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Microwave magnetoelectric effects in ferrite—piezoelectric composites and dual electric and magnetic field tunable filters

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Abstract Magnetoelectric (ME) effects at ferromagnetic resonance (FMR) in ferrite—piezoelectric layered structures have been investigated in lithium zinc ferrite (LZFO)-lead zirconate titanate (PZT) and yttrium iron garnet (YIG)-PZT. When an electric field *E* is applied to PZT, the piezoelectric deformation is transmitted to the ferrite and results in a shift in FMR. Data on the field shift caused by *E* are presented and compared with theory. Although the strength of ME coupling is weaker in YIG-PZT than in LZFO-PZT, the *E*-assisted field shift in YIG-PZT is much higher than the resonance line width and is potentially useful for filter applications. An electric field tunable YIG-PZT microwave band-pass filter based on ME effects at FMR is designed and characterized. The device can be tuned over a wide frequency band with a bias magnetic field and over a narrow band with a voltage applied across PZT. Data on tuning range, insertion loss, and device characteristics are presented.

Keywords Magnetoelectric · Ferrite · Piezoelectric · Ferromagnetic resonance · Microwave filter

1 Introduction

A multiferroic is a material that exhibits two or more of the primary ferroic properties (ferromagnetism, ferroelectricity, ferroelasticity, ferrotoroidicity) in the same phase and have enormous potential for novel device applications. Materials in which ferromagnetism and ferroelectricity occur simultaneously in the same phase and allow coupling between the ferromagnetic and ferroelectric phase are known as magneto-electric (ME) multiferroics [1]. Most of the known single phase bulk ME materials do not exhibit strong ME coupling, have Néel or Curie temperatures far below room temperature, and are often difficult to grow in thin film form [2, 3]. These limitations have to be overcome for practical technological applications of these materials. Unlike single phase materials, relatively large ME coefficients have been obtained in bulk laminated composites consisting of ceramic multilayers of ferroelectric and ferromagnetic materials. When a magnetic field is applied to the composites, the ferrite magnetostrictive phase induces a strain in terms of a shape change, which in turn exerts stress on the piezoelectric phase, resulting in an electric polarization. This extrinsic ME response in the multiferroic composites is due to the composite effect of the two phases, implying that a property like ME effect can be induced in two different phases, which themselves individually are not magneto-electrically active. PZT/ferrite and PZT/Terfenol-D are the most studied composites to date [4, 5]. However, magneto-electric properties of mechanically coupled multilayered (piezoelectric-magnetostrictive) heterostructures can be limited by substrate clamping. Therefore, an alternative is to investigate heterostructures with coupling surfaces in a plane normal to the substrate. Such heterostructures are not expected to be limited by substrate clamping and may exhibit enhanced magneto-electric effects. Zheng et. al [6] have demonstrated growth of self-assembled nanopillars of magnetostrictive material ($CoFe_2O_4$) in a piezoelectric $BaTiO_3$ matrix by Pulsed Laser Deposition (PLD). It is important to note that

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due to increased surface to volume ratio in nanostructures, better elastic coupling and therefore better phase coupling can be achieved in such nanopillars.

Here we focus on engineered multiferroics with ferromagnetic and piezoelectric subsystems. The systems are of interest for studies on the nature of magnetoelectric (ME) coupling and for use in sensors, transducers, and signal processing technologies [4]. Efforts so far on such composites have primarily focused on low frequency (10 Hz–1 kHz) phenomena [5, 7, 8]. Studies on ME coupling at high frequencies could be performed through the measurement of electric field (E) assisted shift of ferromagnetic resonance lines (FMR) for the ferromagnet [9–11]. The shift δH_E arises due strain dependence of the resonance field H_r and its magnitude is determined by the piezoelectric and magnetoelastic constants. Thus δH_E vs E data could be used to understand the nature of ME couplings and determine ME constants. We investigated the resonance ME effects in bulk composites of 90% YIG–10% PZT and observed the expected shift [12]. But for PZT amounts larger than 20%, the main FMR line broadens and masks any E induced shift. Such difficulties, however, are absent in layered structures since the coupling is essentially an interface phenomenon and FMR line broadening due to nonmagnetic PZT is practically absent.

