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

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Abstract
High quality epitaxial bilayer films of spinel CoFe$_2$O$_4$ (CoF) and M-type hexagonal ferrite BaFe$_{12}$O$_{19}$ (BaM) were recently deposited by pulsed laser ablation deposition onto (111) magnesium oxide (MgO) substrates. 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 ($\text{SQ}=M_r/M_s$) 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. 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.

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. 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 ($\text{SQ}=M_r/M_s$) 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. 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. 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 (100) 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 measured to be between 4.0 and 6.0 kOe. 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. In general, the (111) 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 are introduced into the film. 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 10×10×0.5 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 °C<T<900 °C, which showed that CoF films deposited at $T_s \geq 700$ °C and $T_s \leq 400$ °C had more stress than the films at $T_s = 500$ and 600 °C from their lattice dispersions. The oxygen pressure was 30 mTorr for all

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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 \(a = 8.37 \text{ Å} \) 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 \(c = 23.22 \text{ Å} \) was found for the BaM layer of Fig. 1. These results are close to the bulk lattice parameter values \(a = 8.38 \text{ Å} \) for CoF and \(c = 23.16–23.24 \text{ Å} \) for BaM.\(^{3,9,10}\) The lattice constant of MgO is \(a = 4.21 \text{ Å} \) while \(a = 8.38 \text{ Å} \) 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 \(\varepsilon_{CM} = (a_C - a_M)/a_C = -0.003.\) In addition, the lattice mismatches of BaM/CoF and BaM/(111)MgO are \(\varepsilon_{BM} = -0.006\) and \(\varepsilon_{BM} = -0.01,\) respectively, using the bulk value \(a_B = 5.89 \text{ Å} \).

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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>
<thead>
<tr>
<th>Films</th>
<th>(H_c) (kOe)</th>
<th>SQ</th>
<th>(4\pi M_s) (kG)</th>
</tr>
</thead>
<tbody>
<tr>
<td>BaM</td>
<td>0.2–0.6</td>
<td>0.07–0.20</td>
<td>3.4</td>
</tr>
<tr>
<td>CoF at 400 °C</td>
<td>2.94</td>
<td>0.37</td>
<td>4.5</td>
</tr>
<tr>
<td>CoF at 800 °C</td>
<td>0.80</td>
<td>0.13</td>
<td>4.1</td>
</tr>
<tr>
<td>BaM/CoF at 400 °C</td>
<td>1.40</td>
<td>0.36</td>
<td>4.1</td>
</tr>
<tr>
<td>BaM/CoF at 800 °C</td>
<td>1.36</td>
<td>0.29</td>
<td>4.3</td>
</tr>
<tr>
<td>CoF at 400 °C/BaM</td>
<td>0.75</td>
<td>0.19</td>
<td>3.8</td>
</tr>
<tr>
<td>CoF at 800 °C/BaM</td>
<td>0.59</td>
<td>0.14</td>
<td>4.1</td>
</tr>
</tbody>
</table>
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 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.\(^1\) 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-μm-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.\(^3,13\)

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