In-plane *c*-axis oriented barium ferrite films with self-bias and low microwave loss

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Hybrid pulse laser deposition and liquid phase epitaxy methods have been used to produce in-plane *c*-axis (IPCA) oriented barium ferrite (BaM) films on *a*-plane $(11\bar{2}0)$ sapphire substrates with low microwave loss and a high remanence. Total thicknesses were from 5 to 20 μm. A reasonable compromise for low loss and high remanence was reached at a thickness of 7 μm, with a remanence ratio of 0.84 and a 59 GHz peak-to-peak derivative linewidth of 250 Oe. The 20 μm thick film had a linewidth of 110 Oe, one of the smallest values ever obtained for IPCA BaM films. © *2008 American Institute of Physics*. [DOI: 10.1063/1.3010374]

One important challenge for millimeter wave magnetic film device applications is in the development of low loss self-biased high anisotropy hexagonal ferrite films. Such films have potential uses for a wide range of monolithic microwave integrated circuit (MMIC) magnetic devices.[1–4] The critical parameter for self-bias is a high magnetic remanence, that is, a large average magnetization for the film at zero applied field. Low microwave loss is also critical for a real device with an acceptably low insertion loss.

Hexagonal *M* type barium ferrite (BaM) films, with a high uniaxial anisotropy and an effective anisotropy field (H_A) of 17 kOe or so and a nominal saturation induction $(4\pi M_s)$ of 4 kG,[5] are good candidates for such devices. The hexagonal *c*-axis serves as the easy magnetization direction. The high anisotropy field allows one to achieve the desirable millimeter wave frequency, even in the absence of an external biasing magnet.

There has been some progress for BaM films with the *c*-axis normal to the film plane (NCA). Song *et al.*[6] made such films by pulse laser deposition (PLD) with a near single crystal of 60 GHz ferromagnetic resonance (FMR) peak-to-peak derivative linewidth (ΔH_{pp}) of 16 Oe.[6] The remanent magnetization (M_r), however, was quite small. Excessive stress limited film thicknesses to 0.85 μm or so. Thicker NCA BaM films have been made by a hybrid PLD liquid phase epitaxy (LPE) approach.[7,8] A thin high quality PLD seed layer is first deposited. A second layer, up to 45 μm in thickness, is then grown by LPE methods. The nominal 60 GHz ΔH_{pp} values were in the 30–75 Oe range. The thick film remanence ratio M_r/M_s (RR)-values were near zero, as expected for high quality thick films. NCA BaM films have also been made by screen printing.[9,10] These films had moderate loss, with nominal 60 GHz ΔH_{pp} values in the 300–500 Oe range, and a high RR in the range of 0.96 range.

The new challenge, however, is to realize in-plane *c*-axis (IPCA) films with low loss and high RR values. The basic advantage of IPCA films is in the higher ferromagnetic resonance frequency. For given magnetization and anisotropy values, the zero external field FMR frequency for RR=1 goes from $\omega_{FMR}^{NCA}=|\gamma|(H_A-4\pi M_s)$ for a NCA film to $\omega_{FMR}^{IPCA}=|\gamma|\sqrt{H_A(H_A+4\pi M_s)}$ for an IPCA film. Here, γ denotes the gyromagnetic ratio. For the BaM parameters cited above, the $\omega_{FMR}^{IPCA}/\omega_{FMR}^{NCA}$ enhancement factor is about 1.45. This is significant. In principle, the realization of a high RR for IPCA films is also made easier, since the in-plane demagnetizing factors for thin films are close to zero and self-demagnetization effects are minimized.

Progress for IPCA BaM films has been limited. Yoon *et al.*[11,12] were able to make such films both on *a*- and *m*-plane sapphire substrates. High RR values above 0.9 were obtained for PLD films on *a*-plane sapphire substrates up to 7 μm or so in thickness, but the linewidths were large, in the range of kOe.[11] LPE films on *m*-plane sapphire appeared to have local regions with nominal 60 GHz ΔH_{pp} values below 100 Oe, but the overall linewidth was much larger and the films had relatively low RR-values.[12] Up until now, no group has been able to produce IPCA BaM films that combine both a high remanence and low loss.