The focus of this work are (1) an understanding of the effects of magnetic parameters of ferrites on ME coupling in multilayers of $Li_{0.5-x/2}Zn_xFe_{2.5-x/2}O_4$ (LZFO; x=0–0.4) and PZT and in single crystal yttrium iron garnet (YIG)-PZT and (2) microwave filters based on YIG-PZT. Lithium ferrite and YIG, in particular, are appropriate for studies on microwave resonance ME effects because of low losses [13] and thus facilitates accurate determination of δH_E and ME constants. We synthesized lithium zinc ferrite-PZT thick film multilayers by sintering films obtained by tape casting. Films of YIG grown by liquid phase epitaxy on gadolinium gallium garnet (GGG) substrates were used to obtain bilayers with PZT. Microwave ME measurements at 9.3 GHz were performed using a traditional FMR spectrometer. Profiles of FMR absorption vs bias magnetic field H were obtained for a series of electric voltage across PZT. With the application of E, we measured the shift δH_E in the profile by and the ME constant $A=\delta H_E/E$ varied over the range 1–1.5 Oe cm/kV. Since 1 Oe of field shift corresponds to a frequency shift of 2.8 MHz, FMR-based microwave devices can be tuned with E. We designed and characterized a YIG-PZT band-pass filter that can be tuned with an electric field. The ME effect would facilitate rapid voltage tuning for any FMR-based microwave device. Other advantages of microwave ME devices include miniaturization, near-zero power consumption, noise reduction and compatibility with integrated circuit technology [14].

2 Experiments

Multilayer structures consisting of alternate layers of $Li_{0.5-x/2}Zn_xFe_{2.5-x/2}O_4$ (x=0–0.4) and PZT were prepared from thick films synthesized by the doctor blade technique. Films 10–40 μm in thickness were made. Samples with n PZT layers and ($n + 1$) ferrite layers (n=5–30) were prepared. Details of the multilayer synthesis and structural and magnetic characterization are discussed in Ref. [15]. X-ray diffraction studies indicated the presence of two characteristic sets of reflections, corresponding to the ferrite and PZT, and the absence of new phases. Saturation magnetization measured with a Faraday balance was in agreement with expected values for ferrites. Magnetostriction was measured with the standard strain gage technique. The resistivity, dielectric constant and piezoelectric coupling constants were in agreement with expected values for PZT. For YIG-PZT bilayers, we used 58 mm thick (111) YIG on 0.5 mm thick GGG substrates. The film was epoxy bonded to 0.5 mm thick polycrystalline PZT.

For microwave ME effects, ferromagnetic resonance studies using a resonance cavity operating at 9.3 GHz were carried out at room temperature. The static magnetic field was applied either parallel or perpendicular to the sample plane and power absorption by the sample was measured as a function of H. The samples were then subjected to a pulsed (2 ms) electric field across PZT. The use of pulsed field was necessary to avoid any heating of the sample. The resonance absorption versus static magnetic field profile was obtained for a series of electric fields. The resonance field was thus measured as a function of E and the field shift data was used for estimation of ME constants.

3 Results

First we consider studies on LZFO-PZT. Figure 1 shows FMR profiles illustrating the effects of electric field for pure lithium ferrite-PZT. The results are for a multilayer of LFO (x=0)-PZT with 15 micron thick 16 ferrite layers and 15 PZT layers of same thickness. For E=0, FMR with a line-width ΔH on the order of 700 Oe is observed. With the application of E=80 kV/cm, there is a 40 Oe downshift in the resonance field. We measured a linear dependence of δH_E on E and the estimated ME coefficient $A=\delta H_E/E=$ 0.25 Oe cm/kV. Similar measurements were made on samples with thicker PZT layers so that the ferrite-to-PZT volume fraction v is in the range 0.3–1.0. Figure 2 shows data on the variation of the shift δH_E with v for E=22 kV/cm for a sample of LZFO (x=0.2)-PZT. It is clear from the data that a decrease in the ferrite volume leads to an increase in the strength of ME interactions as expected. Similar measurements could not be performed for samples with

