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## ADVERTISEMENT



## Structure, magnetic, and microwave properties of thick Ba-hexaferrite films epitaxially grown on GaN/Al<sub>2</sub>O<sub>3</sub> substrates

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Thick barium hexaferrite [BaO·(Fe<sub>2</sub>O<sub>3</sub>)<sub>6</sub>] films, having the magnetoplumbite structure (i.e., Ba *M*), were epitaxially grown on c-axis oriented GaN/Al<sub>2</sub>O<sub>3</sub> substrates by pulsed laser deposition followed by liquid phase epitaxy. X-ray diffraction showed (0,0,2n) crystallographic alignment with pole figure analyses confirming epitaxial growth. High resolution transmission electron microscopy images revealed magnetoplumbite unit cells stacked with limited interfacial mixing. Saturation magnetization,  $4\pi M_s$ , was measured for as-grown films to be  $4.1 \pm 0.3$  kG with a perpendicular magnetic anisotropy field of  $16 \pm 0.3$  kOe. Ferromagnetic resonance linewidth, the peak-to-peak power absorption derivative at 53 GHz, was 86 Oe. These properties will prove enabling for the integration of low loss Ba *M* ferrite microwave passive devices with active semiconductor circuit elements in systems-on-a-wafer architecture. © 2010 American Institute of Physics. [doi:10.1063/1.3446867]

The present microwave integrated circuit design paradigm has passive circuit elements, e.g., ferrite-based isolators, circulators, phase shifters, etc., fabricated on dielectric substrates, while active circuit elements, e.g., amplifiers, switches, signal processing devices, etc., are fabricated on semiconductor substrates. A long sought goal of the microwave device community has been the integration of passive circuit elements with active semiconductor electronic systems. Such a breakthrough would result in a “system on a wafer” architecture that substantially reduces size, weight, and costs associated with process and assembly, while concomitantly enhancing performance and functionality.<sup>1,2</sup>

The earlier steps toward realizing this goal were taken more than 20 years ago and involved attempts to grow highly oriented low microwave loss ferrite films on silicon (Si) and gallium arsenide (GaAs) (Ref. 3) substrates. Due to excessive interfacial alloying, as in the case of Si, and chemical disassociation, as in the case of GaAs, properties of those ferrite films were not viable for microwave applications. In recent years, emerging semiconductor materials, such as GaN and SiC, have demonstrated advantages in power handling and high frequency operation over mainstay materials such as Si and GaAs. These materials have received great attention from the semiconductor device community due to their attractive thermal conductivities, band gap energies, breakdown voltages, and permittivity, among other properties.<sup>4,5</sup> Additionally, these materials share the same hexagonal crystal symmetry and comparable lattice parameters as hexagonal ferrites and possess the high temperature stability that could enable epitaxial growth of high quality ferrites on semiconductor substrates.

Previously, we have demonstrated the epitaxial growth of Ba *M* on 6H-SiC substrates by pulsed laser deposition

(PLD).<sup>6</sup> Following this breakthrough was the demonstration of nanoscale patterning of Ba *M* films using high rate reactive etching.<sup>7</sup> These advances, taken together, provide a viable pathway for the advanced integration of planar microwave devices with semiconductor integrated circuit platforms. The necessary next step is the growth of thick, 10 s of microns, oriented ferrite films. Such thicknesses are required in many microwave applications including Y-junction circulators<sup>8</sup> that are widely used in transmit and receive modules in phased array radar and communication systems, among other applications. The thickness of PLD prepared ferrite films is typically less than 1 μm owing to the slow deposition rate inherent in PLD (Refs. 9–11). Alternatively, liquid phase epitaxy (LPE) growth rates for ferrites can be much higher, often 10 s of micrometer per hour, and as such is a powerful tool in the growth of near-single crystal thick ferrite films.<sup>12–14</sup> Despite the superior thermal properties of SiC, this substrate has proven unstable in Ba *M* flux melts used in LPE causing the SiC substrates to dissolve. We attribute this in part to the chemical affinity of Si and Fe as is evidenced by the eutectic transition observed in the Fe–Si phase diagram at temperatures near 875 °C; a typical temperature for the LPE growth of Ba *M*.<sup>12–14</sup> In contrast, GaN, as will be shown, is compatible with the LPE technique and therefore is the best hope to realize ferrite integration with semiconductor platforms. The choice of GaN as well reflects its important role in next generation microwave power electronics, such as in high power amplifiers.<sup>15</sup>