Lattice mismatch and surface chemistry provide the key to successful epitaxy. From the narrow FMR linewidth in Ref. 6, it is clear that good epitaxy is achievable for NCA BaM films on sapphire substrates. The inability to produce low linewidth NCA films for thicknesses above 0.8 μm or so, however, also points to the presence of significant stress. From Refs. 11 and 12, it is clear that for IPCA films, both the *a*-plane and the *m*-plane sapphire substrate growth options present problems. For the present work, *a*-plane substrates were used, but hybrid PLD/LPE films were prepared. Relatively thin PLD Ba M epitaxial films were first grown on the *a*-plane substrate. This was done in anticipation of a good RR ratio for thicker LPE films grown on the PLD seed layer in the second step. Following Refs. 7 and 8, it was also expected that the hybrid PLD/LPE approach would also yield narrow linewidths. This expectation was realized, but some fine-tuning in the process parameters was needed. It was necessary, for example, to use lower deposition temperatures for the initial PLD seed layer than normally used to make the highest quality film. It appears that some degradation in the seed layer is needed in order to get adequate strain relief for the subsequent thick LPE layer.

The hybrid approach produced IPCA oriented *m*-plane BaM films with high remanence and moderate loss. Films in the 5–7 μm thickness range appear to offer the best remanence/linewidth compromise. At 7 μm, one has RR

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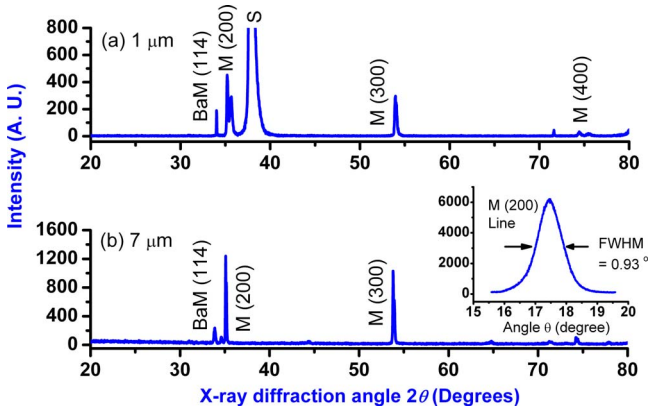

FIG. 1. (Color online) IPCA BaM film XRD 2θ scans. Graphs (a) and (b) show results for a 1 μm thick PLD seed film and the corresponding 7 μm thick hybrid film, respectively. The BaM, M, and S miller indices labels identify the important peaks, with S for the sapphire substrate and M for the characteristic *m* plane peaks. The (b) inset shows a rocking curve for the M (200) diffraction line.

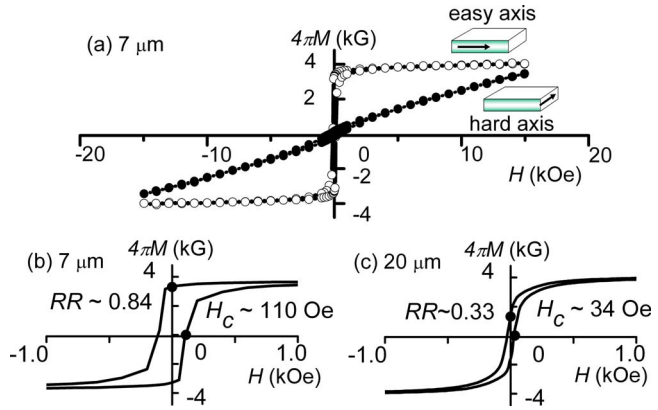

FIG. 2. (Color online) Hysteresis loops of the magnetic induction $4\pi M$ vs in-plane static field H. (a) and (b) show EA and hard axis field loops and the EA loop on an expanded field scale, respectively, for the 7 μm thick film in Fig. 1. (c) shows the expanded EA loop for the 20 μm film. Remanence ratios RRs and coercive fields (H_c) indicated in (b) and (c).

~0.84 and a 59 GHz FMR ΔH_{pp} value of 250 Oe. Thicker films show a decrease in ΔH_{pp} but also a decrease in remanence. At 20 μm, one has RR ~0.33 and ΔH_{pp} ~110 Oe. Thinner films show higher remanence values but degraded linewidths, presumably due to stress effects.

