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Microwave and magnetic properties of barium hexaferrite films having the *c*-axis in the film plane by liquid phase epitaxy technique

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We report the growth of thick barium hexaferrite ($BaFe_{12}O_{19}$,BaM) films on *m*-planes ([$1\bar{1}00$] or [$10\bar{1}0$]) sapphire (Al_2O_3) substrates by the liquid phase epitaxy deposition technique. The procedure entailed the deposition of seed layers of $BaFe_{12}O_{19}$ onto the substrate by the pulsed laser ablation deposition and then dipping the substrate into a molten flux for 2 h. The total thickness ranged from 60 μm to 200 μm. The vibrating sample magnetometer (VSM) measurement data showed that the film exhibited magnetic uniaxial anisotropy axis in the film plane. The coercive field was relatively small, range of H_c was from 0.007 kOe to 0.08 kOe, and the saturation magnetization ($4\pi M_s$)≅4.42 kG. The ferrimagnetic resonance (FMR) linewidth (ΔH) at 59.9 GHz was ~0.08 kOe. This value of ΔH should be contrasted with previous FMR linewidth measurement of films deposited by the PLD technique which showed a ΔH of ~1.20 kOe. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1557791]

I. INTRODUCTION

About 30 years ago there was great interest in depositing epitaxial films of hexaferrite materials ($BaFe_{12}O_{19}$, $SrFe_{12}O_{19}$, etc.)[1–3] in order to provide enhanced performance at high frequencies, such as: smaller size devices and compatibility with monolithic microwave integrated circuit (MMIC) devices. In order to achieve these goals the films needed to be relatively thick (>50 μm) and of high quality (low loss tangents).

The deposition of $BaFe_{12}O_{19}$ or BaM films onto sapphire (Al_2O_3) substrates [(0001), ($11\bar{2}0$), and ($1\bar{1}00$) or ($10\bar{1}0$) sapphire (Al_2O_3) substrates][4–7] and (111) magnesium oxide (MgO)[8,9] substrates by the pulsed laser ablation (PLD), sputtering,[10] chemical vapor deposition (CVD),[11] sol–gel method,[12] and liquid phase epitaxial (LPE) deposition technique[1–3,9] have been investigated. It appears that the LPE deposition technique produces the best quality of BaM films. Previous efforts have attempted deposition of films onto (0001) Al_2O_3,[3] (111) MgO,[9] and (0001) hexagallates ($SrGa_{12}O_{19}$, $BaGa_{12}O_{19}$)[2] substrates where the magnetic easy axis lies normal to the film plane, or the *c*-axis is also oriented along the magnetic easy axis. To our knowledge, there have only been limited attempts to deposit thick BaM films with *c*-axis alignment in the film plane. Our efforts in the past produced BaM films as thick as ~15 μm with the *c*-axis in the film plane using the PLD technique.[7] However, the ferrimagnetic resonance (FMR) linewidth was measured to be ~1.2 kOe at 54 GHz.[7] This is an intolerably high FMR linewidth to fabricate ferrite components at high frequencies. Hence, we have changed our deposition technique from PLD to a LPE technique with the purpose of improving the FMR linewidth, since our modified LPE technique has been so successful in producing[9] narrow FMR linewidth materials of BaM films with *c*-axis normal to the film plane. By modified LPE technique we mean that we require seed layers prior to the ''pure'' LPE deposition technique.

We have deposited by the LPE technique thick films of BaM on both surfaces of *m*-plane [($1\bar{1}00$) or ($10\bar{1}0$) plane] sapphire (Al_2O_3) substrates, where the crystallographic *c*-axis (⟨0001⟩ direction, $a = 5.128$ Å and $c = 12.99$ Å)[4] is along the substrate plane. The object is eventually to use these materials in planar microwave devices such as low bias field phase shifters at high frequency (>50 GHz), self-bias phase shifters,[13] and biasing circuits for Y-type hexaferrite material devices.

