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Dependence of the magnetoelectric coupling in NZFO–PZT laminate composites on ferrite compactness

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Abstract

The synthesis and magnetoelectric (ME) characterization of bilayers of $\mathrm{Pb(Zr, Ti)O_3}$ (PZT) and hot-pressed ferrite $\mathrm{Ni_{0.8}Zn_{0.2}Fe_2O_3}$ (NZFO) are discussed. Very strong ME interactions are measured for the bilayers. The bilayers exhibit superior ME coupling compared to thick film composites prepared with the tape-casting technique. The ME coupling was found to increase with increasing sintering temperature of the ferrite. This supports the idea that the mechanical transport efficiency of the magnetostriction of the ferromagnetic layer is one of the key factors in ME coupling in laminate composites.

1. Introduction

There has been a continual interest in the study of magnetoelectric (ME) materials for many years due to their potential applications in the manufacture of sensors [1, 2]. The ME effect can behave like a magnetic field-induced dielectric polarization [3]. The strength of the effect is generally expressed as the ME voltage coefficient α_E ($\alpha_E = \delta E/\delta H$). Previous reports have indicated that multilayer (ML) composites consisting of piezoelectric and ferromagnetic films or flakes can show a larger α_E than bulk composites since the former can overcome the loss of dielectricity of the piezoelectrics resulting from leakage current though the high conductance of the included ferromagnet [4]. In a ML, ME coupling occurs via mechanical stress. Many layered composites have been investigated and several techniques have been developed to prepare MLs in recent years [5–13].

The first developed layered ME composites were those fabricated by the technique of tape-casting. But the measured α_E s of tape-cast MLs were much smaller than theoretical estimates [9]. Although x-ray diffraction (XRD) shows no impurity phases, the ME parameters

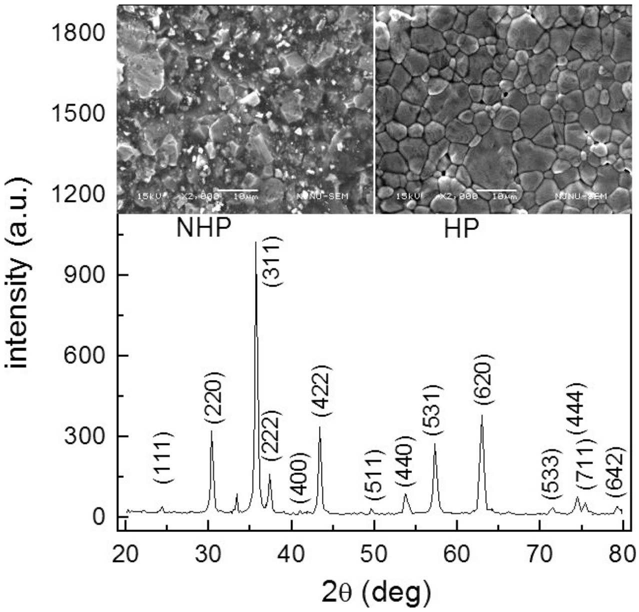

Figure 1. XRD pattern for hot-pressed and 1380 °C sintered $Ni_{0.8}Zn_{0.2}Fe_2O_4$. Inset: scanning electronic microscopy photos for both hot-pressed (HP) and not hot-pressed (NHP) samples sintered at 1380 °C.

were found to be very sensitive to sintering temperature for a ML. It is believed that interface diffusion is the possible cause of poor ME effects [14]. It was shown recently that in addition to strain effects produced at the interface between the piezoelectric and ferromagnetic materials, ME coupling can also be induced by interface bonding [15]. Laminate ME composites formed by bonding ferromagnet and piezoelectric laminates have also been developed. Most laminate ME composites are bilayers or trilayers, so they are expected to avoid much interface diffusion. However, it was found that the α_E s of the laminate composites were also much smaller than theoretical values. It was supposed that the glue used to bond laminates weakened the elastic coupling at the interface(s). Thus, a parameter k was introduced to describe the less-than-ideal interface coupling.

Recently, we found that a possible weakness of interface coupling is not the only factor decreasing the ME voltage coefficient for laminate ME composites. The compactness (or piling density) of the magnet layer is also a key factor affecting α_E. In this paper we present an observation of significantly enhanced ME interactions in ferrite–Pb(Zr, Ti)O_3 bilayers with increasing compactness of the ferrite contained in the bilayers.

2. Sample characterization

The samples under investigation are $Ni_{0.8}Zn_{0.2}Fe_2O_4$–Pb(Zr, Ti)O_3 bilayers formed by bonding sintered discs of Pb(Zr, Ti)O_3 (PZT) and sol–gel derived $Ni_{0.8}Zn_{0.2}Fe_2O_4$ (NZFO) with hot-pressing and sintering at higher temperatures. Layered NZFO–PZT is a typical ME composite, which has attracted much attention in recent years [16].

