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Growth and characterization of 144 \( \mu \)m thick barium ferrite single crystalline film for microwave device application

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Liquid phase epitaxy technique was used to grow 144 \( \mu \)m thick barium ferrite (BaFe\(_{12}\)O\(_{19}\); BaM) single crystalline films on (111) Gd\(_3\)Ga\(_5\)O\(_{12}\) substrate. The growth rate of 72 \( \mu \)m/h was achieved with a flux system of Fe\(_2\)O\(_3\)–BaCO\(_3\)–Na\(_2\)CO\(_3\). The grown BaM films show single crystalline (000\(l\)) orientation that was confirmed by x-ray diffraction and magnetic torque curves. The saturation magnetization (4\(\pi\)Ms) and the anisotropy field (\(H_k\)) were found to be 4.2 kG and 16.0 kOe, respectively. The ferromagnetic resonance linewidth (\(\Delta H\)) at 35 GHz was measured to be 0.1 kOe.


I. INTRODUCTION

Oriented hexagonal barium ferrites (BaM) have been potential candidates for microwave devices in the millimeter wavelength range for the past several decades.\(^1\) The large uniaxial magnetic anisotropy field in these ferrites causes ferrimagnetic resonance (FMR) to occur with a moderate external bias field. In addition, a narrow FMR linewidth (\(\Delta H\)) is desired for millimeter-wave device applications of BaM, which can only be achieved by single crystal ferrite.\(^2\) There has been a great interest in growing BaM thick film by various deposition techniques.\(^3,4\) However, growth of single crystalline film thicker than 100 \(\mu\)m is very challenging. The most successful technique to grow thick BaM film is a flux method known as liquid phase epitaxy (LPE).\(^5\) Deposition of the BaM on (111) MgO substrate by the LPE process resulted in the growth of 22.5 \(\mu\)m thick BaM,\(^6\) indicating that the deposition rate is still too low for circulator application. The maximum attainable growth rate reported for BaM films deposited on \(\alpha\)-Al\(_2\)O\(_3\) (0001) substrate is 100 \(\mu\)m/h.\(^7\) However, the grown BaM films are magnetically in-plane oriented, and also, a seed layer was needed to achieve the above growth rate. On the other hand, the growth rate of BaM film deposited onto Gd\(_3\)Ga\(_5\)O\(_{12}\) (GGG) substrate by the LPE technique was limited to 45 \(\mu\)m/h.\(^5\)

Also, GGG has a cubic structure with a lattice constant of 17.512 \(\AA\) in the (111) direction.\(^8\) However, BaM has a lattice of 5.89 \(\AA\) which is almost 3 times lower than that of the (111) GGG substrate. Therefore, the lattice mismatch between the (111) oriented GGG substrate and the \(\alpha\)-axis corresponding to the 3 unit cells on the basal plane of the BaM is \(\approx\)0.9 %. In addition, the thermal expansion coefficient of GGG and BaM are 9.2 \(\times\) 10\(^{-6}\)/\(^\circ\)C and 10 \(\times\) 10\(^{-6}\)/\(^\circ\)C, respectively. The small thermal expansion coefficient difference between the substrate and the BaM film enables an epitaxial growth and avoids strains and cracking during the LPE growth. Furthermore, it was previously reported that a high growth rate can be expected by employing GGG substrates to deposit thick BaM films when compared to other substrates of MgO or sapphire.\(^9\) In this paper, we report out-of-plane oriented BaM films with a growth rate of 72 \(\mu\)m/h using GGG substrate and a flux system of Fe\(_2\)O\(_3\)–BaCO\(_3\)–Na\(_2\)CO\(_3\).