II. EXPERIMENTAL DETAILS

The BaM seed layers were grown onto *m*-plane, ($1\bar{1}00$) or ($10\bar{1}0$), sapphire (Al_2O_3) substrates using pulsed laser ablation deposition (PLD) technique. Both surfaces of the Al_2O_3 substrates were polished, since seed layers deposition was required onto both surfaces. Total thickness of ~0.2 μm thick was deposited for seed layer onto the front and backside surface of the substrates. Thus, thickness of each seed layer of ~0.1 μm thick was deposited by the PLD technique.[8] After a thin seed layer of BaM was prepared onto the substrate, the substrate was dipped into an isothermal BaM flux melt for liquid phase epitaxy (LPE) deposition for 2 h.[9]

Crystallographic orientation information was obtained from θ–2θ x-ray diffraction (XRD) measurements. In order to measure DC magnetic properties of the BaM films, vibrating sample magnetometer (VSM) measurements were performed. The microwave properties of the BaM films were characterized by the in-plane ferrimagnetic resonance (FMR) measurements between 56 and 60 GHz using a shorted TE_{10} wave guide.

III. RESULTS AND DISCUSSION

X-ray diffraction pattern shows that the film for a 70 μm BaM film have an excellent in-plane ($1\bar{1}00$) orientation, as shown in Fig. 1. The [0001] axes of both the BaM film and substrate are collinear. A lattice spacing in ⟨$1\bar{1}00$⟩ direction

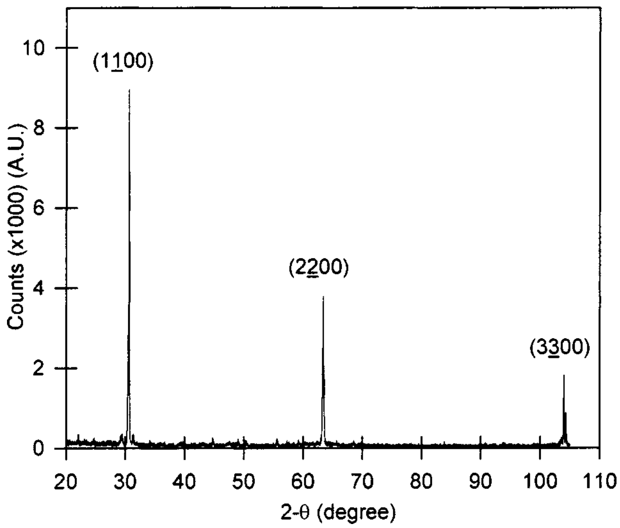

FIG. 1. X-ray diffraction pattern for an ~70 μm LPE BaM film grown on $(1\bar{1}00)$ Al_2O_3 substrate.

of $d_{\langle 1\bar{1}00\rangle}=5.005$ Å was found for the BaM films (thickness of the films $\geqslant 70$ μm) by fitting the centroids of the $(1\bar{1}00)$, $(2\bar{2}00)$, and $(3\bar{3}00)$ reflection peaks. Thus, a lattice constant of $a=5.78$ Å was obtained from $a=d_{\langle 1\bar{1}00\rangle}/\sin(60°)$, which is smaller than the bulk lattice constant of $a=5.89$ Å from literature values.[14] Clearly, the BaM films deposited by us were in compressive stress. This result is quite different from the BaM films deposited in previous researches on (0001) Al_2O_3 substrates, where the BaM films were in tensile stress,[4,15] and the *c*-axis was normal to the film plane. The induced stress may be due to two effects as (1) lattice constant mismatches, ϵ, between the BaM film and substrate. (2) Thermal expansion coefficient mismatches between the BaM film and substrate.[4,6,7,15–17] Both mismatches have been found to be important from previous researches.[15–17] For example, films grown by PLD with *c*-axis in the film plane tended to delaminate for a thickness of $\geqslant 20$ μm whereas films grown now by the modified LPE technique showed no evidence of delamination up to thickness of 70 μm.

