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Tunable fringe magnetic fields induced by converse magnetoelectric coupling in a FeGa/PMN-PT multiferroic heterostructure

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The fringe magnetic field, induced by magnetoelectric coupling in a bilayer Fe-Ga/Pb(Mg_{1/3}Nb_{2/3}) O₃-PbTiO₃ (PMN-PT) multifunctional composite, was investigated. The induced external field is characterized as having a butterfly hysteresis loop when tuned by an applied electric field. A tuning coefficient of the electrically induced fringe magnetic field is derived from the piezoelectric and magnetostrictive properties of the composite. A measured maximum tuning coefficient, 4.5 Oe/ (kV cm⁻¹), is found to agree well with theoretical prediction. This work establishes a foundation in the design of transducers based on the magnetoelectric effect. © 2011 American Institute of Physics. [doi:10.1063/1.3672822]

I. INTRODUCTION

Magnetoelectric (ME) coupling in multiferroic (MF) piezoelectric/magnetostrictive laminate heterostructures has received much attention in recent years, in part due to the use of these structures as electric field tunable phase shifters and other microwave components.^{1,2} These structures exhibit multifrequency operability³ and have electronically tunable magnetic properties.^{4,5} The laminate multiferroic heterostructure is preferable in applications due to its low-power consumption, small profile, and planar design. Previous work on MF composites has been based on direct or converse magnetoelectric coupling, leading to different applications.^{6,7} In direct ME coupling, an applied magnetic field on the magnetostrictive component results in strain exerted upon the piezoelectric element(s), which then induces electric polarization in the piezoelectric element(s). A representative application includes magnetic field sensors.⁸ However, under converse magnetoelectric (CME) coupling, an electric field is applied to the piezoelectric, inducing strain, which in turn strains the magnetostrictive element(s) inducing magnetization in the magnetostrictive element(s). Coupling between the piezoelectric and magnetostrictive elements depends on the connectivity scheme employed in the heterostructure, as well as the coupling coefficients of the distinct elements and their volume fractions.⁹ Additionally, the ME effect can be either static or dynamic, depending upon whether the applied fields are DC or AC.¹⁰ Although the static CME effect has been the focus in the past years, investigation of the fringe magnetic fields of a multiferroic heterostructure has been largely overlooked. Recently, the tuning of ferrite phase shifters using the induced magnetic fields of a Terfenol-D/PMN-PT laminated transducer was demonstrated,¹¹ where the fringe field played the important role of active tuning of the phase shifter while reducing power consumption and increasing switching speed. The Fe-Ga/

^{a)}Author to whom correspondence should be addressed. Electronic mail: fitchorov.t@gmail.com. PMN-PT laminate heterostructure discussed here has also shown potential for electric field assisted tuning of magnetization and other magnetic parameters.¹²

II. EXPERIMENT

A bilayer laminated heterostructure, consisting of a polished polycrystalline FeGa alloy layer with dimensions of 15 mm \times 10 mm \times 0.4 mm and single crystal PMN-PT layer with dimensions of 15 mm \times 10 mm \times 0.5 mm, was assembled. The PMN-PT substrate was obtained commercially. A fast-dry ethyl cyanoacrylate adhesive was used as a bonding material between the two elements, with thickness of 20-30 μ m. The PMN-PT layer was poled along the [011] direction, which is normal to the 15 mm \times 10 mm surface. This singlecrystal PMN-PT possesses a piezoelectric coefficient $d_{33} < 500$ pC/N along the poling direction, and a transverse extension (TE) mode piezoelectric coefficient $d_{31} = 2300 \text{ pC}/$ N. This type of cut was chosen because of its superior piezoelectric performance.¹³ The two faces, with normal vectors parallel and antiparallel to [011], were plated with gold electrodes. The FeGa/PMN-PT heterostructure was positioned between the pole pieces of an electromagnet, so that the direction of the bias magnetic field coincided with the longitudinal direction of the composite. The bias field was incremented in steps of 25 Oe, and then for each bias field, an electric field applied across the PMN-PT was varied over a range of -8 kV/cm to 8 kV/cm. The measurement of the fringe magnetic field exterior to the FeGa layer along a direction parallel to the applied bias magnetic field was accomplished using a Gauss meter. A schematic of the multiferroic composite can be seen in Fig. 1(a). Data were also collected for the fringe field with an out-of-plane bias magnetic field.

