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Microwave tunability in a GaAs-based multiferroic heterostructure: Co₂MnAl/GaAs/PMN-PT

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A strong magnetoelectric (ME) interaction is presented in a magnetostrictive-semiconductor-piezoelectric heterostructure that consists of the Heusler alloy, Co₂MnAl, GaAs, and lead magnesium niobate-lead titanate (PMN-PT). The laminated Co₂MnAl/GaAs/PMN-PT structure, having a thickness of 19 nm/180 μm/500 μm, demonstrates a ferromagnetic resonance (FMR) field shift of 28 Oe with an external electric field of 200 V across the PMN-PT substrate. This corresponds to a resonance frequency shift of ~125 MHz at X-band. It yields a large ME coupling (7 Oe cm/kV) and microwave tunability (~32 MHz/kV cm⁻¹), compared to other trilayer multiferroic composite structures. In addition, static magnetization measurement indicates a reduction in the remanence magnetization while applying the electric field, which corroborates the ME interactions mediated by the translation of magnetoelastic forces in this structure. This work explores the potential of multiferroic heterostructure transducers for use in FMR microwave devices tuned by electric fields. © 2009 American Institute of Physics. [DOI: 10.1063/1.3068543]

I. INTRODUCTION

Magnetoelectric (ME) materials have recently received much attention for both their fundamental physical properties and potential to enable a new generation of electric-field tunable electronic devices.^{1,2} An increasing interest is devoted to a class of materials referred to as multiferroics, which feature the simultaneous occurrence of ferromagnetism and ferroelectricity in a single or multiphase material or heterostructure. Although most known single phase multiferroic materials do not exhibit strong ME coupling at room temperature, the artificially structured materials, typically constructed of layers or granular composites, bring unique opportunities in realizing many devices, such as electric field controlled magnetic memory elements, ferromagnetic resonance devices, microwave gyrators, transformers, and transducers with magnetically modulated piezoelectricity.³⁻⁹ Mechanical coupling with piezoelectric layers provides electrical control of magnetic properties of the magnetostrictive layers, which eliminates the need for applied magnetic fields to control the performance of the ferromagnetic component. Therefore, multilayer heterostructures, consisting of piezoelectric and magnetostrictive layers, function as “true” single-phase multiferroic materials. A logical next step is the integration of the multiferroic element with semiconductor substrates, which would enable on-chip integration and the development of advanced multifunctional electronic devices and systems.¹⁰ In particular, GaAs-based platforms, featuring very low dielectric losses in the rf and microwave bands, are especially

attractive for the incorporation of multiferroic elements in a new class of multifunctional monolithic microwave integrated circuits (MMMICs).¹¹

The present work reports on the ME properties of a layered multiferroic structure consisting of a ferromagnet grown on GaAs substrate and ferroelectric lead magnesium niobate-lead titanate (PMN-PT). Many Heusler alloys, such as Co₂MnAl and Co₂MnSi,^{12,13} are expected to be ideal half-metals with the majority of electrons in one spin state. This makes them useful for spintronic applications, e.g., spin injectors. Furthermore, this class of Heusler alloy films has shown ferromagnetic resonance linewidths of 100–200 Oe at X-band frequencies.¹⁴ They provide new opportunities in highly spin polarized microwave device applications. Therefore, the present research aims to investigate the ME interactions between the Co₂MnAl thin film and ferroelectric crystals at X-band (i.e., $f \sim 10$ GHz) microwave frequencies in this GaAs-based heterostructure. This paper addresses some important results in the study of multiferroic systems, which will enrich research in multiferroic materials and devices.

II. EXPERIMENT

An epitaxial film of Co₂MnAl was grown on a (100)-oriented GaAs substrate by molecular-beam epitaxy using solid source elements. Details of the growth procedure were described in Ref. 12. The Co₂MnAl film, having a thickness of 19 nm, exhibits a cubic structure at room temperature while displaying a phase transition temperature between 760 and 810 K. The heterostructure system employed a PMN-PT with both ferroelectric and piezoelectric properties. The [011]-poled PMN-PT single crystal, with 28%–32% PT, fea-

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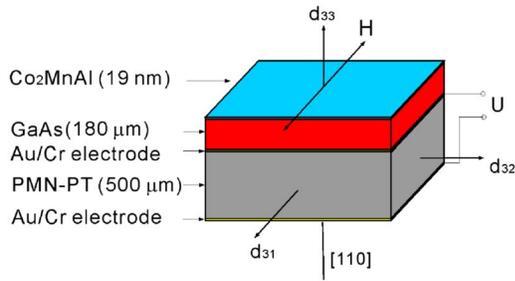