The high degree of magnetic easy direction along in-plane orientation is shown in Fig. 2, where the vibrating sample magnetometer (VSM) hysteresis loops for a ~70 μm 1×1 cm^2 size film are shown with the applied field (H) along the in-plane easy axis and normal to the film surface. Figure 2(a) shows a typical hysteresis loop for double-side seed layers of BaM by the PLD technique before dipping into the melt flux. Size of the substrate was 0.5 mm $\times 1$ cm^2. Thickness of each seed layer was ~0.1 μm which was estimated from VSM magnetization measurements. Figure 2(b) shows the hysteresis loops for the BaM films deposited by the modified LPE technique for 2 h. The external field is applied along the film easy axis, or *c*-axis. The coercive field (H_c) was measured to be $0.009\ \text{kOe}\leqslant H_c \leqslant 0.09$ kOe. This value can be compared with the value obtained from Fig. 2(a), where H_c yielded a value of 2.48 kOe. The demagnetization field was measured to be ~60 Oe from the VSM hysteresis. The saturation magnetization ($4\pi M_s$) was measured as determined from $4\pi(\text{EMU})/\Delta\nu$, where $\Delta\nu$ is the volume of the film, and was equal to 4.42 kG at room temperature. VSM hysteresis loops measurements of the magnetic hard axis of BaM films, out-of-plane VSM (dotted line), are shown in Fig. 2. In general hard axis hysteresis loops for the BaM films cannot be saturated at below 13.0 kOe due to the high uniaxial anisotropy field of $H_A \cong 17.4$ kOe. Coercive fields for the out-of-plane VSM were measured to be $0.008\ \text{kOe}\leqslant H_c\leqslant 0.07$ kOe for 70 μm thick films grown by the LPE technique. This should be compared with H_c of 0.1 kOe as measured from 0.2 μm thick seed layer of the BaM film. The seed layer was deposited by the PLD technique, M_s values measured from in-plane VSM hysteresis at 12.5 kOe were larger than the values obtained from the VSM hysteresis measurements for the out-of-plane. This discrepancy was the result of placing the film closer to the measuring coils of the VSM magnets for the case of H field in the film plane, since the size of the film was significant (1×1 cm^2). The coil separation was about 3 cm.

Ferrimagnetic resonance (FMR) spectra were taken with H placed along the film easy axis. The resulting FMR con-

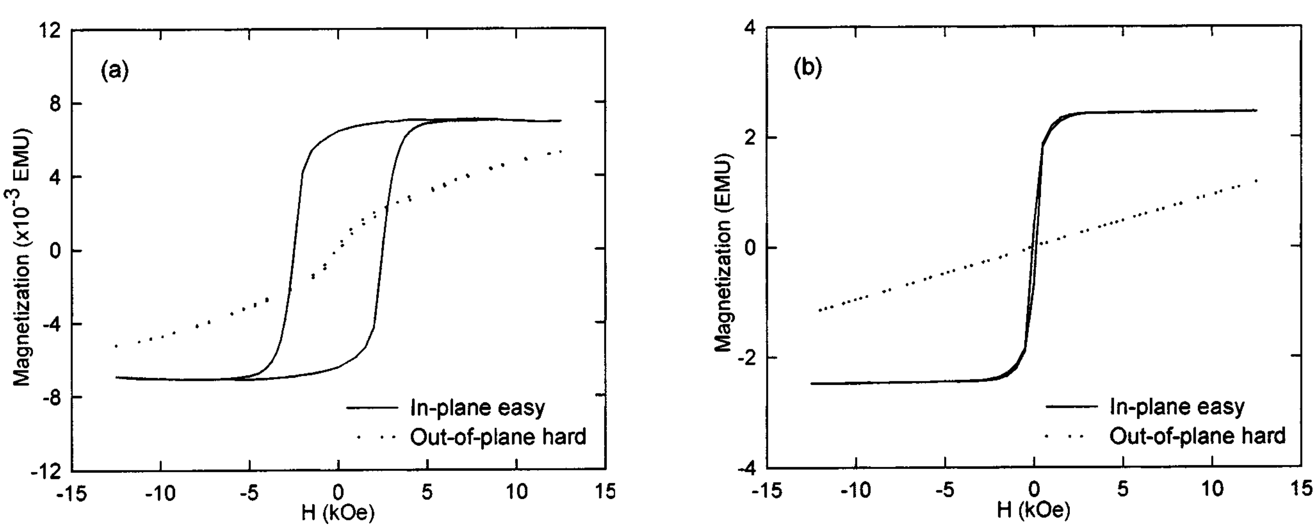

FIG. 2. (a) Hysteresis loops of double side seed layer for a ~0.2 μm PLD BaM film with H along the easy axis and hard axis. (b) Hysteresis loops for an ~70 μm LPE BaM film with H along the easy axis and hard axis.

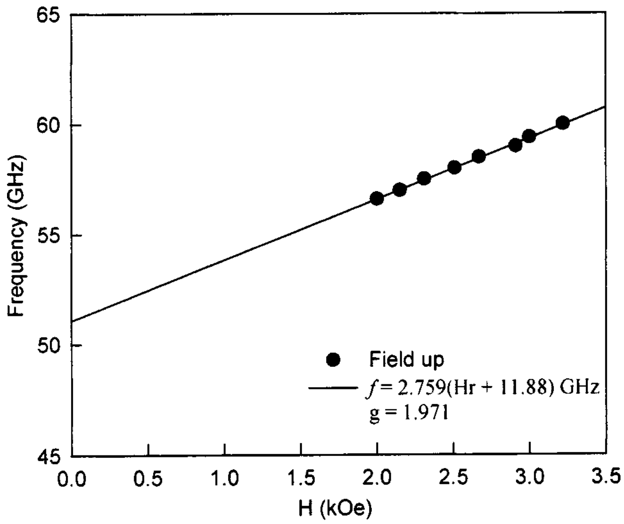

FIG. 3. Ferrimagnetic resonance frequency vs resonance magnetic field.