III. RESULTS AND DISCUSSION

The strain (ε) versus electric field (E) of the PMN-PT component was obtained for the d₃₁ direction and presented in Fig. 1(b). The non-linear behavior of electrically induced



FIG. 1. (Color online) The graphical representation of the multiferroic transducer (a), as well as the piezoelectric response of the PMN-PT component (b).

strain seen in Fig. 1(b) is due to the hysteretic behavior of electronic polarization in the PMN-PT. The strain coercivity can be defined as the electric field at which the measured strain in the PMN-PT reverses polarity. The coercivity is observed to be between 2 kV/cm and 3 kV/cm. The relatively large magnitude of coercivity can be attributed to the difficulty in electric field-assisted domain switching, since the two possible domains are mostly perpendicular to each other.¹⁴ The maximum positive strain is measured at |E| = 2 kV/cm.

The magnetostrictive element, i.e., FeGa alloy of the heterostructure, can be synthesized by the substitution of Ga atoms for Fe atoms in the body-centered-cubic Fe lattice.¹⁵ The addition of Ga enhances the magnetocrystalline anisotropy of the lattice, which results in the stabilization of an easy axis of magnetization along the $\langle 100 \rangle$ direction.¹⁶ The elastic response of FeGa is also anisotropic, increasing in the direction of higher electron density distribution, which is maximized in single crystals and strongly textured polycrystalline forms.¹⁷ The alloy used here is stress-annealed, which improves the magnetostrictive strain through a build-up of internal stress, as compared to the FeGa without pre-stress treatment. The FeGa used in this experiment is additionally strongly textured along the $\langle 110 \rangle$ crystallographic direction, which coincides with the longitudinal dimension of the structure (Fig. 2). A sufficiently strong compressive pre-stress along $\langle 100 \rangle$ is typically required to overcome the magnetocrystalline anisotropy. Once the anisotropy is overcome, the



FIG. 2. The x-ray diffraction scans along the longitudinal and out-of-plane directions of the magnetostrictive FeGa alloy.

magnetic moments are free to align perpendicular to the axis of pre-stress.¹⁸ An applied magnetic field along the longitudinal direction rotates magnetic moments along the magnetization direction, which leads to maximum magnetostriction. It should be noted that the addition of pre-stress results in not only an increased magnetostriction but also an increased bias magnetic field needed for reorienting the magnetic moments in the direction of applied magnetic field.¹⁹ Therefore, there is a trade-off between larger tuning of the fringe field, depending on the magnetostriction, and smaller bias magnetic fields. The B-H loop and magnetostriction versus applied magnetic field for FeGa are shown in Figs. 3(a) and 3(b), respectively. The B-H curve has a shape typical of a soft magnetic material and exhibits saturation at bias fields greater than 500 Oe. Accordingly, the magnetostriction is also saturated near H = 500 Oe. The large saturation field and small slope of the magnetostriction curve are attributed to a weak $\langle 100 \rangle$ texturing in the longitudinal dimension, compared to that in the out-of-pane (Fig. 2).



FIG. 3. The (a) magnetic induction loop and (b) magnetostriction curve of the FeGa alloy as functions of the in-plane bias magnetic field.



FIG. 4. (Color online) (a) Measured fringe magnetic field at the edge of the transducer, as well as (b) tuning coefficient of the fringe field for an in-plane bias magnetic field.

The fringe magnetic field as a function of applied electric field is shown in Fig. 4(a), with the Gauss probe positioned as illustrated in Fig. 1(a). An important characteristic of the external field plot is a pronounced butterfly hysteresis between -4 kV/cm and 4 kV/cm, with two maximums at ± 2 kV/cm. A direct link between the stress-induced magnetic field and electrically induced strain can be observed by comparing the plots in Fig. 1(b) and Fig. 4(a), as both curves follow the same pattern of hysteresis. Two distinct maximums of the fringe field for a given bias field, as seen in Fig. 4(a), coincide with the peaks of the electrically induced strain in the PMN-PT. The presence of fringe magnetic field at zero applied electric field is due to the magnetization induced in the FeGa alloy with the applied magnetic field. It is pointed out that the effect of the demagnetizing field, which opposes any magnetization in the direction of applied bias field, must be considered. The demagnetization factor for a ferromagnetic rectangular prism can be determined analytically.²⁰ For the given dimensions, the demagnetization factor in the magnetization direction is $N_1 \approx 0.035$. To obtain an effective bias field, the demagnetizing field is subtracted from the applied magnetic field.

