Electric Tuning of Ferromagnetic Resonances in Hexagonal-Barium-Ferrite/Barium-Strontium-Titanate Heterostructures

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Abstract—This letter reports the first demonstration of a monolithic heterostructure with a large electric tuning of the ferromagnetic resonance (FMR) at millimeter-wave frequencies. The structure is composed of a barium-strontium-titanate (BST) film and two thin platinum electrodes, all grown on a barium ferrite slab with in-plane uniaxial anisotropy. The electric tuning of the FMR responses around 60 GHz was obtained for bias voltages across the BST layer in the 0–30 V range. The average tuning rate was 1.1 MHz/V. The highest incremental response was 2.1 MHz/V. These rates are significantly higher than the previously reported values for nonmonolithic structures.

Index Terms—Magneto-electronics, multiferroics, hexagonal ferrite, barium strontium titanate, heterostructures, ferromagnetic resonance, millimeter wave.

I. INTRODUCTION

Recent work has demonstrated the use of electric fields for the tuning of the ferromagnetic resonance (FMR) responses in the microwave regime for the ferromagnetic–ferroelectric-layered heterostructures [Ustinov 2006, Das 2009, Demidov 2002] as well as ferromagnetic–piezoelectric-layered heterostructures [Liu 2009, Fetisov 2005, 2008]. For the first class of structures, the tuning relies primarily on the excitation of hybrid magnetoelectric modes. In slightly more detail, a change in the bias voltage across the ferroelectric layer gives rise to a change in its dielectric constant; and this, then results in a change in the hybrid magnetoelectric modes and a corresponding shift in the FMR frequency of the ferromagnetic layer. For the second class of structures, the tuning relies on the effects of electrostriction and magnetostriction. Specifically, a change in the bias voltage across the piezoelectric layer results in a change in its stress; through interfacial coupling, this change then leads to a change in the stress in the ferromagnetic layer. Such stress changes in turn result in a change in the magnetic properties and FMR response in the ferromagnetic layer. These heterostructures have far-reaching implications for the future of electrically tunable microwave magnetic devices. Such devices can have a number of advantages over conventional magnetic-field tuning devices. These include fast tuning, small size, and negligible power consumption.

There is also limited work on layered heterostructures that showed the electric tunability of the FMR responses at millimeter (mm) wave frequencies [Das 2007, Song 2009, Ustinov 2008]. In this case, the heterostructures utilize M-type hexagonal-barium ferrite BaFe\textsubscript{12}O\textsubscript{19} (BaM) thin films as ferromagnetic components. Barium ferrites have a high effective anisotropy field, often in the range of 15–20 kOe. This built-in magnetic bias can yield an FMR response at mm-wave frequencies even for modest external magnetic fields. Song [2009] described a BaM/barium-strontium-titanate (Ba\textsubscript{0.5} Sr\textsubscript{0.5} TiO\textsubscript{3}, BST) heterostructure with a reasonable electric field tuning of the FMR response at about 60 GHz. Ustinov [2008] reported on a BaM/lead-zirconium-titanate (PZT) heterostructure with an electric tunability in the 100 GHz range. These results clearly show the potential of BaM-based heterostructures for low-weight and low-power electrically tunable mm-wave devices for radar and communications.

The missing link for such devices [Song 2009, Ustinov 2008] is in the development of a monolithic structure. This letter reports on the realization of such a structure. From bottom to top, the structure consists of a BaM slab, a thin platinum (Pt) electrode, a BST thin film, and a top Pt electrode. The BaM slab had an in-plane uniaxial anisotropy and was cut from a BaM single crystal prepared by flux-melt growth techniques. The growth of the Pt and BST layers were done by pulsed laser deposition (PLD) techniques. The structure shows an electric field tuning rate of about 1.1 MHz/V for the nominal 60 GHz FMR frequency for a bias voltage in the 0–30 V range. This tuning level is double the values given in Song [2009] and about 100 times higher than those given in Ustinov [2008]. The structure shows a maximum incremental tuning rate of about 2.1 MHz/V at 30 V. This rate is more than 200 times the values reported in Ustinov [2008].

The significant improvement found is from the direct monolithic growth of the Pt and BST layers on the top of the BaM slab. This approach gives good interfacial contact and a strong layer-to-layer coupling. In stark contrast, the active layers in Song [2009] and Ustinov [2008] were mechanically pasted onto each other. It is clear that this mechanical process gives a much weaker layer-to-layer coupling and a corresponding low level of tunability. The current results point the way to enhanced...
tuning in monolithic structures and a new generation of BaM-heterostructure-based monolithic mm-wave devices.