In order to realize heteroepitaxial growth, thermal and structural compatibilities of the substrate’s surface and the growing film are key factors. In the case of Ba *M* grown on GaN, there is a relatively large lattice mismatch of 6.2% between the GaN (001) substrate and the Ba *M* (001) film. Further complicating growth is that GaN is thermally unstable *in vacuo* at temperatures near 900 °C; conditions that

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are typically employed for PLD growth of Ba *M*. In order to mitigate both interfacial strain and surface reactivity, an 8 nm buffer layer of MgO, having (111) crystallographic orientation, was deposited by PLD on the GaN substrate prior to the growth of Ba *M*. To prepare the surface of the substrate for growth, GaN/Al<sub>2</sub>O<sub>3</sub> single crystal wafers were ultrasonically cleaned in acetone and alcohol followed by a rinse in aqua regia solution. These preparation steps were found to result in smooth surfaces on the subnanometer scale, largely free of defects and contamination. Following the preparation of the MgO (111) buffer layer, Ba *M* was ablated from a homogeneous BaFe<sub>12</sub>O<sub>19</sub> target within the same chamber without disrupting vacuum. A KrF Excimer laser, of wavelength 248 nm operating at 250 mJ per pulse, was employed in pulsed laser ablation deposition. An oxygen pressure of 20 mTorr, with a corresponding substrate temperature of 900 °C, was determined to be optimal based upon the structure, magnetic and microwave properties of resulting films. These conditions are consistent with previously published studies of the growth of Ba *M* deposited on lattice-matched oxide substrates.<sup>9–11</sup> The PLD films were then used as seed layers for variant thick film growth by LPE.

After the growth, the crystallographic and magnetic properties of the films were systematically examined by the following techniques. The dc magnetic properties of the as-deposited films were measured at room temperature using vibrating sample magnetometry (VSM, Lakeshore Model 7400). The microwave properties of the films were studied by ferromagnetic resonance (FMR) measurements between 46 to 58 GHz using the shorted waveguide technique. The films were attached to the side wall of the waveguide with the external magnetic field applied perpendicular to the film plane. X-ray diffraction (XRD)  $\theta$ - $2\theta$  patterns and pole figure analyses were obtained using a Cu *K* $\alpha$  source in a Rigaku thin film diffractometer (Model Ultima III). The film interface was imaged in cross section by high resolution transmission electron microscopy (HRTEM), using a Titan 80–300 TEM, equipped with a spherical-aberration (image) corrector, which was operated at an accelerating voltage of 300 kV.

A representative XRD pattern of an as-grown film is presented as the logarithm of intensity versus  $2\theta$  in Fig. 1. All detected diffraction peaks have been indexed to (0, 0, 2*n*) Miller indices of the hexagonal Ba *M* structure indicating a strong crystallographic *c*-axis texture perpendicular to the substrate plane. To further characterize the crystal quality of the Ba *M* films, pole figures were obtained from the (006) and (008) reflections in which the angle between the film normal and the vector bisecting the incident and detected x-ray beams,  $\phi$ , was varied from 0° to 90°, and the azimuthal angle about the bisecting vector,  $\xi$ , was varied from 0° to 360°. The sharp peak at the center of the (006) pole figure [Fig. 2(a)] indicates *c*-axis alignment normal to the film plane with low in-plane dispersion. The sixfold symmetry of low intensity peaks arising from {104} Ba *M* planes have similar values of *d*-spacing. Similarly, threefold symmetry of low intensity peaks derives from the *d*-spacing of {100} MgO planes in which  $2\theta=23.30^\circ$  for Ba *M* (104) and  $2\theta=21.06^\circ$  for MgO (100). The measured interplanar angle between the MgO (100) and Ba *M* (006) planes is 55.00° compared with the expected value of 54.74° estimated for the MgO (111) planes when aligned parallel to Ba *M* (006) planes. With regard to the (008) reflection, three MgO {110} re-

