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The Role of (111)MgO Underlayer in Growth of *c*-axis Oriented Barium Ferrite Films

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Hexagonal barium-ferrite ($BaFe_{12}O_{19}$, magnetoplumbite structure; BaM) film with perpendicularly *c*-axis orientation was successfully deposited on (100) silicon substrates with an MgO (111) underlayer by rf diode sputtering and *in-situ* heating at 920 °C. The magnetic and structural properties of 0.27 μm thick BaM films on MgO (111) underlayers were compared to films of the same thickness deposited onto single-crystal MgO (111) and *c*-plane (000*l*) sapphire (Al_2O_3) substrates by vibrating sample magnetometry (VSM), x-ray diffractometer (XRD), and atomic force microscopy (AFM). The thickness dependence of MgO (111) underlayers on silicon wafer was found to have a large effect on both magnetic and structural properties of the BaM film. The thickness of 15 nm MgO (111) underlayers produced BaM films with almost identical magnetic and structural properties as the single-crystal substrates; this can be explained by the lower surface roughness for thinner underlayer thicknesses. The magnetization saturation (M_s) and the ratio $H_{c\parallel}/H_{c\perp}$ for the BaM film with a 15 nm MgO (111) underlayer is 217 emu/cc and 0.24, respectively. This is similar to the results for the BaM films deposited on the single-crystal MgO (111) and sapphire substrates of 197 emu/cc and 0.10, 200 emu/cc and 0.12, respectively. Therefore, the proposed MgO (111) underlayer can be used in many applications to promote *c*-axis orientation without the cost of expensive substrates.

Key words : Barium ferrite, *c*-axis orientation, MgO underlayer

1. Introduction

There have been many attempts to optimize the magnetic and structural properties of hexagonal barium ferrite ($BaFe_{12}O_{19}$, magnetoplumbite structure, BaM) using various underlayers or single-crystal substrates for magnetic recording media and monolithic microwave integrated circuit (MMIC) applications. The main objective of these investigations is to promote *c*-axis perpendicularly oriented film in either the growth of thick (several μm) films or small magnetic grained (about 10 nm) thin films with high magnetic anisotropy energy to meet above-mentioned applications. Some underlayers used to promote *c*-axis oriented BaM films include Pt, TiO_2, ZnO, and AlN all with varying success [1-6]. However, excellent *c*-axis oriented BaM films, deposited by pulsed laser ablation deposition (PLD) methods, have been shown to grow onto single-crystal MgO (111) and *c*-plane (0001) sapphire (Al_2O_3) substrates [7-9]. If these applications are to be competitive for commercializing application, an easily available substrate and deposition method must be applied. In order to achieve these objectives, BaM films need an underlayer or substrate that can withstand the high temperature annealing required to crystallize the BaM film while minimizing stresses formed from differences in lattice mismatches and a cost effective deposition method.

Many different deposition techniques such as pulsed laser ablation (PLD) [8, 10], evaporation [11], liquid phase epitaxy (LPE) [12, 13], and sputtering have successfully deposited *c*-axis oriented BaM films. The LPE deposition techniques is used to fabricate *c*-axis oriented BaM thick films (50~200 μm) onto single crystal MgO (111) substrates with a ferromagnetic resonance (FMR) linewidth at 60 GHz as low as 27 Oe [13], but this process has restrictions such as unique flux mixtures and a limited substrate selection [14]. The PLD technique has been used to deposit films of similar thickness onto single crystal MgO (111) substrates with a FMR linewidth at 60

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GHz as low as 390 Oe [8]. However the PLD has a shortfall such as a limited deposition area and high power requirement. On the other hand radio frequency (rf) sputtering was selected as the deposition technique for this work due to easy fabrication of targets, low power density, production of large area films, and the ability to use various substrates [3, 15].