The electric field-tunable behavior of the stress-induced fringe field, $H^f_{\sigma}(V)$, can be quantified using the following metric:

$$\alpha = \frac{H_{\sigma}^{t}(V_{0}) - H_{\sigma}^{t}(0)}{\Delta E},$$
(1)

where $\Delta E = E_0 = 8 \text{ kV cm}^{-1}$. The coefficient in Eq. (1) is plotted as a function of bias field in Fig. 4(b). The induced fringe field reaches a maximum, $\alpha = 4.5 \text{ Oe}/(\text{kV cm}^{-1})$, at a bias of 125 Oe. A similar response of the induced internal field as a function of applied bias magnetic field was observed previously.¹² A CME tuning coefficient can be derived from the constitutive relations of the piezoelectric and magnetostrictive components, and the boundary conditions for the stress and strain at the interface of the two elements of the heterostructure.¹ Following a similar procedure, the tuning coefficient for the fringe field is given by

$$\alpha = \frac{\partial H_1}{\partial E_3} = \frac{d_{31}q_{11}^*}{\left(\mu_0 - \mu_{11}^T\right) \left(s_{11}^H + \frac{t_m}{t_p} s_{11}^E\right) + q_{11}^* q_{11}}, \quad (2)$$

where H₁ is the component of the induced fringe magnetic field parallel to the longitudinal dimension of FeGa, E₃ is the applied electric field, d₃₁ is the piezoelectric coefficient of PMN-PT, q_{11} is the piezomagnetic coefficient, q_{11}^* is the stress sensitivity of the magnetic layer, s_{11}^H and s_{11}^E are the elastic compliance coefficients at constant magnetic and electric field, and μ_{11}^T is the permeability of FeGa at constant stress. In Eq. (2), $t_m = 0.4$ mm and $t_p = 0.5$ mm are the thicknesses of the magnetostrictive and piezoelectric components, respectively. We have assumed that $q_{11}^* \approx q_{11}$ for the stress and bias field under consideration. Using $d_{31} = -2300$ pC/N, $q_{11} = 9$ nm/A, $\mu_{11}^T = 15 \mu_0$, $s_{11}^H = 15$ pm²/N, and $s_{11}^E = 126$ pm²/N, the fringe field tuning coefficient is predicted to be 13 $Oe/(kV cm^{-1})$. The piezoelectric coefficient is obtained from the slope of Fig. 1(b), i.e., $d_{31} = \partial \varepsilon / \partial E$. The piezomagnetic coefficient is determined from the λ -H curve in Fig. 3(b), i.e., $q_{11} = \partial \lambda / \partial H_i$, where $H_i = H_a - N_1 M$ is the internal field. The result predicted by Eq. (2) is within the same order of magnitude as the experimentally determined value of 4.5 $Oe/(kV cm^{-1})$. The theoretical coefficient is derived using the magnetic field at the surface of the FeGa layer, and since the magnetic layer can be viewed as an approximation of a large magnetic dipole, the external magnetic field is expected to decrease with increasing distance from the lateral side of the heterostructure. Therefore, it is acceptable that the slight discrepancy in tuning coefficient between the measured and predicted value arises from an error in measuring position.

The experimentally determined fringe field tuning coefficient for an out-of-plane bias is shown in Fig. 5. There is an upward trend with increasing bias field. However, the magnitude of the coefficient is much lower in comparison to the tuning coefficient obtained with an in-plane bias magnetic field. It should be noted that this field configuration corresponds to a transversely magnetized-transversely poled (T-T) mode. In this case, the coupling is pronouncedly determined by both the piezoelectric coefficient d_{33} and the out-of-plane magnetostrictive coefficient (λ_{oop}) . It is clear that either d_{33} or λ_{oop} is much smaller than d_{31} or the in-plane magnetostrictive coefficient (λ_{ip}) . Additionally, the bias magnetic field is transverse to the measured fringe field, intruding an extra torque on the magnetic dipoles. It also acts to reduce the in-plane fringe field. As a result, an observation of low fringe field and ME



FIG. 5. Experimentally determined fringe field tuning coefficient for an outof-plane applied bias field.

coupling is predictable. Furthermore, the maximum tuning coefficient occurs at an applied field of more than 2000 Oe for an out-of-plane measurement. This can be explained by the large demagnetization factor of 0.9 in the thickness dimension.

IV. CONCLUSIONS

It is concluded that the FeGa/PMN-PT laminate composite can produce electrically tunable external magnetic field. When the external field is plotted as a function of applied electric field for a given bias magnetic field, we observe a butterfly hysteresis that is also typical of the electrically induced strain in the piezoelectric PMN-PT. This is suggestive of the strain-mediated magnetoelectric effect. It is shown that an in-plane bias magnetic field produces the largest in-plane fringe magnetic field. This paper has established a foundation for future work on the tuning of devices using a fringe magnetic field induced by the magnetoelectric effect. Further work needs to be done to determine the uniformity of the fringe field, especially in the presence of more than one multifunctional composite in a planar configuration.

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