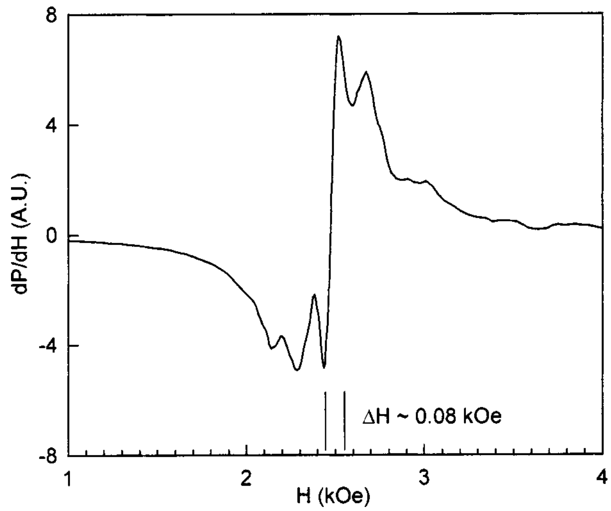

FIG. 4. Ferrimagnetic resonance spectrum at 59.9 GHz.

dition when H and M lie parallel along the film easy axis is then[18]

$$f=\gamma' \sqrt{(H_r+H_A+(N_z-N_x)M_s)(H_r+H_A+(N_y-N_x)/M_s)}, \quad (1)$$

where H_r is the resonant magnetic field, $H_A=2|K_u|/M_s$, $\gamma'=\gamma/2\pi$, and γ is the gyromagnetic ratio. This FMR condition was evaluated by minimization of total magnetic free energy at equilibrium. In Fig. 3, a plot of microwave frequency versus resonant field for the 70 μm thick BaM film is shown. The solid line shows the linear regression of the FMR condition where the fit parameters were $\gamma'=2.759 \pm 0.009$ GHz/kOe, $H_A=17.46\pm 0.03$ kOe, and $4\pi M_s = 4.42\pm 0.02$ kOe. We deduced the g-factor as $g=1.971 \pm 0.006$. It is noted in Fig. 3 that the FMR frequency at 51.09 GHz can be reached at relatively small external field.

Most of the FMR spectra measured in a frequency range from 56 GHz to 60 GHz exhibited Lorentzian shapes with many magnetostatic modes below and above the main FMR mode. The FMR linewidth of $\Delta H \cong 0.08$ kOe at 59.9 GHz, see Fig. 4, was obtained for the main FMR mode. This mode is recognized as the most intense mode and it obeys Eq. (1) very well. Presence of magnetostatic modes in the FMR spectra is a result of the fact that the sample size is finite in the lateral dimensions, since we had to fit the sample into a wave guide. However, for similar materials with larger ΔH,[8] it may not be possible to observe magnetostatic modes, since the resolution between modes is ''washed'' away by the large absorption over frequency and field.

IV. CONCLUSIONS

It is possible to deposit thick layers of $BaFe_{12}O_{19}$ or M-type hexaferrites on m-plane sapphire substrates. Total thickness of 60–200 μm for 2 hours deposition time was achieved of sufficient good quality onto one side of the substrate. Therefore, short deposition times ($\sim$30 μm/1 h) is highly desirable compared to growth of BaM films on (111) magnesium oxide by LPE technique, where growth times were in the order of 6–8 hours. The ferrimagnetic resonance (FMR) linewidth (ΔH) of these films is reasonably narrow and equal to 0.1 kOe. This represents considerable progress in view of the fact that similar films prepared by PLD exhibited FMR ΔH of $\sim$1.2 kOe. Overall these results indicate that the BaM films can be utilized at frequencies $>\sim$51 GHz with low field bias. In addition it is highly possible to grow scandium or aluminum doped m-type hexaferrite films[19] which are magnetic easy in the film plane for operating frequencies from 1 to 100 GHz.

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