The lattice mismatch between BaM and substrate is about 1% for MgO (111) and about 7% for sapphire. For this reason we have decided to deposit an MgO (111) underlayer on silicon (100) substrates. With the development of an MgO (111) underlayer our research group successfully optimized rf sputtering conditions for selective growth of (111)-, (110)-, and (100)-oriented MgO underlayer film on silicon (100) substrate. To our knowledge there have been no attempts to utilize sputtered MgO (111) film as an underlayer to promote *c*-axis orientation perpendicular to the film plane in BaM film.

In this work we have deposited BaM film by rf sputtering onto silicon substrates with an MgO (111) underlayer as a viable means for replacing the expensive substrates required for *c*-axis BaM growth. BaM films with a varying MgO (111) underlayer thickness were compared to BaM films deposited onto single-crystal MgO (111) and sapphire substrates with the same sputtering conditions to promote *c*-axis perpendicular film at a reduced cost.

2. Experiment

The MgO underlayer film was deposited on a naturally oxidized silicon (100) substrate at room temperature using rf reactive sputtering with a working pressure of 27 mTorr and a gas mixture of Ar+50% O_2. The BaM film on the MgO (111) underlayer was deposited by rf reactive sputtering with *in-situ* heating at 920 °C and a working pressure of 5 mTorr and a gas mixture of Ar+20% O_2. Commercially available stoichiomertric $BaFe_{12}O_{19}$ and Mg 8 in. targets (William Advance Materials Inc.) were used to grow the films. Silicon substrates were chosen because they are easily available, cost effective, and can withstand the high temperatures required for *in-situ* heating. Table 1 shows the optimized sputtering conditions for the BaM and MgO (111) underlayer films including substrate temperature and the distance between substrate and target. The thicknesses of MgO (111) underlayers (5, 15, 50, and 100 nm) were examined to determine the effect of underlayer thickness on both magnetic and structural properties of the BaM film. The BaM film thickness was kept constant at 0.27 μm. The as-deposited films were crystallized due to *in-situ* heating, so further post-annealing process did not apply to this work.

The magnetic properties of the BaM film were investigated utilizing torque magnetometer and vibrating sample magnetometer (VSM). The structural properties such as crystallographic characteristics and surface morphology, as well as the effect of MgO (111) underlayer thickness, were characterized with x-ray diffractometer (XRD) and atomic force microscope (AFM).

3. Results and Discussion

To promote *c*-axis orientation of the BaM film, it has been shown that differences in lattice parameters and coefficients of thermal expansion must be minimized. The

Table 1. Sputter deposition conditions for underlayer and BaM films.

Thin Film	Method	Sputtering gas	Substrate temp.	Total gas pressure (mTorr)	Thickness (nm)	Distance of substrate to target (cm)
BaM	rf	Ar + 10% O_2	920 °C	5	270	5
MgO (111)	rf	Ar + 50% O_2	Room temp.	27	5, 15, 50, 100	3.5

Table 2. Magnetic properties of BaM film deposited on various underlayer thickness and substrates.

		Thickness (underlayer) (nm)	R_{ms} underlayer (nm)	$H_{c\parallel}/H_{c\perp}$	$4\pi M_S$ (G)	SQ(⊥)	SQ(∥)
BaM//MgO (111) underlayer/(100) Si		5	–	0.94	2934	0.50	0.24
		15	9	0.15	2772	0.44	0.10
		50	15	0.40	2378	0.43	0.21
		100	25	0.30	1975	0.39	0.27
BaM//single crystal substrate	MgO (111)	–	–	0.10	2476	0.49	0.07
	Sapphire (0001)	–	–	0.12	2515	0.47	0.06
BaM//(100) Si		–	–	0.58	2049	0.54	0.48

selective growth of MgO (111) underlayer was developed onto cost effective oxidized silicon substrates using rf deposition method. It is confirmed by x-ray diffraction pattern that through control of the sputtering conditions, in particular the target to substrate distance, the MgO (111) orientation was successfully controlled regardless of film thickness. It is observed that the MgO (111) underlayer surface roughness decreases with decreasing MgO (111) thickness, which helps with the lattice match and film crystallinity, as shown in Table 2 and Figure 1. It is determined that 15nm thick MgO (111) underlayer has developed to fabricate BaM films with the best magnetic and structural properties, this can explained by the low surface roughness of these films as compared to the thicker underlayers.

