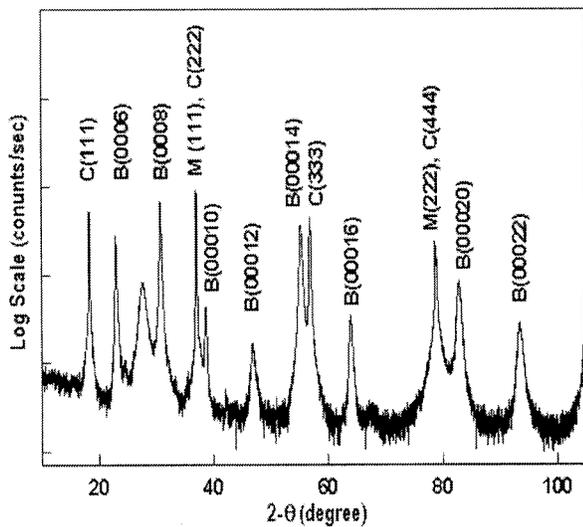


FIG. 1. X-ray diffraction pattern for the BaM/CoF(400°C) bilayer. B, C, and M correspond to the BaM, CoF, and MgO peaks.

depositions. Each CoF film was annealed *in situ* at 900°C for 60 to 90 min at pressure before depositing the BaM overlayer.

The x-ray diffraction (XRD) spectrum was measured using a Rigaku 300 diffractometer for selected films. Vibrating sample magnetometer (VSM) and torque magnetometer measurements were performed in order to analyze the magnetic orientations of the films. The film surface morphology was also measured by a JEOL 6100 scanning electron microscope (SEM).

### III. RESULTS AND DISCUSSION

An XRD pattern for the BaM/CoF (400°C) bilayer film is shown in Fig. 1, where the indexed (0001) and (111) peaks correspond to diffraction from *c*-axis oriented BaM, (111) oriented CoF, and the MgO substrate. A lattice constant of  $a = 8.37 \text{ \AA}$  was deduced for the CoF layer of Fig. 1 by fitting the center of the (111) and (333) peaks to the Bragg's law. Moreover, a lattice constant of  $c = 23.22 \text{ \AA}$  was found for the BaM layer of Fig. 1. These results are close to the bulk lattice parameter values  $a = 8.38 \text{ \AA}$  for CoF and  $c \approx 23.16\text{--}23.24 \text{ \AA}$  for BaM.<sup>3,9,10</sup> The lattice constant of MgO is  $a = 4.21 \text{ \AA}$  while  $a = 8.38 \text{ \AA}$  for the unit cell of CoF spinel, where the spinel unit cell contains 8 formula units. Thus, the room temperature lattice mismatch between CoF and (111) MgO is  $\epsilon_{C/M} = (a_C - a_M)/a_C = -0.003$ . In addition, the lattice mismatches of BaM/CoF and BaM/(111)MgO are  $\epsilon_{B/C} = -0.006$  and  $\epsilon_{B/M} = -0.01$ , respectively, using the bulk value  $a_B = 5.89 \text{ \AA}$ . Here, it is assumed that the close-packed oxygen planes are maintained across the interfaces. Therefore, atoms on the BaM (0001) plane are expected to be less strained on the CoF (111) plane than those of films where the BaM film is deposited directly onto MgO (111). In addition to the lattice mismatch, there are different thermal expansion mismatches between materials. The measured linear thermal expansion coefficient of MgO ( $\alpha = 13.6 \times 10^{-6}/\text{K}$  from 275–1300 K)<sup>8</sup> is greater than that of BaM ( $\alpha_a = 8\text{--}10 \times 10^{-6}/\text{K}$  from 275–1100 K)<sup>9,10</sup> and CoF ( $\alpha = 7.3 \times 10^{-6}/\text{K}$  from 297–

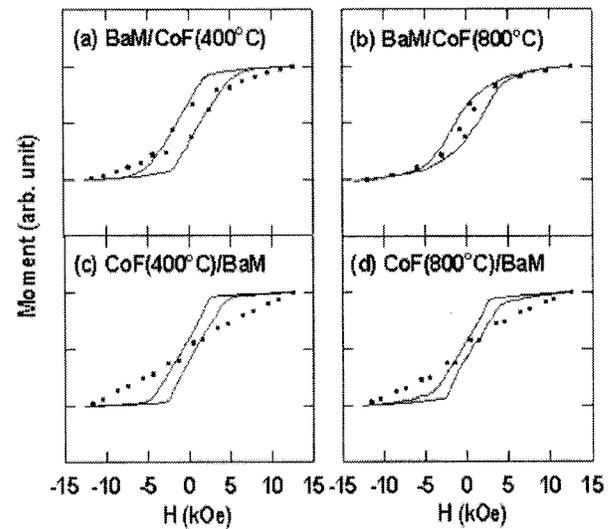


FIG. 2. Hysteresis loops for bilayer films on (111) MgO: perpendicular orientation (solid lines) and in-plane orientation (dashed lines).

