Is the magnetoelectric coupling in stickup bilayers linear?

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The magnetoelectric (ME) characterization of stickup bilayers Ni\textsubscript{0.8}Zn\textsubscript{0.2}Fe\textsubscript{2}O\textsubscript{3}–Pb(Zr, Ti)O\textsubscript{3} and trilayers of Ni\textsubscript{0.8}Zn\textsubscript{0.2}Fe\textsubscript{2}O\textsubscript{3}–Pb(Zr, Ti)O\textsubscript{3}–Ni\textsubscript{0.8}Zn\textsubscript{0.2}Fe\textsubscript{2}O\textsubscript{3} are discussed. The ME interactions in the trilayers were found much stronger than that of bilayers prepared with the same technique. The maximum transverse ME voltage coefficient of trilayer can reach 430 (mV/cm Oe) under a magnetic field $H=215$ Oe. This value is much closer to the theoretical estimate for the ME coupling for bilayers. The frequency dependence of $\alpha_E$ also reveals the difference of the elastic and magnetoelectric coupling between the bilayers and trilayers. Analysis suggests that a more approach linear response of the piezoelectric layer to the magnetostrictive of the ferrite may be responsible for the significant enhanced ME effect in the trilayers.  

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I. INTRODUCTION

The magnetoelectric (ME) effect has stimulated a continual interest for dozens of years due to its potential applications in the manufacture of sensors.\textsuperscript{1,2} Various kinds of ME materials have been investigated in recent years. They include some manganese perovskites, such as HoMnO\textsubscript{3} or YMnO\textsubscript{3},\textsuperscript{3–5} bulks composing both ferroelectrics and ferromagnet,\textsuperscript{6–8} and layered materials consisting of ferromagnetic and ferroelectric layer(s).\textsuperscript{9–13} Two kinds of layered ME composites have been developed so far, tape-cast multilayers of thick films and stickup laminates. Current investigations focus on the stickup layered composites since it needs not to consider both the loss of dielectricity of the ferroelectrics due to the conductance of the ferromagnet composed and the interlayer diffusion, which cannot be neglected for tape-cast multilayers.\textsuperscript{14} Results reported show that the ME voltage coefficients $\alpha_E(=\delta E/\delta H)$ in stickup bilayers is usually much less than that of theoretical estimate. That was believed to result from the weakness of the elastic coupling at the interface(s) due to using glue to bond the laminates. However, the $\alpha_E$ in trilayers made from the same materials and with the same technique are often several times of that in the bilayers and closer to the theoretical estimate for the bilayers.\textsuperscript{14,15} Considering that (1) the electromagnetic coupling in layered composite occurs via interlayer mechanical stress, (2) there is only elastic connecting but no electric connecting in the stickup laminates, thus it is difficult for us to attribute the much enhanced ME coupling in stickup trilayers only to some possible improvement of interface coupling. The improvement of the linearity of the mechanical strain may be a more important factor for the increase of the ME coupling in the layered composite.

Here, we present an investigation of ME effect on layered Ni\textsubscript{0.8}Zn\textsubscript{0.2}Fe\textsubscript{2}O\textsubscript{3} (NZFO) and PZT-NZFO/NZFO-PZT-NZFO are much larger than that of the bilayers NZFO-PZT. Then we will demonstrate that the elastic strain in trilayers is quite different from that in bilayers. Through simple analysis, we will deduce a more linear response of the piezoelectric layer to the magnetostrictive of the ferrite is responsible for the significant enhanced ME effect in the trilayers.

II. SAMPLE CHARACTERIZATION

Disks of the ferrite NZFO used were prepared from nanoparticles obtained by sol-gel techniques as described previously.\textsuperscript{16} The powder was pressed into rods of 10 mm in diameter and about 8 mm in length and treated at 70 MPa and 900 °C for 1 h with a hot-pressing facility. The hot-pressed samples were further sintered at 950–1380 °C to get NZFO rods with different compactness. The rods then were cut into pellets with 0.7 mm thick. The samples are characterized by x-ray diffraction (XRD) in a rotating anode diffractometer. The patterns of XRD revealed the absence of any impurities except spinel structure.