II. EXPERIMENTAL

Thick BaM films were grown on (111) GGG substrate by LPE technique. BaM flux melt was prepared from a mixture of iron oxide (Fe\(_2\)O\(_3\)), barium carbonate (BaCO\(_3\)), and sodium carbonate (Na\(_2\)CO\(_3\)).\(^9\) The mole ratio of 63.17 (Fe\(_2\)O\(_3\)): 10.53 (BaCO\(_3\)): 26.3 (Na\(_2\)CO\(_3\)) was used for the growth of the BaM thick film. The mixture was ground, and calcined at 980 \(^\circ\)C for 12 h. A platinum (Pt) crucible was heated initially to melt the mixture at an elevated temperature and was replenished with the mixture at regular intervals. Then, the crucible was shifted to a LPE system where it was held at a temperature of 1250 \(^\circ\)C for 10–12 h to homogenize the melt. Prior to dipping the substrate into the melt, the temperature was slowly cooled to the growth temperature ranged from 1162 to 1150 \(^\circ\)C. The substrate was preheated for 20 min at a position of 5-10 mm above the melt in order to reach thermal equilibrium with the solution, and then it was slowly dipped into the melt to allow the growth of the BaM film. The melt was cooled at 4.5 \(^\circ\)C/h during the growth, while maintaining rotation of the substrate at 20–50 rpm to minimize the temperature gradient in the melt. Also, during the LPE growth, the substrate was only partially immersed in the liquid solution. This approach has been chosen because of two main reasons. First, preliminary test experiments showed
that at the interface where the substrate was in contact with the Pt clamp fixture, the substrate always cracked. This occurs because of thermal shocks caused by the difference in thermal conductivities of BaM and Pt. However, when the substrate was partially dipped in the melt, the Pt wires stay above the liquid level and no substrate cracking was observed. Second, the nonimmersed part of the substrate did not exhibit any layer growth. Therefore, the uncovered surface can also be used as a reference plane to measure precisely the layer thickness. Finally, the deposited substrate from the melt was cleaned in a solution of hot dilute nitric acid to remove nonferrite residues.

The thick BaM ferrite films were characterized for their magnetic and physical properties by a vibrating sample magnetometer, x-ray diffraction (XRD), and scanning electron microscopy (SEM). We have used a standard field-swept shorted waveguide technique with an ac field modulation and lock-in detection to characterize microwave properties of the films. The sample is attached on the side wall of a Ka band shorted waveguide and adjusted to the maximum ac magnetic field position. The static magnetic field is applied in the parallel direction to the easy axis of the BaM film, which is the perpendicular direction to the sample film surface. In addition, to minimize the linewidth broadening, the smallest modulation field was chosen in the lock-in technique.

III. RESULTS AND DISCUSSION

After 2 h of LPE growth, the total thickness of BaM film on GGG substrate was measured to be 144 μm by cross-sectional morphology of SEM as shown in Fig. 1. The growth rate of the BaM films on the GGG substrate is 72 μm/h, which is the highest known growth rate achieved by the LPE technique. This higher growth rate, as compared to the previously reported rate of 45 μm/h, may be attributed to the use of a new flux system of Fe₂O₃–BaCO₃–Na₂CO₃ and partial immersion of substrate in the melt.

The crystal structure of the BaM thick film was confirmed by an x-ray diffractometer. The film reflects all the (000l) planes of BaM, which identifies the c-axis orientation of the BaM thick film as shown in Fig. 2. The crystalline orientation was also determined by the magnetic torque curves, at an applied field of 10 kOe. The plane of the disk was rotated both clockwise and counterclockwise in the applied field. As shown in Fig. 3, the film exhibits a uniaxial symmetry when the applied field (H_app) is normal to the film plane near 10° and 190°. This implies that the anisotropy energy is minimum in the easy direction of the magnetization. The curves corresponding to the angles of 100° and 280° show a positive slope, implying hard alignment of magnetization in the in-plane direction at 10 kOe.

As shown in Fig. 4, it is confirmed that the 144 μm thick LPE processed BaM film possesses uniaxial magnetic anisotropy. The saturation magnetization (4πM_s) in the out-of-plane direction was found to be 4.2 kG at room temperature. The in-plane magnetization, however, shows no saturation at a maximum applied field of 10 kOe due to the high crystalline anisotropy field (H_K) of the film. The coercive field of the LPE processed thick film was 3.8 Oe, implying that the films have relatively small defects and voids. The uniaxial anisotropy field for the 144 μm film was estimated by the following equation:

\[ H_{K} = \frac{\pi M_s}{2} \]

FIG. 1. (Color online) SEM showing the cross-sectional area of 144 μm thick LPE processed BaM film on (111) GGG substrate.

