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Strong magnetoelectric coupling at microwave frequencies in metallic magnetic film/lead zirconate titanate multiferroic composites

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Strong magnetoelectric coupling was observed at microwave frequencies in metallic magnetic film/lead zirconate titanate [Pb(Zr,Ti)O_3] multiferroic composites, in which the magnetic films were either FeCoB or FeGaB with relatively high saturation magnetostriction constants between 40 and 70 ppm and narrow ferromagnetic resonance linewidths of ~20 Oe at 10 GHz. Large electrostatically induced ferromagnetic resonance frequency shifts of 50–110 MHz at ~2.3 GHz were observed. These metallic magnetic film/Pb(Zr,Ti)O_3 multiferroic composites with large electrostatic tunability of the ferromagnetic resonance frequency provide great opportunities for integrated microwave multiferroic devices. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2902316]

Strong magnetoelectric (ME) coupling has been observed in ME composites with ferri/ferromagnetic and ferroelectric materials,[1–6] which has led to many devices at low frequencies such as a picoTesla sensitivity magnetometer,[2] and at microwave frequencies such as electrostatically tunable bandpass filters,[3] bandstop filters,[4] resonators,[5] phase shifters,[6] etc. These ME devices are based upon bulk ME materials, in which strong ME coupling has been achieved.

In order to achieve strong ME coupling at microwave frequencies, the magnetic materials in ME composites need to have a large saturation magnetostriction constant (λ_s) and high permeability,[7] i.e., a low saturation magnetic field (H_k), high saturation magnetization ($4\pi M_s$), and narrow ferromagnetic resonance (FMR) linewidth (ΔH). High quality single-crystal yttrium iron garnet (YIG) material has been the material of choice for microwave ME composite materials and devices.[4–6] These high quality YIG crystals typically need a high temperature synthesis process at temperatures >700 °C, making it difficult for microwave ME devices to be utilized in integrated circuits. In addition, YIG has a very low λ_s of 0.2 ppm, which is not ideal for achieving strong ME coupling. An alternative microwave magnetic material for ME composites is the class of metallic magnetic films, of which the eddy current loss at microwave frequencies can be negligible when the thickness of the film is less than its skin depth. Metallic magnetic films have their own advantages as a microwave magnetic material compared to ferrites. These metallic magnetic films can have large λ_s,[8] relatively low ΔH,[9–13] high $4\pi M_s$ of up to 24.5 kG,[9] high squareness of ~100%,[10–13] high self-biased FMR frequencies in the gigahertz range,[10] and low processing temperature.

Metallic magnetic films with excellent magnetic softness and low ΔH, however, typically have very low λ_s. For example, several of the most well known soft magnetic films such as Permalloy ($Ni_{81}Fe_{19}$ wt %), sendust (FeAlSi), CoCrTa, CoZrNb, FeXN films, etc., all have nearly zero λ_s. Soft magnetic thin films based on $Fe_{70}Co_{30}$, such as the $(Fe_{70}Co_{30})_{100-x}N_x$ (Ref. 10) and $(Fe_{70}Co_{30})_{100-x}B_x$,[12] exhibit excellent soft magnetic properties with low ΔH, low H_k, together with a decent λ_s of 40–45 ppm.[10,13] Most recently, we developed FeGaB films with a narrow ΔH of 15 Oe at x-band (10 GHz), a low H_k of ~20 Oe, and a large λ_s of 70 ppm.[13]

In this work, we investigated the ME coupling of metallic magnetic film/Pb(Zr,Ti)O_3 (or PZT) multiferroic composite materials at microwave frequencies. Both FeCoB/PZT and FeGaB/PZT composite materials were studied. Strong ME coupling was observed at microwave frequencies showing large FMR frequency shifts of 50–110 MHz at ~2.3 GHz. The metallic films used in this study are described in Table I. FeGaB films were cosputtered from FeGa and B targets with detailed deposition conditions available in Ref. 13. FeCoB films were cosputtered from FeCo and B target,[12] similar to the FeGaB films deposition conditions.