It is well known that the dynamic ME coupling is directly proportional to the piezomagnetic coefficient defined by $q=\delta\lambda/\delta H$. Magnetic characterizations of NZFO included magnetization and magnetostriction $\lambda$. Figure 1 shows the magnetic field $H$ dependent magnetization and magnetostrict-
trend to saturation when the primary axes larger than that along the directions for epoxy layer ranged from 0.01 to 0.02 mm.

The thickness of perpendicular to the sample plane. It was then bonded to NZFO. PZT was first poled by heating to 150 °C and cooling back to room temperature in an electric field of 30–50 kV/cm perpendicularly to the sample plane and perpendicular to the sample plane and perpendicular to

III. EXPERIMENTS AND DISCUSSION

For ME characterization, we measured the electric field produced by an alternating magnetic field applied to the composite. The samples were positioned in a measurement cell and subjected to a bias magnetic field \( H \) and an alternating current (ac) magnetic field \( E \). The voltage \( \delta V \) across the sample was amplified and measured with an oscilloscope or a lock-in-amplifier. The ME voltage coefficient was estimated from

\[
\alpha_{E,31} = \frac{\delta V}{\delta H} = \frac{\delta V}{t \delta H},
\]

where \( t \) is the thickness of PZT. The measurements were done for two different field orientations. The transverse coefficient \( \alpha_{E,31} \), which is often much larger than the longitude one \( \alpha_{E,33} \), was measured for the electric fields \( H \) and \( E \) along the direction parallel to the sample plane and perpendicular to \( \delta E \).

Figure 2 shows the \( H \) dependence of the \( \alpha_{E,31} \) for the trilayers NZFO-PZT-NZFO composed by NZFO with different \( T_s \) at room temperature and an ac magnetic field at a frequency of 100 Hz. It shows that \( \alpha_{E,31} \) appears as a peak with increasing bias field. But the peak value \( \alpha_{E,31,\text{max}} \) increases with increasing the \( T_s \) of NZFO and trends to saturation when \( T_s \) goes to about 1400 °C, as shown in the inset of Fig. 2. The former has been carefully discussed elsewhere. Here we focus just on the later. We find that the \( T_s \)-dependent \( \alpha_{E,31,\text{max}} \) is very similar to the \( T_s \)-dependent compactness, as also shown in the inset, which also increases with increasing \( T_s \) and trends to saturation. The compactness was defined as the ratio of the measured density to the density estimated from structure parameters that was obtained from XRD data.

Figure 3 shows the \( H \) dependence of the \( \alpha_{E,31} \) for the bilayers NZFO-PZT composed by NZFO with different \( T_s \) with the same conditions of measurement as we have done for trilayers. The inset shows the \( T_s \)-dependent \( \alpha_{E,31,\text{max}} \) for the bilayers, which shows quite fluctuated.

Comparing Fig. 3 with Fig. 2, we find that the ME coupling in bilayers is much weaker than that in trilayers composed by the same temperature sintered NZFO. Additionally, the value of \( \alpha_{E,31} \) for the trilayer composed with 1380 °C...
sintered NZFO is closer to the theoretical estimate for bilayers than that measure from the bilayer also composed with

\[
E_{E, 31} = \frac{-v(v-1)\beta d_{31}(m q_{11} + m q_{21})}{(m s_{11} + m s_{12})E_{33} v + (p s_{11} + p s_{12})E_{33}(1-v) - 2(\beta d_{31})^2(1-v)},
\]

where \(v = p_v / (p_v + m_v)\) and \(p_v\) and \(m_v\) denote the volume of piezoelectric phase and magnetostrictive phase, \(m S_{ij}\) and \(m Q_{ij}\) are compliance and piezomagnetic coefficients; \(p S_{ij}\) and \(p d_{ij}\) are compliance and piezoelectric coefficients; and \(\epsilon_{kn}\) is the permittivity matrix.