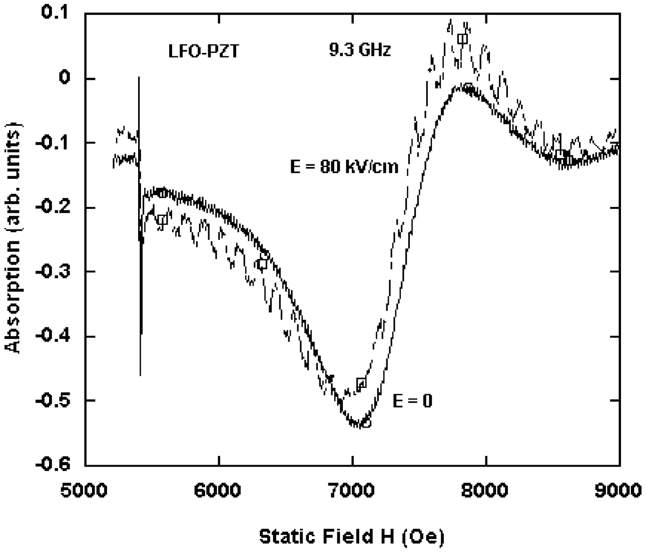

Fig. 1 Resonant magnetoelectric effect measured in a multilayer composite of lithium ferrite (LFO)-lead zirconate titanate (PZT). The sample contained 16 layers of LFO and 15 layers of PZT. The thickness of each layer is 15 µm. The static field *H* is perpendicular to the sample plane and 2 ms pulses of *E* is applied perpendicular to the plane. Absorption vs *H* profiles are shown for *E*=0 and *E*=80 kV/cm

higher x because the FMR absorption was very broad with $\Delta H \sim 1000$ Oe. The large ΔH masked any *E*-induced shift in the resonance field.

In our theoretical model for high frequency ME effects in layered composites, we assumed a ferrite-piezoelectric bilayer [9, 10]. An external field E causes an interface strain due to piezoelectric effect and produces shift of the resonance magnetic field. We obtained the following expression for the field shift

$$\delta H_E = \frac{3\lambda_{100} d_{31} E_3}{M_0\left[\left({}^{p}s_{11} + {}^{p}s_{12}\right)(v/1+v) + \left({}^{m}s_{11} + {}^{m}s_{12}\right)v\right]} = AE \tag{1}$$

where A is a magnetoelectric constant, M_0 is a saturation magnetization and λ_{100} is the magnetostriction constant. In Eq. 1 m denotes the magnetostrictive phase and p the piezoelectric phase, d is the piezoelectric coefficient, and s is the compliance coefficient. Using the following values for material parameters ${}^{p}s_{11}=15*10^{-12}$ m^2/N, ${}^{p}s_{12}=-5*10^{-12}$ m^2/N, ${}^{m}s_{11}=6.5*10^{-12}$ m^2/N; ${}^{m}s_{12}=-2.4*10^{-12}$ m^2/N, $d_{13}=-175$ pm/V,. $\lambda_{100}=23\cdot 10^{-6}$, and $4\pi M_0=3600$ G, we obtain the ME constant A=0.2 Oe cm/kV. The estimated value is in excellent agreement than the measured value of 0.25 Oe/(kV/cm) for LFO-PZT multilayers. The volume dependence of the shift δH_E was estimated for LZFO (x= 0.2)-PZT and compared with data in Fig. 2. There is very good agreement between theory and data.

We also investigated the microwave ME coupling in YIG-PZT bilayers prepared by bonding (111) YIG film on to PZT discs. FMR profiles as in Fig. 1 were obtained for a series of *E* values. Figure 3 shows the variation with E of the shift in the resonance field at 9.3 GHz for bias magnetic field parallel to the sample plane. The shift is linear with an ME constant *A*=0.98 Oe cm/kV. Thus the ME coupling is a factor of four stronger in YIG-PZT than in LZFO-PZT.

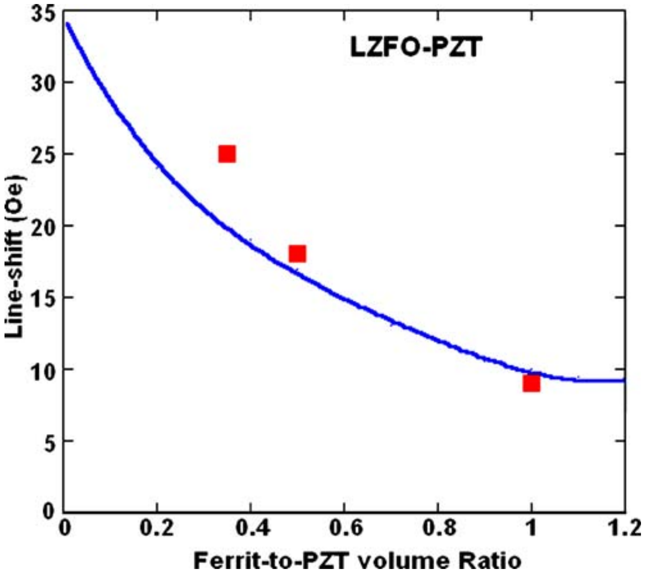

Fig. 2 Dependence of the resonance line shift on ferrite-to-PZT volume ratio of $Li_{0.4}Zn_{0.2}Fe_{2.4}O_4$-PZT multilayer sample at *E*=22 kV/cm. The composite contained 16 layers of $Li_{0.4}Zn_{0.2}Fe_{2.4}O_4$, with the layer thickness of 38 µm, and 15 layers of PZT with the same layer thickness