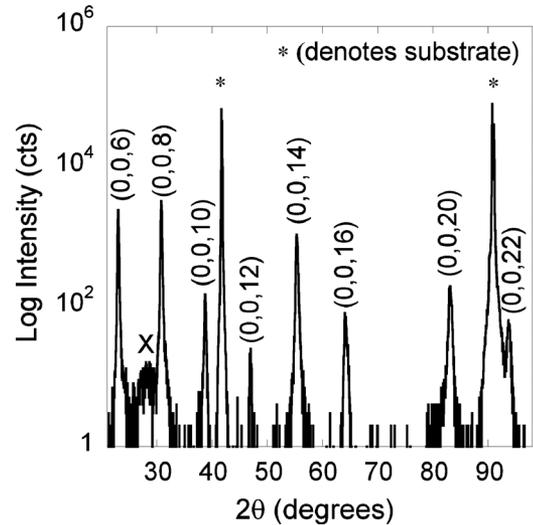


FIG. 1. XRD pattern for an as-deposited Ba *M* film. All significant diffraction features are indexed to (0,0,2*n*) Miller indices having space group *P6/mmc*. X signals a single diffuse peak that has not been identified. \* denotes GaN/Al<sub>2</sub>O<sub>3</sub> substrate peaks.

flections having interplanar angle of 35.26° with respect to the MgO(111)//Ba *M*(008) planes appear with threefold symmetry measured at 35.00°. These results confirm the epitaxial growth of Ba *M* (001) on MgO(111)/GaN(001)/Al<sub>2</sub>O<sub>3</sub>(001). Quantitative analysis of the (006) pole figure by the peak angular integration suggests that more than 95% of the Ba *M* grains have *c*-axes aligned within 5° of the film normal.

Figure 3 presents HRTEM cross section images of the Ba *M* films grown on GaN with a buffer layer of MgO (111). The images reveal regions of GaN, MgO, and hexaferrite phases. Importantly, a small region of intermixing, approximately ~1 nm, is measured at the MgO/GaN interface. This level of intermixing is relatively small when compared to other reports detailing the growth of hexaferrites on semiconductor substrates.<sup>16</sup> Interestingly, there appears a second

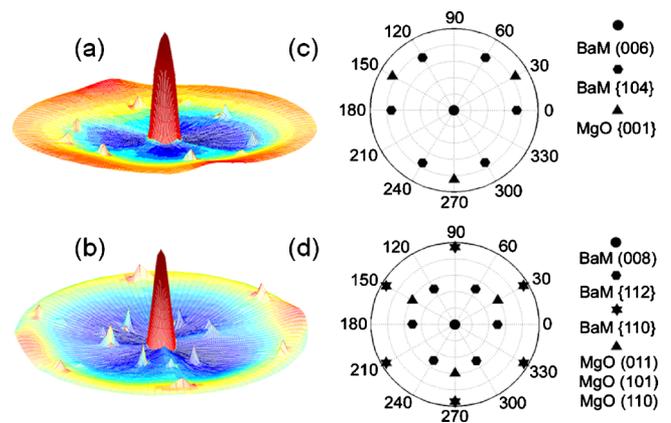


FIG. 2. (Color online) Pole figures obtained for (a) the (006) and (b) (008) reflections, and the corresponding two-dimensional projections (c) and (d), with  $2\theta$  values fixed at 23.00° and 30.30°, respectively. The single dominant peak in (a) corresponds to  $\phi=\xi=0^\circ$  for the Ba *M* (006) reflection. The weaker peaks in (a) and (c) exhibiting sixfold symmetry corresponds to closely spaced Ba *M* (104) reflections. The minor peaks exhibit threefold symmetry in (a) and (c) deriving from closely spaced MgO (100) reflections illustrating the epitaxial nature of the Ba *M* films grown on MgO/GaN/Al<sub>2</sub>O<sub>3</sub>. Similarly, (b) and (d) show similar data for the (008) reflection further supporting the epitaxial growth of films.

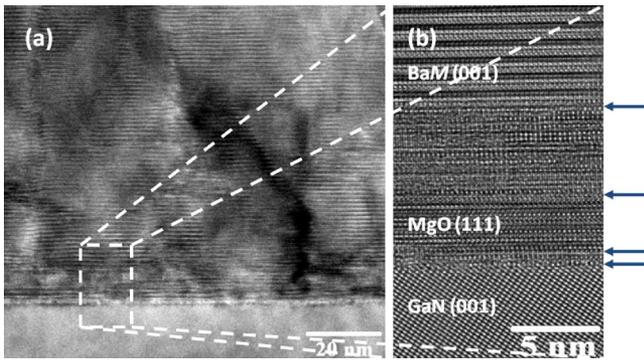


FIG. 3. (Color online) Transmission electron microscopy images of the Ba *M* film grown on MgO/GaN/Al<sub>2</sub>O<sub>3</sub> in cross section. The image (b) is an expanded view of the section denoted in (a). The regions denoted by arrows signify intermixing.

region of intermixing at the MgO/Ba *M* interface that encompasses  $\sim 5$  nm in thickness. We conjecture that this region consists of a Mg(Ba)-spinel ferrite phase. We expect that this spinel phase is comparatively magnetically soft and indeed in Fig. 4(a) one observes a pronounced change in slope near remanence in the hysteresis loop collected along the hard axis. This change in slope signals the switching of a soft magnetic phase that is decoupled from the harder Ba *M* phase. Because this interfacial phase is magnetically soft, and constitutes a small volume fraction of the total magnetic structure, it does not degrade the microwave performance.

