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S. A. Oliver

S. D. Yoon

I Kozulin

M.L.Chen

C. Vittoria Northeastern University

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Growth and characterization of thick oriented barium hexaferrite films on MgO (111) substrates

S. A. Oliver^{a)}

Center for Electromagnetic Research, Northeastern University, Boston, Massachusetts 02115

S. D. Yoon, I. Kozulin,^{b)} M. L. Chen, and C. Vittoria

Department of Electrical and Computer Engineering, Northeastern University, Boston, Massachusetts 02115

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Highly oriented films of BaFe₁₂O₁₉ have been deposited onto MgO (111) substrates by pulsed laser ablation deposition. In contrast to epitaxial $BaFe_{12}O_{19}$ films grown on Al_2O_3 (001) substrates, these films experience an in-plane biaxial compressive stress, and do not crack or delaminate to thicknesses of at least 28 μ m. X-ray diffraction, magnetometry, torque magnetometry, and ferrimagnetic resonance results all indicate excellent c-axis orientation normal to the film plane, and magnetic properties comparable to bulk values. The thickness and properties of these films approach those required for applications in low-loss self-biased nonreciprocal microwave devices. © 2000 American Institute of Physics. [S0003-6951(00)01324-3]

One critical concern during the growth of the thick (>25) μ m) highly oriented films of ferrimagnetic hexaferrite materials that are needed for planar nonreciprocal microwave devices is the control of the deleterious effects of large thermally induced biaxial stresses.^{1,2} Since the stress relationship in epitaxial films is intimately tied to the differences between the lattice parameters and coefficients of thermal expansion of the film and underlying substrate, it is essential that these differences be minimized in order to avoid film cracking and delamination. However, in contrast to the film growth conditions that can be varied systematically, such as the substrate temperature, ambient atmosphere, target stoichiometry, etc., the number of substrates that have physical parameters complementary to the hexaferrites is severely limited due to crystallographic and economic considerations.

The present development of epitaxial hexaferrite films, such as barium hexaferrite (BaFe₁₂O₁₉), has exclusively examined films deposited onto sapphire (Al₂O₃) substrates having various crystallographic orientations.^{2–4} Here sapphire has the rhombohedral (corundum) structure that reasonably well matches the lattice parameters of $BaFe_{12}O_{19}$, which itself consists of a hexagonal close packed structure of oxygen planes, where the magnetic cations are present at interstitial sites. The room temperature lattice mismatch has been estimated at 7% for (001) BaFe₁₂O₁₉ films deposited on (001) Al₂O₃,⁵ where it has been noted that a significant shear strain is also present at the interface due to the mismatch in crystallographic lattices.⁶ More importantly for thick film growth, the coefficient of thermal expansion of (001) Al₂O₃, at $7.80 \times 10^{-6} \circ C^{-1}$ for $0^{\circ} - 927 \circ C^{7}$ is less than that of $BaFe_{12}O_{19}(10 \times 10^{-6} \circ C^{-1})$,⁸ such that the $BaFe_{12}O_{19}$ films sustain significant biaxial tensile stress upon cooling from the deposition temperature of 900-925 °C. These large stresses are manifested through the fracture and delamination of the films, where the delamination is observed to occur either through spalling in the film, spalling in the substrate, or debonding at the interface. In practice, the maximum attainable thickness of $BaFe_{12}O_{19}$ (001) films on Al_2O_3 (001) substrates is less than 20 μ m, which yields films that are too thin for practical application in low-loss microwave devices operating at frequencies below 40 GHz.9