BaM film deposited onto a 15 nm thick MgO (111) underlayer are compared to films of the same thickness and sputtering conditions deposited onto single-crystal MgO (111) and sapphire substrates. Figure 2 shows the in- and out-of-plane *MH* loops for the BaM film on single-crystal MgO (111), sapphire substrates, and an MgO (111) underlayer. The magnetic properties of all films are summarized in Table 2. The perpendicular coercivity ($H_{c\perp}$) of the films deposited on an MgO underlayer, MgO substrate, and single-crystal sapphire substrate is almost the same, 1120 Oe, 1177 Oe, and 1058 Oe, respectively. The same result was found for the magnetization saturation (M_s) and the ration of $H_{c\parallel}$ to $H_{c\perp}$ for each film as 217 emu/cc and 0.15, 197 emu/cc and 0.10, 200 emu/cc and 0.12, respectively. It is also noted that films deposited onto bare silicon had a very high in-plane squareness and coercivity indicating random orientation of the film due to the existence of nucleation sites for randomly oriented BaM crystallites in the BaM film above 0.1 μm [3, 16]. The slight difference in the film properties between films deposited on MgO (111) underlayers and those deposited on MgO (111) and sapphire single crystal substrates may be due to the diffusion of Mg into the BaM film during deposition at 920 °C, and the fact that the MgO underlayer is hydroscopic.

The structural properties of the 0.27 μm BaM film deposited onto a 15 nm thick MgO (111) underlayer were characterized by x-ray diffraction pattern (XRD) and atomic force microscope (AFM). The data in Figure 3 shows *c*-axis oriented BaM. The 218 peak explains the coercivity in the in-plane direction of the VSM hysteresis loop in Figure 2. This data is almost identical to the films deposited onto MgO and sapphire single-crystal substrates.