The 50 nm thick FeCoB films on 0.1 mm thick glass substrates and 50 nm $Fe_{72}Ga_{10}B_{18}$ films on 0.24 mm Si substrates were each epoxy bonded to a 0.5 mm thick Pb(Zr,Ti)O_3 ceramic beam with a dimension of $35\times 4\ \mathrm{mm}^2$ (PIC151, PI Ceramic Co.) to form the composites. When the PZT is subjected to a transverse voltage V across its thickness t, a biaxial strain of $\Delta l/l=d_{31}(V/t)$ is induced, where d_{31} is the piezoelectric charge coefficient. This phenomenon is known as the inverse piezoelectric effect. Deformation of the PZT will lead to strain in the metallic magnetic films (FeCoB or FeGaB) which are tightly bonded to the PZT. This PZT induced strain in the magnetic film will lead to a change in its in-plane anisotropy field due to the well known inverse magnetoelastic effect, which is reflected in the shift in the FMR frequency of the magnetic film at microwave frequencies.

TABLE I. Summary of magnetic properties of films used in this study.

Composition [at %]	$4\pi Ms$ (kG)	H_c (Oe)	H_k (Oe)	ΔH at x-band (Oe)	λ_s (ppm)
$Fe_{75}Co_{15}B_{10}$	17	<1	19	<20	40
$Fe_{72}Ga_{10}B_{18}$	12	<2	21	15	50–70

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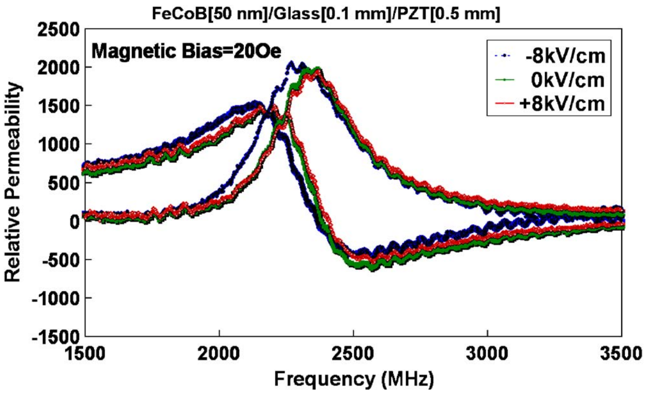

FIG. 1. (Color online) Permeability spectrum of FeCoB/PZT composite vs electrostatic bias at a fixed magnetostatic bias of 20 Oe (~2.3 GHz).

A broad band permeability measurement technique was used to measure the permeability of the composites, in response to both an in-plane magnetostatic field along the easy axis direction and a transverse electrostatic bias field across the thickness of the PZT slab. An electrostatic bias between +/−8 kV/cm was applied to the PZT layer by biasing the electrodes at different voltages between −400 V and +400 V. The voltage was swept from −400 to +400 V and back to −400 V to observe the hysteretic behavior of the polarization of the PZT.

The measured permeability spectra of the FeCoB/PZT composite film at different applied voltage across the PZT layer are shown in Fig. 1 under a constant bias field of 20 Oe. A FMR frequency shift of 50 MHz was obtained at 2.3 GHz together with a negligibly small change of the initial permeability. The FeGaB composite was biased at 35 Oe to observe the ME effect at the same frequency. The permeability spectra of the FeGaB/PZT ME composite are shown in Fig. 2 at different applied voltages across the PZT layer. The measured FMR frequency shift at +/−8 kV/cm for the FeGaB/PZT composite is ~110 MHz, which is much larger than the 50 MHz FMR frequency shift for the FeCoB/PZT ME composites. It is notable that the 110 MHz FMR frequency shift is comparable to the largest FMR frequency shift reported.[14,15]