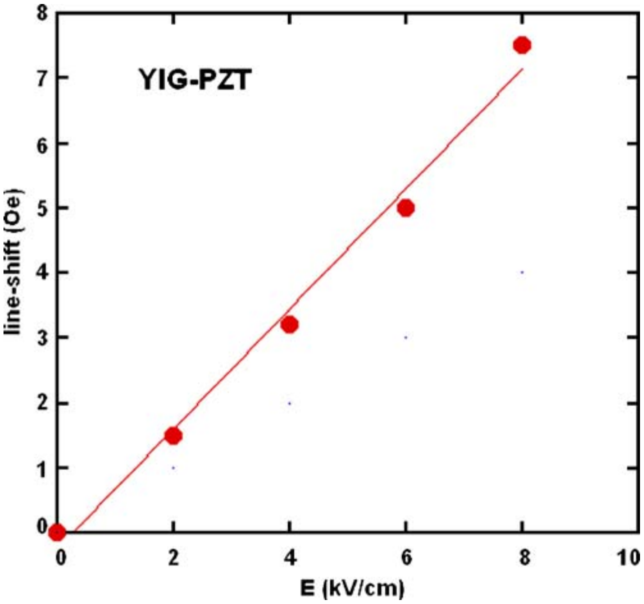

Fig. 3 Resonance field shift at 9.3 GHz as a function of *E* for a bilayer of 58 mm (111) YIG and PZT

4 Ferrite-Piezoelectric Filters

The data in Figs. 1, 2 and 3 provide evidence for electric field tunability of ferrite-PZT devices that are based on FMR, such as resonators, filters and phase shifters. Ferrites are used in tunable microwave and millimeter-wave devices and the tunability is traditionally realized through the variation of a bias magnetic field [16]. This magnetic tuning could be achieved over a very wide frequency range, but is relatively slow, noisy, and requires high power for operation. Similar devices but with some unique advantages could be realized by replacing the ferrite with a ferrite-ferroelectric composite [14]. A critical requirement for such tuning is the width of FMR in the ferrite. Polycrystalline LZFO shows a very large line-width, on the order of 800 Oe which is 10 times higher than the E-assisted in Fig. 1. But LPE films of YIG have very narrow FMR line-width of 0.5–1 Oe which is an order of magnitude smaller than the E-assisted shift in Fig. 3. We, therefore, designed and characterized YIG-PZT band pass filters. Our studies indicate very good tunability due to a strong ME interaction and an acceptable insertion loss.

Single and dual element YIG-PZT filters were studied. The dual-element filter, shown in Fig. 4, consists of a 1 mm thick dielectric ground plane (permittivity of 10), input and output microstrips of nonresonance lengths, and two YIG-PZT bilayers. The input-output decoupling is determined by the gap between the microstrips. The input and output microstrip transducers are of 1 mm in width and 18 mm in length. Power is coupled from input to output under FMR in the ME element. The ME element consisted of epitaxial YIG film bonded to PZT. A 110 μm thick LPE-YIG film on a (111) gadolinium gallium garnet substrate was used. The film had a saturation induction 4πM of 1,750 G and FMR line-width of 1 Oe. The PZT disc was initially poled by heating up to 150°C and cooling back to room temperature in an electric field of 10 kV/cm perpendicular to the sample plane. The bilayer was made by bonding the YIG film surface to PZT with 0.08 mm thick layer of ethyl cyanoacrylate, a fast-dry epoxy. The samples were placed between transducers as in Fig. 4 and were subjected to a bias magnetic field *H* either parallel or perpendicular to the sample plane.