FIG. 1. (Color online) Schematic diagram for a $\text{Co}_2\text{MnAl}/\text{GaAs}/\text{PMN-PT}$ multiferroic heterostructure.

tures anisotropic transverse piezoelectric coefficients, d_{31} and d_{32} , i.e., $d_{31} = -1800$ pC/N and $d_{32} = 900$ pC/N.¹⁵ In comparison, the more widely used lead zirconate titanate ceramics have an isotropic piezoelectric coefficient, d_{31} , far smaller at ~ -400 pC/N.³

The multiferroic composite explored here was designed to operate in the L-T ME coupling mode (i.e., longitudinal magnetized/transverse polarized) and is a laminated structure consisting of Co_2MnAl film and PMN-PT single crystal poled along with [011]. Figure 1 is a sketch of the composite structure. Note, that the PMN-PT crystal has a thickness of 0.5 mm and was bonded to a 180 μm thick GaAs substrate with a quick curing ethyl cyanoacrylate glue. Unlike previously reported multiferroic heterostructures,¹⁶ the bonding of the Co_2MnAl magnetic film to the PMN-PT crystal is mediated by the GaAs substrate. This structure has a ratio of (film+GaAs) to PMN-PT thickness of 180/500 or 36%. This is an important parameter in the structural design of the MF element since it is difficult to realize strong ME coupling if this ratio is too large.

Ferromagnetic resonance (FMR) measurements were carried out using a microwave cavity excited in a TE_{102} mode at X-band ($f = 9.55$ GHz), where both the external field H_0 and microwave magnetic field \vec{h} were applied in the plane of the sample structure. Here, H_0 is along with the d_{31} direction of the PMN-PT crystal for ME coupling measurements. The magnetostriction constant of the Co_2MnAl film was measured by using a high precision optical technique over a magnetic field range of 0–250 Oe.¹⁷

III. RESULTS AND DISCUSSION

Since FMR measurements were carried out with an applied magnetic field aligned along the in-plane [110] direction for the Co_2MnAl film at X-band, without application of an electric field, the resonance condition can be simply expressed as follows:¹⁸

$$f = \gamma' \sqrt{(H - H_k) \left(H + \frac{1}{2} H_k + 4\pi M_s \right)}, \quad (1)$$

where H , H_k , and $4\pi M_s$ denote the external field for in-plane measurement, magnetocrystalline anisotropy field, and the saturation magnetization, respectively. The measurement indicates a FMR linewidth $\Delta H = 210$ Oe, which roughly corresponds to a half-width of frequency, $\Delta f \approx 920$ MHz at the resonance frequency $f = 9.55$ GHz.

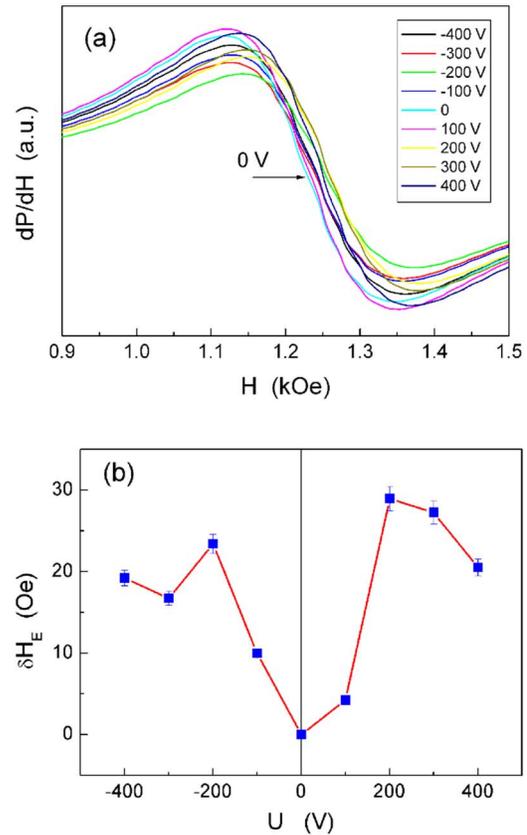


FIG. 2. (Color online) (a) FMR absorption derivative varies with a magnetic field at different electric fields, and (b) an induced internal magnetic field is tuned by an electric field across PMN-PT crystal.