Both in-plane and perpendicular hysteresis loops of the as-deposited Ba *M* films are presented as Fig. 4(a). It is shown that the easy axis of the Ba *M* film is aligned out of the film plane consistent with the crystallographic *c*-axis aligning perpendicular to the sample plane. The hysteresis loops collected along the easy direction showed a maximum squareness ( $M_r/M_s$ ) of more than 90% providing these sample potential for self-biased applications that, for example, allow for the elimination of bias magnets in such applications as circulators and isolators.<sup>17</sup> The anisotropy field is estimated at  $16.0 \pm 0.3$  kOe and the saturation magnetization is  $4.1 \pm 0.3$  kG. A low coercive field,  $\sim 10$  Oe, was measured along the magnetic hard axis. Low coercivity along the hard direction reflects the less effective pinning of domain walls from the defected interface volume and has

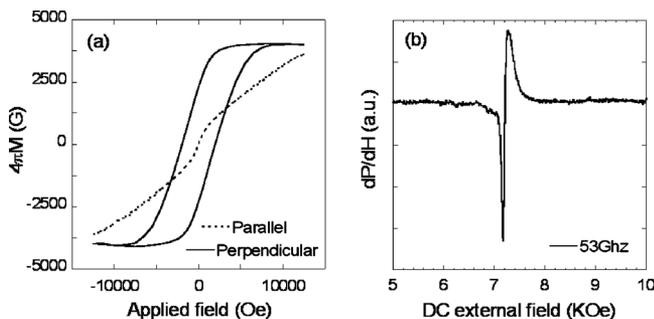


FIG. 4. (a) Hysteresis loops obtained by VSM with a maximum applied field of 13 000 Oe aligned perpendicular (solid) and parallel (dashed) to the film plane. (b) Power derivative as a function of applied magnetic field in the region near the FMR at 53 GHz. The linewidth measured as the peak-to-peak power derivative is 86 Oe for the  $4 \mu\text{m}$  thick as-deposited Ba *M* film grown by PLD and LPE in tandem on MgO(111)/GaN(001)/Al<sub>2</sub>O<sub>3</sub>(001).

been phenomenologically linked to low microwave losses in Ba *M* films. These values closely match those of other high quality Ba *M* films deposited on lattice matched oxide substrates.<sup>9–11</sup>

The microwave properties were measured by FMR. The power derivative as a function of applied magnetic field in the region near the FMR at 53 GHz is shown in Fig. 4(b). The measured resonance frequency matched very well to the calculated theoretical resonance;  $f = \gamma'(H + H_a - 4\pi M_s)$ . The FMR linewidth of the thick LPE film ( $\sim 4 \mu\text{m}$ ),  $\Delta H$ , is 86 Oe. This value compares well with values reported for Ba *M* films grown on lattice matched oxide substrates<sup>9–11</sup> and is superior to those previously grown on 6H-SiC.<sup>6</sup>

In summary, barium hexaferrite films of thicknesses of  $0.4 \mu\text{m}$  having  $4\pi M_s$  of  $4.1 \pm 0.3$  kG and  $H_A = 16.0 \pm 0.3$  have been grown by PLD on GaN substrates buffered with a 8 nm crystalline PLD-grown MgO (111) layer. Liquid phase epitaxial growth on the PLD-grown seed layers allowed for the synthesis of thick films up to  $4 \mu\text{m}$  having near single crystal properties and low microwave FMR linewidth ( $\Delta H$ ) of 86 Oe. These results demonstrate that high crystal quality, low microwave loss, barium hexaferrite films can be grown on high performance wide band gap semiconductor substrates, specifically GaN. This demonstration provides a pathway to realizing the integration of high performance ferrite microwave passive devices with active circuit elements on a common semiconductor substrate; a necessary step in creating “system on a wafer” architectures.

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