The first step was to lay down the thin seed layer. A nominal 1 μm thick epitaxial PLD seed layer was grown on *a*-plane $(11\bar{2}0)$ sapphire. The energy fluence and repetition rate for the 248 nm KrF excimer laser was 1.7 J/cm^2 and 25 Hz, respectively. The deposition time was 25 min, the substrate temperature was 870 °C, and the target substrate distance was 6.6 cm. These conditions gave bare unannealed PLD *m*-plane films with RR ~0.9 and a nominal 60 GHz ΔH_{pp} of about 1.2 kOe. X-ray diffraction (XRD) data showed that higher deposition temperatures can give somewhat better epitaxy but no improvement in linewidth. It is to be emphasized that the XRD data showed the growth of *m*-plane films. The large linewidths are likely due to the large *m*-plane BaM *a*-plane sapphire mismatch induced strain. The overall mismatch is complicated because of the overlay of these different classes of planes.

The second step is to lay down the thicker LPE film on the seed. A mixture of $BaCO_3$, Fe_2O_3, and B_2O_3 with mole ratio of 1.0:0.25:0.571, as in Ref. 8, was mixed thoroughly at 1250 °C for 6 h. The homogeneous melt was then cooled to the desired LPE growth temperature. A growth temperature of 860 °C, as measured with an external thermocouple, was found to give good epitaxial films. Lower temperatures gave mottled films. Higher temperatures yielded no film growth at all. As noted above, film thicknesses ranged from 5 to 20 μm. Thickness was controlled by the immersion time for the seed film in the supersaturated melt. The growth rate was about 0.2 μm/min. The film thicknesses were determined from cross section scanning electron microscopy (SEM).

Graphs (a) and (b) in Fig. 1 show standard $\theta/2\theta$ diffraction scans for a nominal 1 μm thick PLD film and a 7 μm thick hybrid PLD/LPE film, respectively. The inset in (b) shows the M (200) rocking curve. The diffraction peaks in (a) come mostly from the $(\ell 00)$ *m*-plane scattering of the hexagonal structure, except for a small peak from the (114) plane. One can also see the *a*-plane sapphire "S" peak. In (b), one sees the same $(\ell 00)$ *m*-plane reflections as in (a). Here, however, there is no sapphire peak and the intensities of the $(\ell 00)$ peaks in (b) are about three times higher than those in (a). The full width at half maximum of the XRD rocking curve is 0.93°. While the film is not perfectly oriented, the film is reasonably epitaxial. All told, the XRD data underscore the epitaxial growth of IPCA BaM PLD/LPE hybrid by the recipe given above.

The graphs in Fig. 2 shows representative room temperature vibrating sample magnetometry hysteresis loop data in an induction $4\pi M$ versus static field H format. The data in (a) and (b) are for the same 7 μm film as in Fig. 1. (a) shows data with the field H applied along the in-plane (IP) easy and hard directions, as indicated. (b) shows the easy axis (EA) IP loop on an expanded field scale. (c) shows similar data as in (b) but for a 20 μm film. The data in (a) show that the film has an in-plane uniaxial axis. The 7 μm film saturation induction $4\pi M_s$ and effective uniaxial anisotropy field H_A values are about 3.9 kG and 17.3 kOe, respectively. These are close to the reported values for single crystal NCA BaM films.[6] The corresponding $4\pi M$ and H_A values for the 20 μm thick film were 4.0 kG and 17.3 kOe, respectively. The 20 μm thick film data in (c) shows much lower coercive field and RR-values than do the 7 μm film data (b).

Graphs (a) and (b) in Fig. 3 show in-plane EA field 59 GHz FMR data (open circles) and Lorentzian derivative fits (solid curves) for the same films as in Fig. 2, shown in an absorption derivative versus field format. The fits are reasonably good. The FMR fields for the (a) and (b) data, taken as the zero crossing points from the fits, are about 2.07 and 2.24 kOe, respectively. The derivative linewidths are as indicated in the figure. For IP EA field ferromagnetic resonance, the applicable FMR frequency (ω) field (H_0) connection is given by

$$\omega = |\gamma| \sqrt{(H_0 + H_A)(H_0 + H_A + 4\pi M_s)}, \quad (1)$$

Based on the operating frequency of 59 GHz, $|\gamma|/2\pi = 2.8$ GHz/kOe, and the $4\pi M$ and H_A values cited above, one obtains theoretical FMR field values of 1.91 kOe (7 μm film) and 1.87 kOe (20 μm film). The matches with the data are reasonable and well within the range of error set by the uncertainties in $4\pi M$ and H_A.