However, the measured \(E_{E, 31}\) of most stickup ME bilayers is much less than the calculated value from Eq. (1).\(^{19}\) So is it for the present case, as can be seen in Fig. 4. We ever

devolved a relatively comprehensive theory in which the composite is considered as a homogeneous medium with piezoelectric and magnetostrictive subsystems.\(^{20}\) A novel technique was employed to take into account the actual interface conditions, i.e., by introducing an interface coupling parameter \(k\). \(k=1\) means ideal interface coupling. Using open circuit conditions, one obtains the following expressions for the transverse ME voltage coefficients:

\[
E_{E, 31} = \frac{-k v (v-1)\beta d_{31}(m q_{11} + m q_{21})}{(m s_{11} + m s_{12})E_{33} v + (p s_{11} + p s_{12})E_{33}(1-v) - 2(\beta d_{31})^2 k(1-v)},
\]

For the bilayer composed with 1380 °C sintered NZFO, \(k\) is about 26%. For most ME bilayers reported, \(k\) is inter-\(\varepsilon\)tive in 20%–40%.\(^{19}\) And \(k\) value cannot be effectively improved no matter what kind of glue has been used. On the other hand, the \(E_{E, 31}\) for the trilayer composed by 1380 °C sintered NZFO can reach 429 mV/cm Oe. This value is almost three times of that for the bilayers made by the same technique. So, it is doubtful to attribute the weakness of ME coupling in bilayers only to nonideal interface coupling. For a good model that can be practically used, it is necessary to take both nonideal interface coupling and flexural deformations of the layers into account.

The electromagnetic coupling occurs via interlayer mechanical stress in layered composite, so, whether the me-
Mechanical strain or elastic coupling between or among the layers is important. Since the rigidity, the Young’s modulus and thermal property of the piezomagnetic used is not necessarily to be compatible with that of the piezoelectric material, a nonlinear elastic coupling could occur in bilayers. The possible nonlinear mechanical strains are shown in Fig. 5, where Fig. 5(a) shows a distortion of PZT due to its lack in rigidity, and Figs. 5(b) and 5(c) show some typical flexural deformation due the difference of Young’s modulus between the PZT and the ferrite. Nonlinear distortion of the piezoelectric layer could much reduce ME effect considering about the tensor characteristic of piezoelectric coefficient. While the elastic strain in trilayers should be linear since the mechanical stress in those is symmetry about the center (piezoelectric) layer. Thus we have reason to attribute the weakness of ME coupling in bilayers partly to their nonlinear mechanical strains.

More possible evidence about the nonlinear distortion in bilayers could be obtained by comparing the $\alpha_{E,31}$ versus frequency of the bilayers and trilayers to that of theoretical values.

The frequency dependence of $\alpha_{E,31}$ was studied for the bilayer and trilayer composed by 1380 °C sintered NZFO. The bias field was set at the field corresponding to maximum in $\alpha_{E,31}$ in Fig. 4. The voltage coefficients were then measured as the frequency $f$ of the ac field $\partial H$ was varied. The $\alpha_E$ vs $f$ profiles of the bilayer and the trilayer at room temperature for transverse fields are shown in Fig. 6. Upon increasing $f$, $\alpha_{E,31}$ increase gradually with a minor peak at 190 and 237 kHz for the bilayer and trilayer, respectively. At higher $f$, we observe a rapid increase in $\alpha_{E,31}$ to a maximum of 2.916 V/cm Oe at 275.5 kHz and 8.408 V/cm Oe at 339.4 kHz for the bilayer and trilayer, respectively. A similar $\alpha_E$ vs $f$ profiles occurred for longitudinal fields. The peaks occur at the same frequency as for the transverse fields, but with a much smaller maximum $\alpha_E$ compared to the transverse fields.

We identified the resonance at $f_r$ with electromechanical resonance (EMR) in PZT. The resonance is characterized by a discontinuity in impedance versus $f$ data. It is obvious from Fig. 6 that EMR leads to a very significant enhancement in the strength of ME coupling, by a factor of about 20 compared to the low frequency value (Fig. 4). The resonance in ME coefficient occurs when the ac field is tuned to EMR.

IV. CONCLUSION

The nature magnetoelectric interactions have been investigated in trilayers and bilayers consisting of sol-gel prepared Ni$_{0.5}$Zn$_{0.5}$Fe$_2$O$_4$ and piezoelectric PZT. Maximum ME coupling is measured when the layered processed by hot-pressing and sintered at 1380 °C. It was found that the ME coupling in the trilayers is 3.5 times of that in the bilayers and more approach to the theoretical estimate for bilayers. The electromechanical resonance frequency in the trilayer is also closer to the theoretical estimate for bilayers than that measured in the bilayer. These suggest that the ME coupling in trilayers is much better in linearity than that in bilayers.

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