Figure 2 presents the results of in-plane microwave measurements under an applied electric field aligned along the d_{31} direction of the PMN-PT crystal, i.e., [110] direction. In general, the direction of the field shift is determined not only by the configuration of magnetic and electric fields but also by the magnetoelastic nature of the magnetic film and ferroelectric/piezoelectric crystal used in the MF element. For example, while an electric field is applied along the d_{33} direction, the d_{31} direction experiences compressive strain if $d_{31} < 0$. In the case when an external magnetic field is applied parallel to the d_{31} direction of PMN-PT crystal and the magnetostriction constant of the magnetic film is positive, an internal stress-induced magnetic field aligns perpendicular to the external magnetic field in the film plane. This scenario is illustrated in Fig. 1. Since this strain-induced anisotropy field is obviously uniaxial, the magnetic resonance field corresponding to zero electric field is therefore minimum as illustrated in Fig. 2(a). While carefully considering the FMR measurement results in Fig. 2(a), we note a complex correlation between resonance field shift and applied electric field. Figure 2(b) clearly indicates that a 200 V electric field results in a maximum induced magnetic field of 24–28 Oe. Larger electric fields do not result in larger field shifts or ME coupling.

Although the underlying physical principles of the heterostructure ME response are relatively straightforward, an accurate calculation of the ME interaction remains challenging.¹⁹ Here, an estimate of the strain-induced field is derived as below,⁶

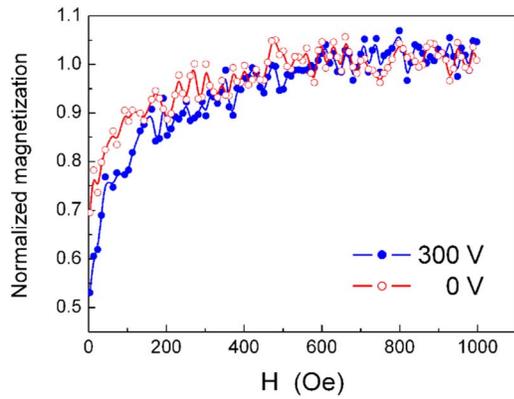


FIG. 3. (Color online) Recoil magnetization curves at an application of electric field of 300 V and at zero field for a $\text{Co}_2\text{MnAl}/\text{GaAs}/\text{PMN-PT}$ multiferroic heterostructure.

$$\delta H_E = \frac{3\lambda\sigma}{M} = AE, \quad (2)$$

where A is the ME constant; λ and σ are magnetostriction constant and the stress in the Co_2MnAl film, respectively. An estimated induced field, δH_E is 20–40 Oe in terms of the parameters: $\lambda = 12 \pm 4$ ppm, $C_{11} = 1.52 \times 10^{12}$ dyn/cm², $C_{12} = 1.43 \times 10^{12}$ dyn/cm²,²⁰ and $M = 630$ G for the Co_2MnAl film, as well as $d_{31} = -1750$ pC/N, $d_{32} = 900$ pC/N, and $E = 4$ kV/cm for the PMN-PT crystal. Therefore, this experiment demonstrates a good agreement between the theoretic prediction and the measured values. Furthermore, a ME coupling constant, $A = 7$ Oe cm/kV, is much larger than previously reported values for other trilayered structures, such as YIG/GGG/PMN-PT (Ref. 21) and FeGaB/Si/PMN-PT.²²

In addition, we also calculated the electric field tunable FMR frequency shift $\delta f = 125$ MHz, while applying an electric field of 200 V across the PMN-PT slab. This result substantiates the microwave tunability, 31.3 MHz/kV cm⁻¹, of this heterostructure, which is twice the value of the YIG/GGG/PMN-PT structure, where YIG and GGG represent yttrium iron garnet and gadolinium gallium garnet, respectively.²¹ We are also able to obtain a ratio of frequency shift to half-width frequency $\delta f/\Delta f$ of 13.6%: a parameter that is of interest in the design of microwave devices as well as a means of evaluating and comparing the microwave tunability of MF heterostructures.

Finally, we also observed a dc ME coupling effect for this structure by the measurement of the static recoil magnetization curves under an electric field during vibrating sample magnetometry. Figure 3 clearly shows a change in remanence magnetization, i.e., the remanence reduces from 0.7 to 0.55 under the application of a 300 V electric field. This represents a 20% reduction in magnetization due to the ME coupling. This static tunability arises from ME interactions mediated by the translation of magnetoelastic forces through the heterostructure.

IV. CONCLUSION

We have successfully fabricated a layered multiferroic heterostructure that consists of a Co_2MnAl film grown on (100) GaAs substrate bonded to a PMN-PT. The trilayer structure exhibits a significant FMR field shift of 28 Oe and a frequency shift of 125 MHz under a relatively low electric field of 200 V. It in turn leads to a large ME coupling constant of 7 Oe cm/kV and microwave tunability of 32 MHz/kV cm⁻¹. These results mark the first report of a MF heterostructure based on a GaAs substrate and may enable the integration of multiferroic elements in a new class of MMMICs.

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