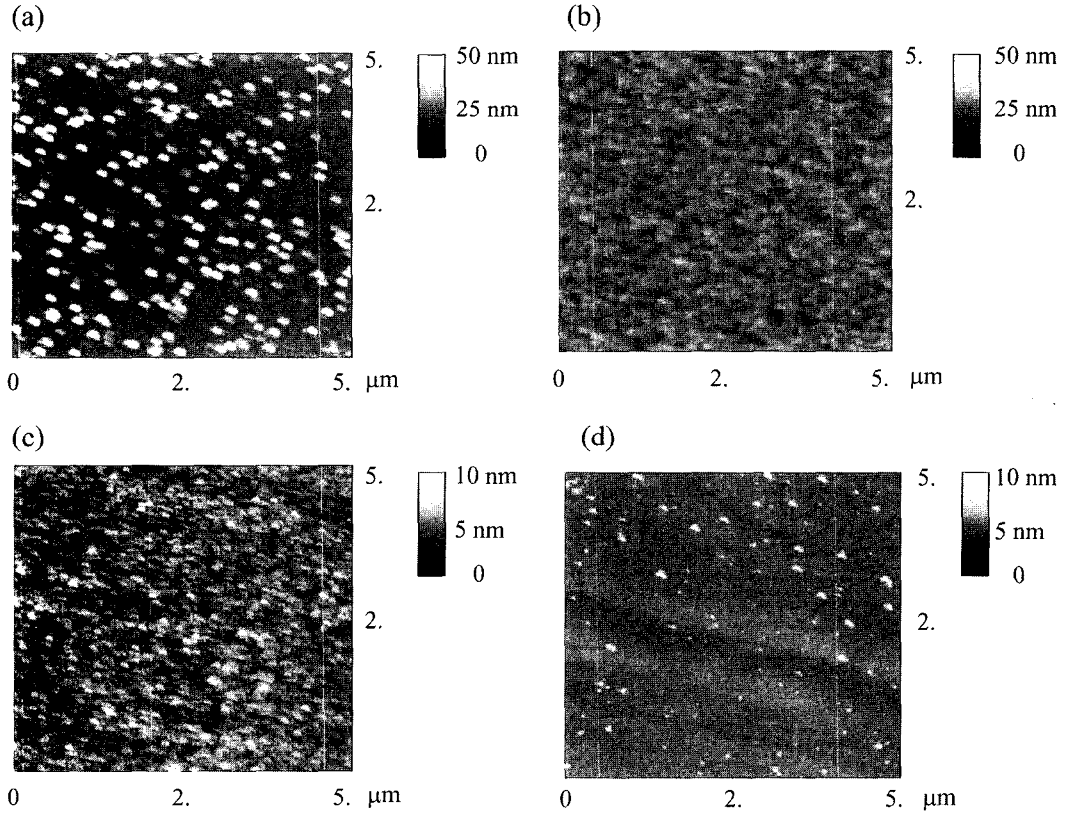

Fig. 1. AFM images of the MgO (111) decreasing surface roughness with decreasing thickness deposited on Si (100) substrates at room temperature; (a) 100 nm; (b) 50 nm; (c) 15 nm; (d) 5 nm.