In order to develop the thick oriented hexaferrite films required for microwave devices, we have deposited and characterized the properties of $BaFe_{12}O_{19}$ films grown on (111) MgO substrates by pulsed laser ablation deposition. Here, the ionic MgO crystalline lattice has a face centered cubic structure, such that the MgO (111) crystal plane is close packed and retains the continuity of oxygen planes from the (001) BaFe₁₂O₁₉ film. Thus, from straightforward geometrical considerations it is expected that the BaFe₁₂O₁₉[11 $\overline{2}$ 0] crystal axis will lie collinear to the $[1\overline{1}0]$ axis in the MgO (111) crystal plane. The lattice parameter of MgO is a_{MgO} =4.213 Å,¹⁰ such that a lattice mismatch of $\epsilon = (a_{\text{BaM}})$ $-a_{MgO}/a_{BaM} = -0.01(BaM = BaFe_{12}O_{19})$ is expected between atoms on the MgO (111) plane and those of (001) $BaFe_{12}O_{19} (a_{BaM} = 5.893 \text{ Å}).^{11}$ More importantly, the measured thermal expansion coefficient of MgO (13.6 $\times 10^{-6} \circ C^{-1}$ from 0 to $1000 \circ C)^7$ is greater than that of BaFe₁₂O₁₉, such that the barium hexaferrite film will experience biaxial compressive stress upon cooling. Now the stress relationship for the BaFe₁₂O₁₉/MgO system indicates that the ultimate obtainable film thickness will be governed by the fracture strength of the MgO substrate instead of the BaFe₁₂O₁₉ film.

The barium hexaferrite films characterized here were typically deposited onto 0.5 mm thick (111) MgO substrates by pulsed laser ablation deposition using a KrF excimer laser $(\lambda = 248 \text{ nm})$ at an energy density of 4–5 J/cm² and a repetition rate of 50 Hz. The ambient oxygen pressure was set at 20 mTorr, which corresponds to the optimal pressure found for growth of thick BaFe₁₂O₁₉ films on Al₂O₃ (001) substrates.^{2,12} The substrate temperature was maintained at either 900 or 925 °C, where an in situ halogen lamp was used in addition to a conductive heater in order to maximize the

^{a)}Author to whom correspondence should be addressed; electronic mail: saoliver@neu.edu

^{b)}Present address: Raytheon Systems Company, Sudbury, MA 01776.



FIG. 1. X-ray diffraction pattern obtained from a 1 μm BaFe $_{12}O_{19}$ film. Miller indices are shown for both film and MgO (111) substrate.

temperature of the film surface temperature throughout the deposition. Targets consisted of 2 in. diam pressed powder and sintered BaFe₁₂O₁₉, which were mounted 4 cm from the substrate and rotated and rastered to maximize target surface usage. All films were observed to be dense and void free from cross-sectional electron micrographs, with the film thicknesses ranging from 1 to 28 μ m. Selected films were characterized by x-ray diffraction, magnetometry, torque magnetometry, and ferrimagnetic resonance measurements.

X-ray diffraction measurements were obtained using a Bragg-Bretano powder diffraction system. Figure 1 shows the diffraction pattern obtained on a 1 μ m thick BaFe₁₂O₁₉ film, where the observed peaks have been labeled by the Miller indices for both MgO and BaFe₁₂O₁₉.^{10,11} All of the International Center for Diffraction Data listed $BaFe_{12}O_{19}(00n)$ peaks are present in this pattern, along with the (0,0,22) peak that does not appear in standard databases. A value for the *c*-axis lattice parameter of c = 23.290 Å was found by fitting the centroids of the four tallest (00n) peaks to the Bragg diffraction equation. This value is greater than either the *c*-axis values for bulk $BaFe_{12}O_{19}^{11}$ or for those of films deposited under identical growth conditions onto (00n)Al₂O₃,¹² and is thus suggestive that the *c*-axis lattice parameter is affected by the biaxial compressive stress. However, both oxygen defects and stacking faults may also contribute to this effect. A similarly enlarged c-axis lattice parameter was also found from the diffraction pattern for a 16 μ m film, where this film showed only Al₂O₃(00n) and highly broadened $BaFe_{12}O_{19}(00n)$ peaks.

Excellent magnetic orientation was observed for these films from magnetometry and torque magnetometry measurements. Figure 2 shows the hysteresis loop behaviors for a 27 μ m film where the applied field *H* was oriented either in the film plane (||) or normal to the film surface (\perp). Both results demonstrate the excellent magnetic orientation present in this film, which arises because the magnetic easy axis of BaFe₁₂O₁₉ coincides with the crystallographic *c* axis. Here the normal measurement shows the good square loop behavior expected when the magnetic easy axis lies out of the film plane, where the hysteresis curve is skewed due to the demagnetizing field internal to the film. A mean value for the volume saturation magnetization for these films of



FIG. 2. Hysteresis loops are shown for a 27 μ m BaFe₁₂O₁₉ film with *H* parallel with (||) or normal to (\perp) the film plane.

