Structure, magnetism, and tunable microwave properties of pulsed laser deposition grown barium ferrite/barium strontium titanate bilayer films

R. Heindl, H. Srikanth,[a] S. Witanachchi, and P. Mukherjee
Department of Physics, University of South Florida, Tampa, Florida 33620

T. Weller
Department of Electrical Engineering, University of South Florida, Tampa, Florida 33620

A. S. Tatarenko and G. Srinivasan
Department of Physics, Oakland University, Rochester, Michigan 48309

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Ferrite/ferroelectric films are of interest as they afford dual tunability of the permeability and permittivity using magnetic and electric fields. We have grown bilayered thin films of ferrimagnetic $BaFe_{12}O_{19}$ (BaM) and ferroelectric $Ba_{0.5}Sr_{0.5}TiO_3$ (BST) on *A*-oriented polished sapphire substrates using pulsed laser deposition. X-ray thin film analysis (θ-2θ and azimuthal scans) established highly oriented crystalline films. Magnetic hysteresis loops indicated large magnetic anisotropy between the in-plane and out-of-plane field orientations and the *M*-*H* characteristics of BaM were influenced by the presence of the BST layer. Electrical and magnetic tunability studies of the bilayers in the frequency range of 1–65 GHz are also reported. © *2007 American Institute of Physics*.
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Rapid advances in rf and microwave devices require utilizing improved high-frequency tunable dielectric and magnetic materials. Ferrimagnetic *M*-type barium hexaferrite ($BaFe_{12}O_{19}$, BaM) and ferroelectric barium strontium titanate ($Ba_{0.5}Sr_{0.5}TiO_3$, BST) have shown excellent properties in the microwave and millimeter-wave regions of the electromagnetic spectrum and have been commonly used in tunable filters, phase shifters, isolators, and circulators.[1–4] BaM is characterized by relatively low losses at gigahertz frequencies and high magnetic anisotropy that could be useful in self-biased microwave magnetic device applications. BST is a well studied and technologically important material. Due to its inherent nonlinear dielectric properties, its microwave response can be tuned by applying electric fields.

While BaM and BST have individually been well studied, there are obvious advantages in growing bilayers of these materials on compatible substrates due to the need for multifunctional materials in the present day era of microelectronics and rf microelectromechanical system (MEMS) technology. This can be achieved through formation of composite films or films grown in the stacked bilayer geometry. Magnetodielectric multilayers have also been of current interest because of the possibility of magnetoelectric coupling and tunability of ferromagnetic resonance (FMR) that could potentially lead to other devices.[5,6]

Over the past few years, we have investigated the growth, microstructure, and magnetism in BaM/BST structures produced by magnetron sputtering. Our studies have led to fine-tuning of the growth parameters and observation of interesting magnetic properties that indicated the important role of interfacial properties between the BaM and BST layers (in bilayers) and BaM and BST grains (in composite films).[7,8] In this paper, we report on high quality bilayer films of BaM/BST grown on *A*-oriented sapphire substrates using pulsed laser deposition (PLD). Along with the structural and magnetic properties, we also demonstrate the electrical and magnetic tunabilities of these bilayers at microwave frequencies up to 65 GHz.

BaM/BST bilayer thin films were grown on *A*-oriented polished sapphire substrates using PLD with the substrate heated up to 650 °C. Conductive silver paste was applied at the substrate-heater interface to promote uniform heating during deposition. From our past experience and published literature from other groups, it is well documented that substrate heating is critical for the formation of single phase, homogeneous barium ferrite. The deposition pressure was set at 250 mtorr of high purity O_2. Several different combinations of BaM/BST and substrates were experimented with. In this paper, we discuss only the structure with BaM as the bottom layer grown on Al_2O_3 with the top layer being BST. The films were postannealed in a quartz tube furnace in flowing oxygen at 900 °C for 8 h and the thickness of each layer was estimated to be around 0.5 μm.