II. STRUCTURE PREPARATION AND CHARACTERIZATION

For the present structure, the BaM base element was in the shape of a 1.8 mm by 1.5 mm rectangle and with a nominal thickness of 300 \( \mu \text{m} \). The PLD-deposited BST film was about 3.4 \( \mu \text{m} \) thick and was put down at 550 \( ^\circ \text{C} \). The bottom and top Pt electrodes were about 0.13 and 0.2 \( \mu \text{m} \) thick, respectively, and were put down at room temperature. The PLD process used a 248-nm KrF excimer laser. For the BST layer, the laser energy fluence was 1.7 J/cm\(^2\), the repetition rate of laser pulses was 25 Hz, and the target–substrate distance was 7.8 cm. For the Pt electrodes, the energy fluence was 3.5 J/cm\(^2\), the pulse repetition rate was 5 Hz, and the target–substrate distance was about 3.0 cm.

The physical properties of the BaM/BST heterostructure were determined by scanning electron microscopy (SEM) and X-ray diffraction (XRD). The static magnetic properties of the BaM component were measured by the vibrating sample magnetometry at room temperature. The dielectric constant of the BST component was obtained from capacitance measurements at 100 kHz. The FMR responses were measured at 60 GHz with a V-band-shorted waveguide system, field modulation, and lock-in techniques.

III. RESULTS AND DISCUSSION

Fig. 1 shows details of the heterostructure assembly. Fig. 1(a) shows a schematic of the layers, Fig. 1(b) shows an SEM image in cross-section view, and Fig. 1(c) gives a representative XRD spectrum of the assembly. The labels in Fig. 1(c) give Miller indexes for the indicated peaks. Several results are evident from the panels in Fig. 1. With regard to the Pt layers, the SEM data in Fig. 1(b) confirm the Pt layer thicknesses previously cited and show that the bottom Pt layer is continuous and has a uniform thickness; and the XRD data in Fig. 1(c) indicate that the Pt layers are (1 1 1)-oriented. With regard to the BaM layer, the two main XRD peaks in Fig. 1(c) indicate the in-plane c-axis orientation of the component. This orientation is essential to the realization of in-plane uniaxial anisotropy in the BaM slab. With regard to the BST component, the XRD data show that this layer is well oriented. Finally, the XRD spectrum shows no other peaks for components other than those previously noted.

The bottom Pt layer plays a critical role in the oriented growth of the BST film. First, as the lattice mismatch between the BST (1 1 1) and Pt (1 1 1) planes is only about 0.5%, the well-oriented Pt layer provides a good template for epitaxial BST growth. The bottom Pt layer also appears to have a second positive effect, the amelioration of the stress between the highly mismatched BaM (l 0 0) and BST (1 1 1) lattice planes. This, along with a properly chosen thickness for the Pt layer, serves to promote oriented BST growth. The selected Pt layer thickness of 0.13 \( \mu \text{m} \) is in the right range. Thinner layers typically break up into islands during deposition, while thicker Pt layers tend to peel.

Fig. 2 shows the measured relative dielectric constant \( \varepsilon_r \) of the BST film component as a function of bias voltage. As indicated, the solid and open circles show the data for increasing and decreasing voltage sweeps, respectively. These data show three things. First, the electric tunability is about 20% at the relatively low-bias voltage limit of 30 V. Second, the \( \varepsilon_r \)–V responses for the increasing and decreasing voltage sweeps are slightly shifted. These shifts are due to the ferroelectric hysteresis for the BST. Finally, the data indicate that the response is linear for voltages above about 5 V and more rounded for lower voltages. This is likely due to the shape of the ferroelectric hysteresis loop at low voltage.

The data in Figs. 1 and 2 demonstrate the good quality of the embedded BST layer. As earlier noted, the realization of this good BST layer is largely due to the development of a proper bottom Pt layer. This high-quality tunable BST component plays an essential role in the realization of a large electric tuning of the FMR response considered next.
The FMR data in Fig. 3 demonstrate the strong electric tuning response. The FMR responses were measured at 60 GHz for different bias voltages applied across the BST layer. Graph in Fig. 3(a) shows FMR absorption derivative profiles for BST bias voltages of 0, 20, and 30 V, as indicated. Graph in Fig. 3(b) shows the FMR field, defined as the midpoint in field for the derivative profile, as a function of the bias voltage. Graph in Fig. 3(c) shows the equivalent shift in the FMR frequency versus bias voltage had the experiment been done at a fixed magnetic field. The conversion was based on the standard BaM thin-film FMR equation with an effective in-plane uniaxial anisotropy field of 16.7 kOe and a practical gyromagnetic ratio of $\gamma = 2.87 \text{ MHz/Oe}$ [Buffler 1962].

Fig. 3(a) shows four important things. First, there is a clear shift of the FMR profiles to lower fields with an increase in the bias voltage. This shift provides direct evidence for the electric tuning of the FMR response. Second, in spite of the substantial shift, the FMR profiles remain almost unchanged in shape and the linewidth is preserved. This fact is important for practical devices. Third, with the BaM parameters previously listed, one obtains a theoretical 60 GHz FMR field value of 3200 Oe. This value is in close agreement with the data. Finally, all the profiles in Fig. 3(a) indicate peak-to-peak FMR linewidths in the 30 Oe range. This matches with the linewidth value of the initial BaM slab. Such a match in linewidth indicates that the deposition of the BST and Pt layers from top did not affect the $Q$ factor of the FMR in the BaM slab. Note that, in previous work on mechanically assembled BaM/BST heterostructures, the pasting of the BST film also made no difference to the $Q$ factor of the FMR [Song 2009].