 $4\pi M_s = 4.2 \pm 0.2$ kOe was obtained from the demagnetizing field values and magnetic moments by averaging over five films having thicknesses above 20 μ m. Meanwhile, the inplane hysteresis curve shows the linear approach to saturation expected for a highly oriented crystal where the uniaxial anisotropy field value H_A exceeds the 12.5 kOe measurement field.

Torque magnetometry results also confirmed the excellent magnetic orientation present for these BaFe₁₂O₁₉ films. Figure 3 shows the magnetic torque versus rotation angle for a 1.7 μ m film where the film is rotated both clockwise and counterclockwise around an axis in the film plane in an applied field of 10 kOe. A distinct uniaxial symmetry is apparent for both curves, where *H* lies normal to the film plane near 10° and 190°, and the difference between rotation directions is apparent at angles near the magnetically hard inplane orientation since *H* has insufficient intensity to align the magnetization into the film hard plane. In contrast, measurements that are taken with *H* rotating in the film plane angle, indicating that there is no preferential component of in-plane anisotropy.

The microwave magnetic properties of thinner films were appraised by ferrimagnetic resonance (FMR) measurements obtained using a shorted waveguide technique at fre-



FIG. 3. Clockwise (solid line) and counterclockwise (dashed line) torque magnetometer results are shown where a 1.7 μ m film is rotated about an axis in the film plane in an applied field of H = 10 kOe. Arrows denote the angle corresponding to film normal (magnetic easy axis).



FIG. 4. A ferrimagnetic resonance spectra taken on a 1.7 μ m film at 54 GHz is shown, where ΔH is the ferrimagnetic linewidth.

quencies from 40 to 60 GHz. Figure 4 shows the differential power absorption dP/dH vs H for a 1.7 μ m film at a frequency of 54 GHz. This resonant absorption shows a FMR linewidth of $\Delta H = 0.70$ kOe, which is considerably larger than the best previous values of $\Delta H = 0.035 - 0.070$ kOe found for BaFe₁₂O₁₉ films on (001)Al₂O₃ substrates.^{3,13} However, the resonant absorption mode for these films is expected to be highly broadened compared to the reference films since these films contain more strains and defects due to their much higher growth rate and much lower ambient oxygen growth pressure, and due to the localized spin pinning and demagnetizing field effects caused by the outgrowths that appear on the thicker films. The uniaxial anisotropy field for this film was found to be $H_A = 16.4$ kOe by fitting the resonance equation to the measured frequency vs resonant field results, where the Lande g factor was taken as 2.00.³ This value for H_A is also lower than the bulk value of $H_A = 17.6$ kOe,¹⁴ but is similar to the values found for films deposited on Al₂O₃(001) substrates.¹²

The usefulness of these $BaFe_{12}O_{19}$ films can be further enhanced through methods such as reducing the FMR linewidth, depositing thicker films, or through cation substitution to modify the magnetic properties. Reductions in the FMR linewidth can be obtained through annealing procedures that refine the crystal structure and remove defects. For example, the FMR linewidth was found to decrease from 0.45 to 0.35 kOe for a 1 μ m film after calcining at 1000 °C for 2 h. However, based upon the previous results found for BaFe₁₂O₁₉ films on Al₂O₃, it may be necessary to grow films at much higher oxygen pressures than those considered here in order to obtain markedly better FMR linewidths.^{3,13}

As noted above, the usage of MgO as a substrate now means that the ultimate usable film thickness is governed by the fracture strength of the substrate. Fortunately, bimetallic strip models of layered structures having comparable thicknesses indicate that the stress internal to the substrate can itself be controlled through the choice of substrate thickness,¹⁵ thus providing a method for forestalling fractures in the substrate that might otherwise hinder the growth of thick $BaFe_{12}O_{19}$ films. Finally, the usefulness of thick $BaFe_{12}O_{19}$ films can be extended to microwave devices operating over many different frequency ranges through compositional adjustment of the film uniaxial anisotropy field through the substitution of nonmagnetic cations for iron within the magnetic lattice.¹⁶

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