In both the FeCoB/PZT and FeGaB/PZT ME composites, the application of the negative electric field causes a downward shift in the permeability spectrum, while a positive field produces the opposite response. This behavior could be explained by considering the electric field induced the elastic deformation of the PZT and its effect on the anisotropy field. A tensile strain in the metallic magnetic films with a positive magnetostriction constant leads to an increase in the effective in-plane anisotropy field, whereas a compressive strain leads to a decrease. The effective in-plane magnetic field (ΔH_{eff}) in a magnetic film/ferroelectric ME composite bilayer generated from the stressed mediated ME coupling can be expressed as follows (cgs unit): $\Delta H_{\mathrm{eff}} = 3\lambda_s Y d_{31} \times E / M_s$, where M_s is the saturation magnetization, and Y is the Young's modulus of the magnetic film; d_{31} is the piezoelectric coefficient and E is the electric field across the thickness direction of the ferroelectric substrate. Since the FMR phenomenon is used for many microwave magnetic devices,[7,8,16] the tunable range of the FMR frequency is a representation of the tunability of many kinds of such microwave ME devices. The tunable FMR frequency range (Δf_{FMR}) induced by the effective in-plane magnetic field (ΔH_{eff}) of such microwave ME devices, with metallic magnetic film deposited directly on a ferroelectric substrate, can be derived to be $\Delta f_{\mathrm{FMR}} = \gamma\sqrt{\mu_i}\Delta H_{\mathrm{eff}}$, where γ is the gyromagnetic constant ~2.8 MHz/Oe, and μ_i is the initial relative permeability of the magnetic film. We can readily get $\Delta f_{\mathrm{FMR}}/f_{\mathrm{FMR}} = \Delta H_{\mathrm{eff}}/H_{\mathrm{DC}}$, where H_{DC} is the net in-plane uniaxial anisotropy field of the magnetic film, and $\mathbf{f}_{\mathrm{FMR}}$ being the FMR frequency, or operating frequency in many different microwave devices.[7,8,16]

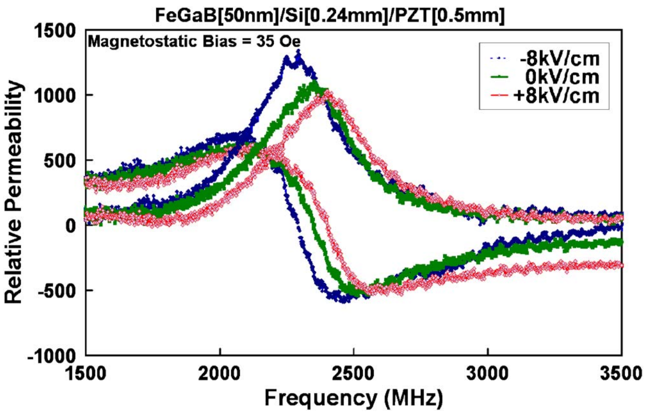

FIG. 2. (Color online) Permeability spectrum of FeGaB/PZT composite vs electrostatic bias at a fixed magnetostatic bias of 35 Oe (~2.3 GHz).

For the microwave ME composite with $Fe_{75}Ga_{13}B_{12}$ (at %) film on a PZT slab, we will get a ΔH_{eff} of ~30 Oe and a large tunable FMR frequency range of $\Delta f_{\mathrm{FMR}} = 650$ MHz at 2.5 GHz when a moderate electric field of 800 V/mm is applied across the PZT layer by the predicted FMR peak shift derived from the Landau–Lifshitz–Gilbert equation.[17] This corresponds to a large tunability of $\Delta f_{\mathrm{FMR}}/f_{\mathrm{FMR}} = 28\%$. This large tunability in the FeGaB/PZT bilayer ME composite material is one order of magnitude higher than what has been reported so far for the ME composites, which are typically in the range of $\Delta f_{\mathrm{FMR}}/f_{\mathrm{FMR}} = 0.5–2.5\%$ with $\Delta f_{\mathrm{FMR}} = 30–125$ MHz.[6,14,15]

For the ME composites we reported, there is always a substrate for the metallic magnetic film, i.e., FeCoB/glass/PZT and FeGaB/Si/PZT. The presence of a substrate for the metallic magnetic films as well as the nonideal strain coupling at the interface of the ME composite may explain the discrepancy between the observed and predicted performances. Putting metallic magnetic films onto PZT directly is hindered by the large magnetic linewidth of the metallic magnetic films, which we are working on right now.

In conclusion, we have demonstrated the strong ME coupling at microwave frequencies in metallic magnetic film/PZT ME composites, namely the FeCoB/PZT and FeGaB/PZT composites. Large FMR frequency shifts of 50 and 110 MHz are observed in the FeCoB/PZT and FeGaB/PZT composites, respectively, when tuned with an e field of +/−8 kV/cm. Large FMR frequency tunability of 28% at 2.5 GHz is predicted for FeGaB/PZT ME composite when FeGaB is deposited directly on PZT, which provides tunable microwave ME devices with large tunability.

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