The crystallographic orientation and texture were determined using an x-ray diffractometer (Bruker Instruments, Cu $K\alpha$ radiation) and the magnetic properties were measured in a 7 T physical properties measurement system (PPMS) from Quantum Design. In order to measure electrical tunability, coplanar transmission lines were deposited on top of the bilayer structure using a standard optical lift-off lithography process and thermal evaporation of 15 nm of Cr and ≈1 μm of Au. The dimensions of the signal conductor and the gap between the signal and the ground conductors were 11.5 and

[a]Author to whom correspondence should be addressed; electronic mail: sharihar@cas.usf.edu

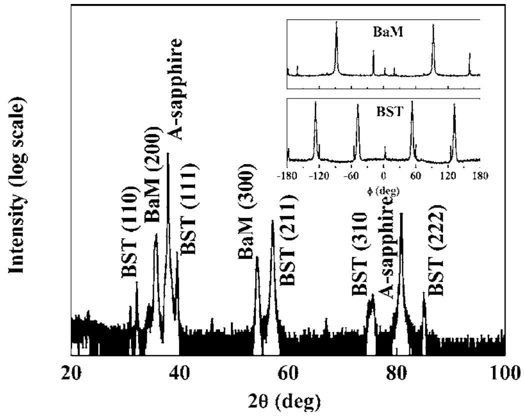

FIG. 1. X-ray diffraction analysis: $\theta-2\theta$ scan. Inset: azimuthal (φ) scan of BaM (220) peak at $2\theta=63.060°$ and $\Phi=30°$ and BST (200) peaks at $2\theta=63.060°$ and $\Phi=65°$.

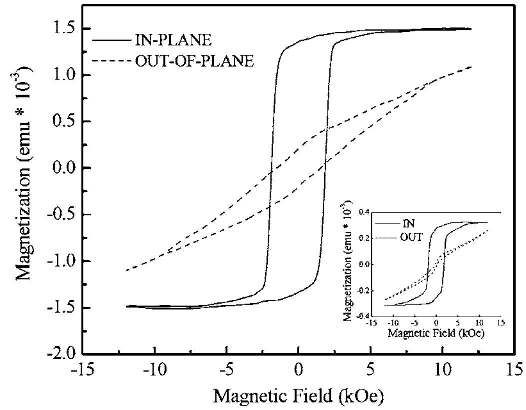

FIG. 2. Magnetization hysteresis for magnetic fields parallel and perpendicular to the film surface. Inset shows the magnetization of a single layer BaM film.

5 μm, respectively. Electric fields were applied through a bias tee between the signal and the ground conductors. A standard short-open-load-thru (SOLT) calibration was performed and *S* parameters were measured using an Anritsu 37937C vector network analyzer in the frequency range of 1–65 GHz. Magnetic tunability of the microwave absorption was measured at Oakland University using a resonant cavity method based on a *U*-band waveguide-coaxial adapter.

Figure 1 shows the x-ray diffraction (XRD) θ-2θ scan for a BaM/BST bilayer grown on *A* sapphire. Two prominent peaks coming from the BaM layer can be seen, belonging to the (h00) group, which indicates that the *c* axis of the BaM lies in the film plane. BST shows a polycrystalline structure, with strongest reflection from the (211) plane. The azimuthal scans (φ scans) of the BaM (220) and BST (200) atom planes shown in the inset reveal the in-plane orientation of the film. The BaM has one set of two large peaks at $\phi=90°$ and $\phi=270°$. This is expected from crystallites with their *c* axes in the film plane with only two possible reflections coming from the planes parallel to (0001) and $(000\bar{1})$. There is also another set of two smaller peaks coming from another set of in-plane oriented crystallites rotated by 70°. No out-of-plane orientation was recorded, indicating a very good crystallographic quality of the BaM phase. The bottom panel, corresponding to the BST film, shows a set of four peaks, consistent with the cubic crystal structure. Some additional peaks appearing in the φ scans indicate a somewhat lower degree of film texture, due to the presence of multiple crystallites, consistent with the θ-2θ scan.