940 K),<sup>11</sup> such that the BaM and CoF films should experience compressive stress. However, SEM micrographs of all films, in fact, showed no indication for buckling and few surface irregularities.

Hysteresis loops for bilayer films deposited under each set of conditions are shown in Fig. 2. All films showed uniaxial magnetic anisotropy out of the film plane except for BaM/CoF (800°C), which exhibited no distinct uniaxial anisotropy. The loops also showed the BaM/CoF bilayers had higher  $H_c$  than CoF/BaM bilayers. Table I lists  $H_c$  and SQ values of BaM, CoF, and bilayer films where the external magnetic field was applied perpendicular to the film plane. These results indicate that  $H_c$  was enhanced for BaM/CoF bilayer films compared to BaM films having been deposited under similar conditions. However,  $H_c$  of the BaM/CoF bilayers was 50% lower than  $H_c$  of the single CoF layer film, which was measured to be 3.0 kOe, see Table I. This result implies that the combined effects of the *in situ* annealing and the high deposition temperature used for the overlying BaM film reduced  $H_c$  for these bilayer films. Hysteresis loops of the CoF/BaM bilayers showed their loop behavior was similar to single layer BaM films. Moreover, their  $H_c$  values

TABLE I. Out-of-plane  $H_c$  and SQ for the films on the (111) MgO substrate. Temperatures shown are those of the CoF film deposition.

Films	$H_c$ (kOe)	SQ	$4\pi M_s$ (kG)
BaM	0.2–0.6	0.07–0.20	3.4
CoF at 400°C	2.94	0.37	
CoF at 800°C	0.80	0.13	4.5
BaM/CoF at 400°C	1.40	0.36	4.1
BaM/CoF at 800°C	1.36	0.29	4.3
CoF(at 400°C)/BaM	0.75	0.19	3.8
CoF (at 800°C)/BaM	0.59	0.14	4.1

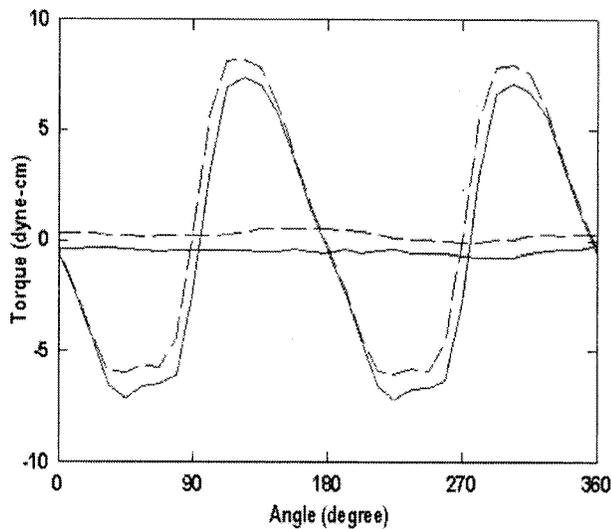


FIG. 3. Out-of-plane and in-plane torque measurements for the BaM/CoF(400 °C) bilayer film. Clockwise (solid lines) and counterclockwise (dashed lines).

were found to be similar to BaM single layer films. Thus, CoF overlayers did not significantly effect the magnetic properties compared to single BaM films.

Saturation magnetization ( $4\pi M_s$ ) values are listed in Table I except for a value for the CoF layer deposited at 400 °C, which could not be saturated. These values were measured from their hysteresis loops by the intercept method or their saturation emu values. Apparently, the net  $4\pi M_s$  values are lower than their bulk values. Indeed, the bulk  $4\pi M_s$  of CoF is near 5.7 kG (Ref. 12) while the bulk  $4\pi M_s$  of BaM is 4.7 kG. Such lowered  $4\pi M_s$  values are commonly observed in the growth of thin BaM films, although bulk values are recovered for thicker films.<sup>1</sup> Thus, it is anticipated that thicker bilayer films will show higher  $4\pi M_s$  values.

Figure 3 shows out-of-plane and in-plane torque magnetometer measurements for the BaM/CoF (400 °C) bilayer film. This result agreed with the VSM hysteresis loop of Fig. 2 in that the easy direction was out of the film plane. However, the torque values ( $L$ ) shown in Fig. 3 were much smaller than the  $L = 60$  dyn cm found for a 1- $\mu$ m-thick BaM film. This result indicated that net anisotropy energy for the BaM/CoF (400 °C) bilayer film is smaller than that of a single BaM film. It is speculated that Co may have diffused into the BaM overlayer during deposition. This may have caused a change in the anisotropy field ( $H_A$ ) and saturation magnetization ( $4\pi M_s$ ) of the BaM overlayer.<sup>3,13</sup>

#### ACKNOWLEDGMENT

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