FIG. 2. XRD patterns of 144 μm thick BaM film on (111) GGG substrate.

FIG. 3. (Color online) Torque measurement of 144 μm thick BaM film on GGG substrate at H_app=10 kOe.
The estimated demagnetization in the film plane of the crystal is given by

\[ N_n = \frac{H_{\text{sat(easy)}}}{4\pi M_s} = \frac{3900}{4205} = 0.92. \]

The demagnetizing factor \( N_n \) in the easy plane direction of the crystal is obtained by

\[ H_{\text{sat(hard)}} = H_k + 4\pi N_n M_s. \]  

For \( H_{\text{app}} \) along the easy direction, the magnetization is linear in \( H_{\text{app}} \) below saturation, which occurs at \( H_{\text{sat(easy)}} = 3.9 \text{ kOe} \), and \( H_{\text{sat(hard)}} \) is estimated to be 16.0 kOe by extrapolating the out-of-plane and in-plane curves to their intersection point. Assuming a density of 5.29 g/cm\(^3\) for BaM, the saturation magnetization is obtained to be 4205 G. The demagnetizing factor \( N_n \) gives the easy plane direction of the crystal is given by

\[ N_n = \frac{H_{\text{sat(easy)}}}{4\pi M_s} = \frac{3900}{4205} = 0.92. \]

The estimated demagnetization in the film plane of the crystal \( N_n = (1-N_n)/2 = 0.04 \) and \( 4\pi M_s = 4205 \text{ G} \) yield 0.168 kOe of \( 4\pi N_n M_s \). Therefore, \( H_{\text{sat(hard)}} \approx H_k \) by Eq. (1) since \( 4\pi N_n M_s \) is small and negligible. This means that crystalline anisotropy of the 144 \( \mu \text{m} \) thick BaM film is close to 16 kOe of \( H_{\text{sat(hard)}} \) and this film is of very high quality.

With regard to microwave properties, room temperature FMR measurements have been performed on the 144 \( \mu \text{m} \) thick BaM film with a shorted waveguide technique. The FMR condition used for the measurement is

\[ f_r = H_r + H_k - 4\pi M_s, \]

where \( f_r \) is the resonance frequency in gigahertz, \( H_r \) is the resonance external field in kilo-oersted, \( H_k \) is the anisotropy field, and \( \gamma' = \gamma/2\pi = 2.788 \text{ GHz/kOe} \). Figure 5 shows the differential power absorbed \( dP/dH \) with the applied field. A ferrimagnetic resonance linewidth \( (\Delta H) \) of 0.1 kOe at 35 GHz was obtained for 144 \( \mu \text{m} \) thick BaM film from resonance absorption spectra. However, the linewidth of 0.1 kOe obtained is considerably larger than the best previous values reported for the thick BaM films grown on GGG substrates \((\approx 60 \text{ Oe at } 59 \text{ GHz})\). This might be attributed to the extrinsic strains and defects on the film surface caused by higher temperature and also high growth rate of 72 \( \mu \text{m/h} \) during the LPE process. In addition, localized spin pinning and demagnetizing field effects by any outgrowths on the thicker films might have also caused the linewidth broadening. Further reduction in FMR linewidth can be obtained by polishing and/or annealing the samples that enhance the crystal structure and reduce defects on the film and also any strains at the film substrate interface.

**IV. CONCLUSIONS**

Single crystalline BaM films have been successfully grown on (111) oriented GGG substrate by LPE process. These films were \( c \)-axis oriented. The growth rate of the single crystal BaM film was 72 \( \mu \text{m/h} \), which is the highest known growth rate. Accordingly, a total thickness of 144 \( \mu \text{m} \) was achieved in 2 h. Room temperature FMR measurements shows the FMR linewidth \( (\Delta H) \) of 0.1 kOe at 35 GHz.

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