Magnetization versus applied field (M-H) data at room temperature (300 K) for the BaM/BST bilayer are presented in Fig. 2. The main figure shows the hysteresis loops with *H* applied parallel (in plane) and perpendicular (out of plane) to the film surface. The inset shows the data for an individual BaM single layer film on *A*-oriented sapphire substrate (made under similar growth conditions) for comparison. Large anisotropy difference of the loops can be observed for the in-plane and out-of-plane orientations, comparable to the results in literature.[9] Large squareness is also noticeable in the in-plane hysteresis loops (0.87 for the single layer and 0.91 for the bilayer). This is consistent with the microstructural results suggesting that the easy axis of magnetization lies in the film plane with the hard axis perpendicular to the plane. A comparison between the out-of-plane magnetization of the single BaM layer and that of the BaM/BST bilayer shows that there is some difference in the magnetization characteristics. It appears that the BST layer has some influence on the shape of the *M-H* curve. For example, the coercivity is higher compared to that of the single BaM layer. The coercivities of the in-plane *M-H* loops are about 1820 and 1870 Oe for the single layer and bilayer, while the coercivities of the out-of-plane loops are 230 and 1525 Oe for the single layer and bilayer, respectively. This is an important finding, indicating possible interaction of the layers presumably mediated by strain at the BaM/BST interface. The bottom of the BST layer likely influences the orientation of the BaM crystallites close to its interface, causing them to be different (i.e., *c*-axis oriented out of plane) compared to the overall nature of the BaM growth (as seen in the XRD) which favors the in-plane geometry. We have repeatedly seen the influence of BST on the orientation of BaM crystallites over the years in samples grown under various conditions as well as different techniques such as sputtering and PLD. These facts indicate the interesting potential for magnetodielectric coupling effects in BaM/BST layered as well as composite films.

The electrical tunability of the bilayer films was measured using coplanar transmission lines as described earlier. The phase shift of the transmission coefficient S_{21} is described as[10]

$$\Delta\Phi = \Phi_2 - \Phi_1 = \frac{360fl}{c}\left(\sqrt{\varepsilon_{\rm eff}^{\rm field}\mu_{\rm eff}^{\rm field}} - \sqrt{\varepsilon_{\rm eff}^{\rm no\_field}\mu_{\rm eff}^{\rm no\_field}}\right), \quad (1)$$

where *f* is the operating frequency, *l* is the line length, *c* is the speed of light, and $\varepsilon_{\rm eff}$ and $\mu_{\rm eff}$ are effective permittivity and permeability of the sample, respectively. From Eq. (1), it can be seen that the relative change in effective permittivity as a function of externally applied electric field would result in a phase shift of the transmission coefficient. Figure 3 shows the measured phase shift per unit length at different applied voltages in the frequency range of 1–65 GHz. The phase shift increases with the frequency for a given applied voltage bias and the slope becomes larger as the electric field is increased. These trends are consistent with microwave di-

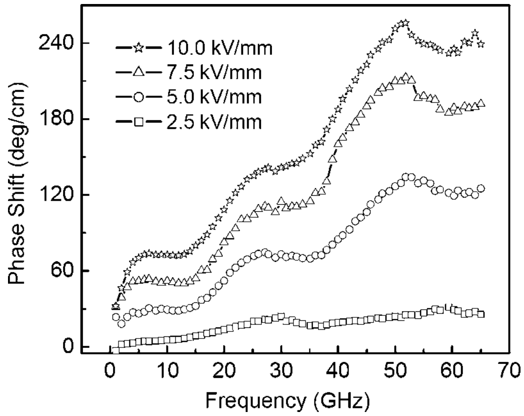

FIG. 3. Differential phase shift at various applied electric fields.

electrics based on BST. Another study of a dual-tunable device consisting of a BST film deposited on an yttrium iron garnet (YIG) substrate[10] reported the electrical phase shift from the BST film to be 17°/cm with 2.1 kV/mm at 10 GHz, which is comparable to the data reported here. The insertion loss was relatively high most likely due to the poor impedance match at the BST layer and the signal ports. The poor impedance match can also explain the wavy nature of the curves in Fig. 3. Resonances in the BST/BaM/substrate structure may also contribute to this fine structure.