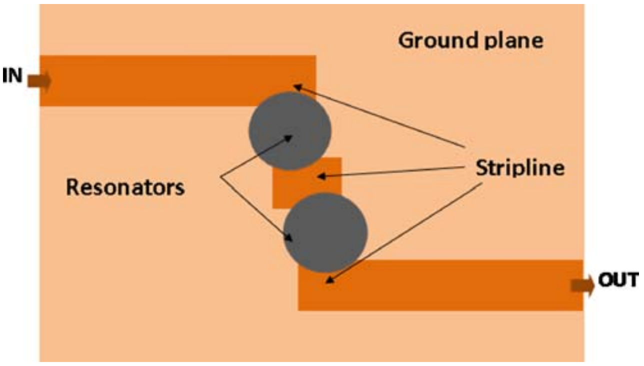

Fig. 4 Schematic diagram showing a dual-cavity magnetoelectric (ME) band-pass filter. The ME resonators consisted of a 110 μm thick (111) yttrium iron garnet (YIG) on GGG substrate bonded to PZT

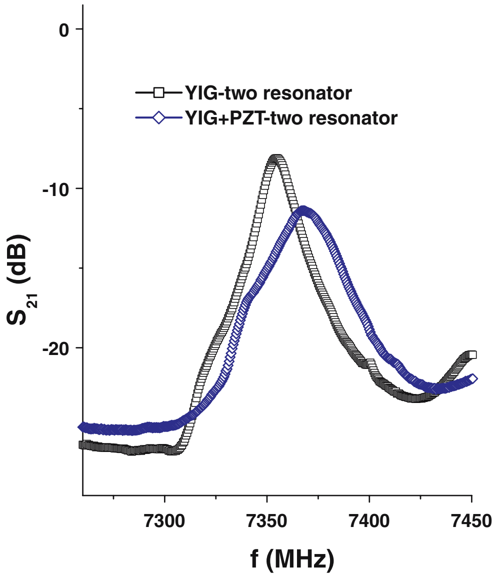

Fig. 5 Insertion loss *L* vs frequency *f* profiles for YIG-only and YIG-PZT filters for *E*=0

The device characterization was carried out with a vector network analyzer (PNA E-8361). An input continuous-wave signal $P_{in}(f)$=1 mW was applied to the filter. The frequency *f* dependence of the insertion loss *L*, i.e., the transmitted power through the ME elements, was measured at 4–10 GHz for a series of *H*. Then the electric field tunability was investigated by obtaining *L* vs *f* profiles with *E* applied across PZT. The frequency dependence of *L* was measured for single and dual YIG-PZT filters and compared with data for YIG-only filters. Representative profiles

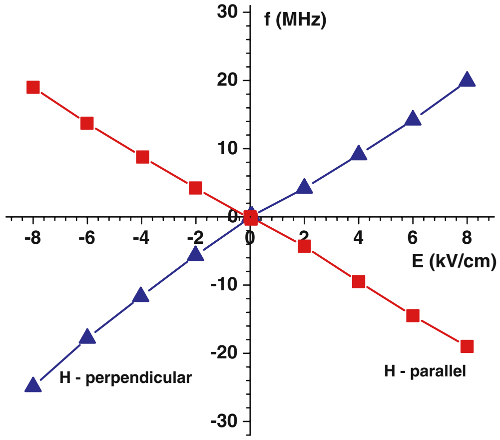

Fig. 6 The dependence of the frequency shifts δf vs *E* for bias magnetic field *H* parallel and perpendicular to plane of the resonators

are shown in Fig. 5 for dual YIG-only and YIG-PZT filters. The profiles for $E=0$ show maximum input-output coupling for a central frequency $f=7.36$ GHz (corresponding to an in-plane bias field $H=1915$ Oe). The loss increases sharply at other frequencies and the off-resonance isolation is 20–25 dB. The YIG-only filter has an insertion loss of 8 dB and $Q=735$. But the insertion loss is much higher for YIG-PZT and Q drops to 100. A significant modification of L vs f profile was observed when E was applied across PZT. The profile was shifted in frequency by δf. Figure 6 shows the dependence of δf on E. One observes a near-linear variation in δf with E. We observed a reversal in δf (up- or down shift) when the direction of E was reversed by reversing the polarity of applied voltage and is attributed to a switch from compressive to tensile strain in YIG. The overall δf for $E=\pm 8$ kV/cm is close to 1% of the central frequency.

5 Conclusion

The nature of microwave ME coupling has been investigated in layered samples of lithium zinc ferrite and PZT. Information on microwave ME effects has been obtained through the effects of external electric field on ferromagnetic resonance for the ferrite. The ME constant estimated from such data is indicative of strong high frequency ME interactions, in agreement with theory. Similar studies on YIG-PZT bilayers reveal a much stronger ME coupling than in LZFO-PZT. A dual magnetic and electric field tunable band-pass filter based on FMR in YIG has been designed and characterized. The filter can be tuned by 1% of the central frequency with a nominal electric field of 8 kV/cm.

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