The data in Fig. 3(b) and (c) give an FMR field shift of about 11 Oe and an equivalent frequency shift of 32 MHz at a bias voltage of 30 V. The data also yield an average incremental tuning rate of about 1.1 MHz/V. Previous work has yielded much lower tuning rates of about 0.01 and 0.55 MHz/V for BaM/PZT and BaM/BST nonmonolithic structures, respectively. One can see that the tuning rates obtained here are notably higher than that for mechanically pasted heterostructures. From Fig. 3(c), one can see that the incremental frequency tuning actually increases with bias voltage. The data give a maximum incremental tuning rate of about 2.1 MHz/V at 30 V. This is almost twice the rate for the BaM/BST nonmonolithic structures in Song [2009]. Relative to the results for stress-tuned BaM/PZT nonmonolithic structures [Ustinov 2008], the current incremental tuning rate is more than 200 times larger. It should be noted that the tuning rate here was defined as the rate of the frequency change with bias voltage, and this rate is of interest for device applications. The rate in terms of the change of frequency with electric fields, however, was also often used in literature. In terms of frequency change with electric field, the tuning rates for the heterostructure presented here are actually of the same order as that for the previous BaM/PZT structure.

From a more general perspective, there are also four key points for emphasis. First, “monolithic is better.” From the point of view of practical devices, a monolithic heterostructure offers more benefits than a mechanically pasted heterostructure. These include a larger tunability, a lower bias voltage, and a better compatibility with monolithic integrated circuits. The tunability enhancement derives from the strong coupling between the ferromagnetic and ferroelectric layers. In previous mechanically assembled heterostructures, there exists an interlayer, either a layer of glue or an air gap, in between the ferromagnetic and ferroelectric (or piezoelectric) layers. This interlayer limits the coupling between the two active layers and, hence, limits the tunability. It should be noted that the tunability enhancement through the monolithic approach was first demonstrated with a yttrium-iron-garnet/BST heterostructure at microwave frequencies [Das 2009]. This work with this study clearly shows the advantage of monolithic heterostructures over mechanically assembled heterostructures. Second, “in-plane configuration is better.” When the structure is magnetized in-plane, the dipolar fields in the ferromagnetic film are perpendicular to the film plane, go into the ferroelectric layer, and provide the strong coupling. Third, “thin embedded platinum electrodes are better.” The tunability enhancement demonstrated here is due to the strong BaM/BST coupling as well as the high quality of these layers. One can expect even more enhancement in the layer-to-layer coupling with a reduced and optimized embedded Pt layer thickness. Finally, it is the large in-plane uniaxial anisotropy of the BaM slab that facilitates FMR operation at 60 GHz with modest magnetic fields. For other materials, one needs much higher magnetic fields for FMR operations at the same frequency. For example, an in-plane magnetized yttrium iron garnet film requires a magnetic field of about 23 kOe; and a BaM film with perpendicular-to-plane uniaxial anisotropy requires a field of about 9 kOe.

The previously demonstrated frequency shift is substantial for a low bias voltage of only 30 V. Many practical devices, however, need a much wider frequency tuning range. Future work on further enhancement in the frequency tuning range is of great interest. In principle, one can increase the frequency shift by the use of a higher bias voltage or through an enhancement in the tuning rate. From a point of view of
device applications, however, low bias voltages are always preferred. In order to increase the tuning rate, possible approaches include: 1) optimization of the thicknesses of all the layers and the lateral dimension of the heterostructure; 2) enhancement of electric tuning in the ferroelectric layer; and 3) the use of electrode-ferromagnetic–ferroelectric-electrode configuration. For approach (1), the main idea is to change the curvature of the dispersion curve of hybrid modes and the wavenumber of the resonance mode for optimal tuning [Demidov 2002]. Approach (2) can be realized through a change in the composition of the ferroelectric film [Im 2000] as well as the optimization of the deposition processes. For approach (3), the growth of a ferroelectric layer on the top of a ferromagnetic layer offers a direct layer-to-layer contact and, therefore, yields stronger layer-to-layer coupling and a corresponding large tuning rate [Song 2010].

IV. CONCLUSION

In summary, this letter shows that one can make BaM/BST monolithic mm-wave heterostructures with a very high level of electric tuning of the magnetic response. The structure was made by the PLD, in turn, of a bottom Pt electrode, a BST layer, and then, a top Pt electrode onto an in-plane c-axis-oriented BaM slab. The Pt and BST layers were well oriented, and the BaM slab retained its initial predeposition quality. The monolithic fabrication and the quality of the structure yield a substantial increase in the electric tuning of the mm-wave FMR response, relative to all previous work. The results clearly demonstrate the tremendous potential for BaM hybrid-mode-based monolithic heterostructures as electrically tunable on-chip mm-wave devices.

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REFERENCES