Study of the magnetic tunability of the microwave response in magnetodielectric films is less common and is generally restricted to FMR experiments.[11] While we have not done conventional FMR measurements on these bilayers, we have attempted to get some information using a resonant technique specifically developed to study magnetoelectric phenomena in materials.[12] In this experiment, the BST/BaM samples were placed at the metal short end of a *U*-band waveguide-coaxial adapter and the reflected power was measured at different external magnetic fields ranging from 0 to 14 kOe using an electromagnet. Preliminary results of the return loss are shown in Fig. 4. In the frequency range of 40–60 GHz, multiple resonances were observed with most of them identified as magnetic polaritons.[13] The polaritons are hybrid excitations of electromagnetic waves in the substrate and/or BST and forward volume magnetostatic waves in the BaM film. The top four panels of Fig. 4 show this absorption signal plotted at different fields. The bottom panel is a trace of the minima of the curves as a function of the external field. These data are qualitatively in agreement with results obtained on heterostructures of BaM and strontium gallate.[14] A noticeable feature in this set of data is that there are distinct jumps in the minimum frequency position at characteristic fields above 8 kOe. These jumps are due to mode repulsion of magnetic and dielectric resonances. While more work is needed before one can interpret these complex features, it should be pointed out that the data in Fig. 4 are reminiscent of hybrid spin-electromagnetic waves that have been observed in similar experiments on layered YIG-BST structures.[15] If indeed these are related to spin-electromagnetic modes, it would open up the possibility of microwave and millimeter-wave device structures based on the BST/BaM bilayers. A detailed study of the electrical and magnetic tunabilities in BST/BaM films including the tunability of the FMR signal is beyond the scope of this conference paper and will be presented in a forthcoming publication.

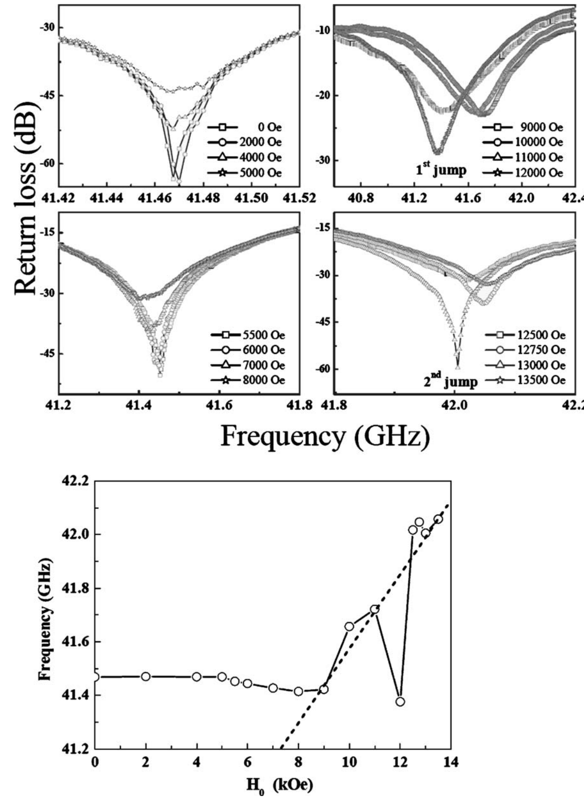

FIG. 4. (a) Microwave absorption and (b) position of the resonant absorption in BaM-BST multilayer as a function of external field.

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