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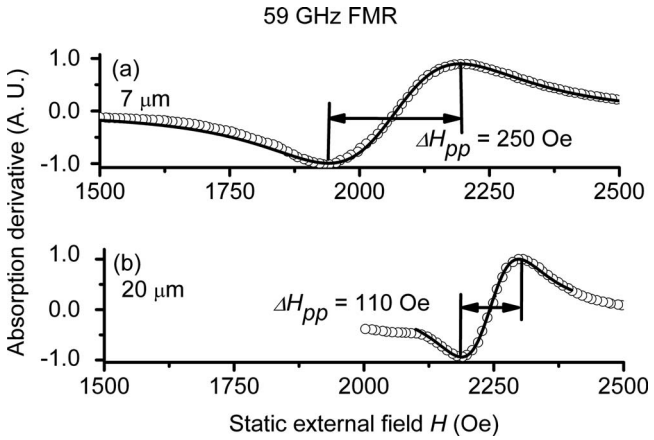

FIG. 3. Graphs (a) and (b) show 59 GHz FMR absorption derivative vs H profiles for the 7 and 20 μm thick films featured in Fig. 2, respectively. The open circles show the data. The solid curves show Lorentzian derivative best fits. The peak-to-peak derivative linewidths (ΔH_{pp}) are as indicated.

Turn now to the linewidths. Recall that the initial seed layer alone has $\Delta H_{pp} \approx 1.2$ kOe. The 250 Oe linewidth for the 7 μm hybrid film is about a factor of five smaller and the 110 Oe linewidth for the 20 μm hybrid film is more than a factor of 10 lower than the seed linewidth. It is clear that the thicker the hybrid film, the better the crystal alignment, the more the strain relief, and the better the overall film quality as measured by the linewidth.

Figure 4 shows (a) RR and (b) 59 GHz ΔH_{pp} linewidth values as a function of film thickness. From (a), one sees that the RR value decreases more or less smoothly from about 0.9 at 1 μm down to about 0.3 at 20 μm. As discussed in connection with Fig. 2, the decrease in the RR accompanied by a decrease in the coercive force as well. These decreases are tentatively ascribed mainly to the strain relief that comes

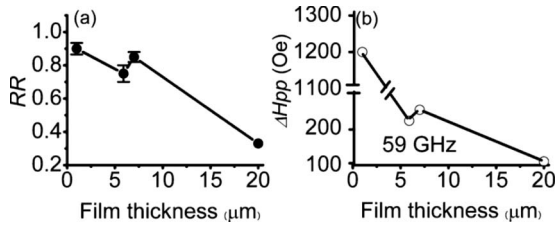

FIG. 4. (a) RR vs film thickness. (b) 59 GHz peak-to-peak derivative linewidth ΔH_{pp} vs film thickness. The data points for the 1 μm film are a pure PLD film, with the FMR measurements done at about 66 GHz.

with thicker LPE layers and the corresponding reduction in local stress related obstacles to domain wall movement. Graph (b) shows a similar trend for the linewidth. The nominal V band linewidth decreases from about 1200 Oe at 1 μm down to about 110 Oe for 20 μm thickness. Roughly speaking, one can see that a thickness around 7 μm or so offers a reasonable compromise for a high RR and low linewidth, with RR $\sim$0.84 and $\Delta H_{pp} \sim 250$ Oe. Thinner films offer high RR and high loss, while thicker films offer small RR and low loss.

In summary, hybrid PLD and LPE techniques have been used to produce IPCA BaM films with low microwave loss and a high remanence. The use of an initial thin PLD seed film of moderate quality with a LPE grown thicker film on top, appears to provide a viable way to produce useful films for planar high frequency MMIC devices. It is noteworthy, moreover, that the 110 Oe derivative linewidth for the 20 μm film is one of the smallest values achieved for IPCA oriented barium ferrite film on any substrate.

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