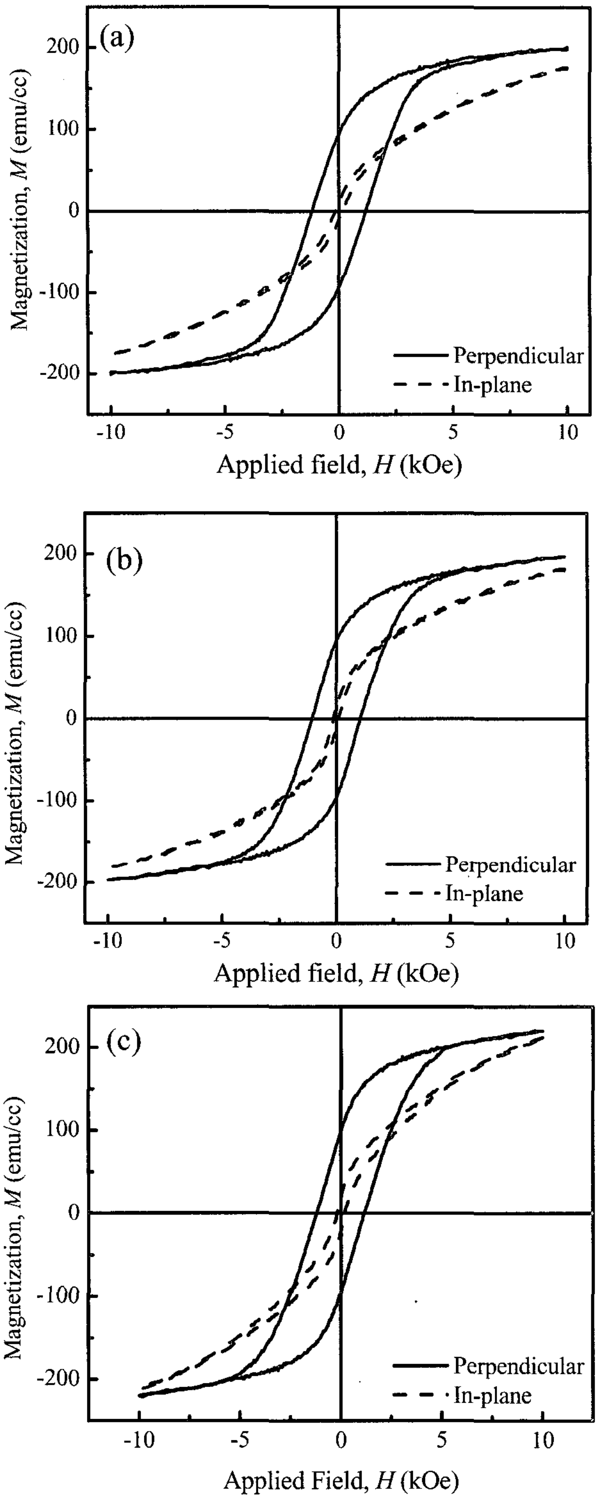

Fig. 2. VSM hysteresis loop of BaM deposited by rf sputtering at 920 °C on (a) sapphire (0001) substrate; (b) MgO (111) substrate; (c) 15nm MgO (111) underlayer on silicon.

Figure 4 shows the AFM image of the BaM film; the columnar structure associated with BaM is shown and is superior to any other film greater than 0.1 μm in thickness deposited on silicon with varying underlayers. This implies that BaM//MgO (111)/Si can compete with expensive substrates and produced BaM film of similar quality with economic cost. By utilizing this underlayer, thick and thin films may be possible, though only 0.27 μm thickness was considered in this study.

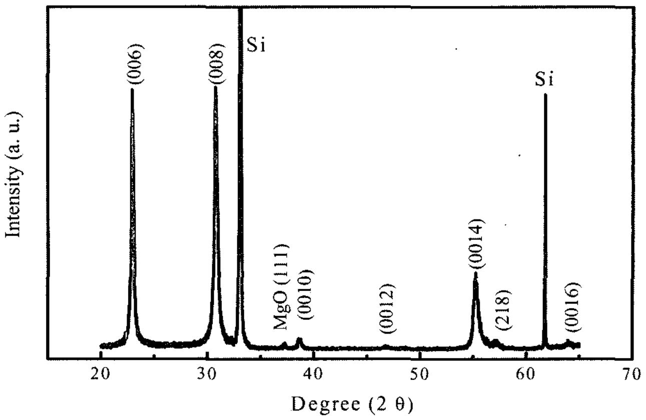

Fig. 3. The X-ray diffraction pattern of a 0.27 μm thick BaM film on 15 nm thick MgO (111) underlayer.

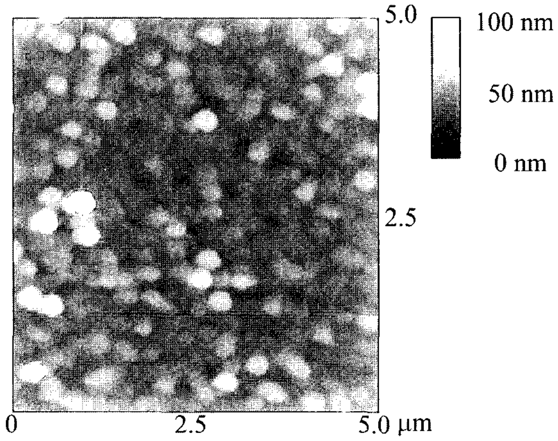

Fig. 4. AFM image of a 0.27 μm thick BaM film on 15 nm thick MgO (111) underlayer.

4. Conclusion

The selective growth of MgO (111) thin film used as an underlayer for BaM to take advantage of the small 1% lattice mismatch between the BaM and MgO (111). BaM film was deposited on (100) silicon substrates with an MgO (111) underlayer by rf reactive sputtering and *in-situ* heating, 920 °C. This film shows magnetic properties similar to BaM film deposited with the same conditions on MgO (111) and sapphire single-crystal substrates. It has studied the thickness dependence of the MgO (111) underlayer and determined that 15 nm thick underlayer films is a prime candidate to promote *c*-axis oriented BaM film growth. This implies that BaM//MgO (111)/Si can compete with expensive substrates and produce BaM film of similar quality at a decreased cost. By utilizing this underlayer, thick and thin films may be possible to fabricate, though only thin films were considered in this study.

Acknowledgement

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