Samples of $Ni_{0.8}Zn_{0.2}Fe_2O_3$ (NZFO) were prepared from nanoparticles obtained by sol–gel techniques as described previously [17]. The powder was pressed into pellets of thickness 7 mm and treated at 70 MPa and 950 °C for 1 h with a hot-press facility. The hot-

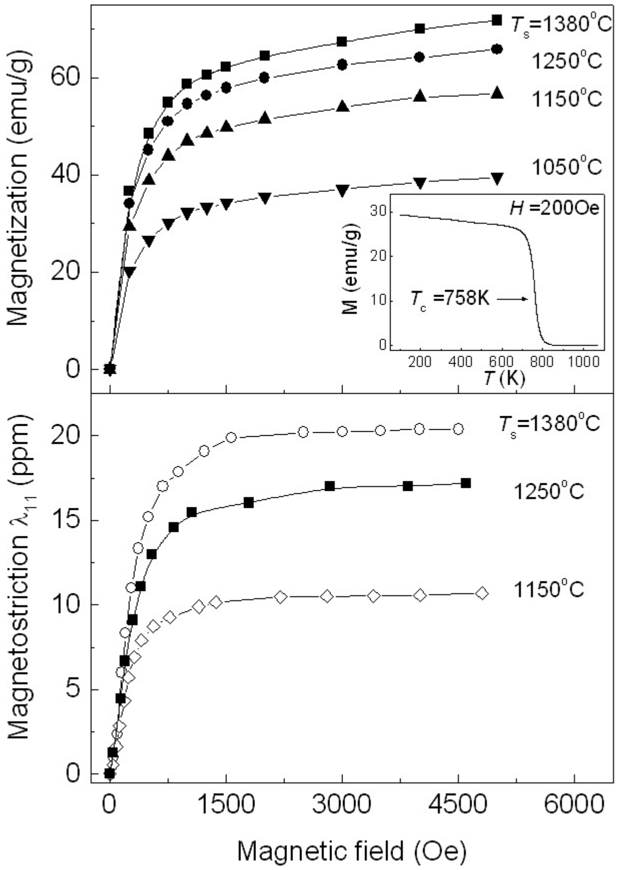

Figure 2. Magnetic field dependence of magnetization (upper panel) and magnetostriction (lower panel) for $Ni_{0.8}Zn_{0.2}Fe_2O_4$ sintered at different temperatures. The field is parallel to the sample plane. Inset of upper panel: temperature dependence of magnetization for 1380 °C sintered $Ni_{0.8}Zn_{0.2}Fe_2O_4$. The dots are measured data and the lines are guides to the eye.

pressed samples were further sintered at 1000 to 1380 °C to get NZFO samples of different compactness.

The samples are characterized by XRD in a rotating anode diffractometer. Figure 1 shows the XRD spectrum of NZFO sintered at 1380 °C. The pattern reveals the absence of any impurities. The inset of the figure shows scanning electronic microscopy photos for both hot-pressed (HP) and not hot-pressed (NHP) NZFO (all sintered at 1380 °C). It can be observed that the compactness of HP NZFO is much larger than that of NPH NZFO. Here, the compactness is defined as the ratio of the density measured to that of the theoretical estimate from the structure parameters obtained from the XRD data. The density was measured by using a floating method of with alcohol.

3. Experimental results and discussion

The hot-pressed and higher temperature sintered NZFO show better magnetic parameters. Magnetic characterizations of the NZFO included magnetization and magnetostriction, as shown in figure 2. The former (the upper panel) was measured with a vibrating sample magnetometer, and the letter (the lower panel) was obtained by the standard strain gauge

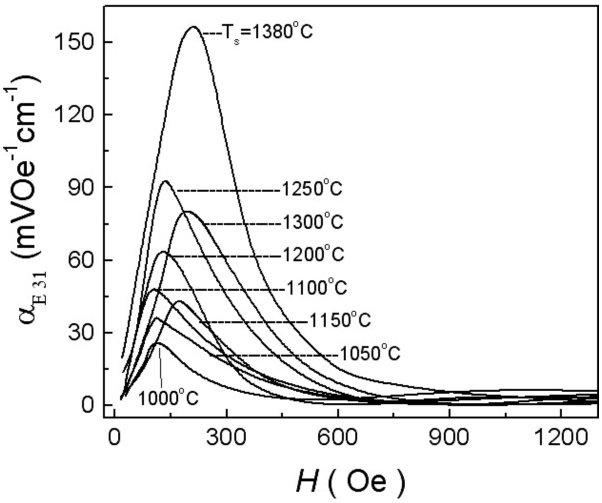

Figure 3. Bias magnetic field dependence of transverse ME voltage coefficient for NZFO–PZT bilayers composed of NZFO sintered at different temperatures.

technique. It can be seen that the magnetization of NZFO increases with increasing sintering temperature T_s. The temperature dependence of magnetization for the NZFO shows the same Curie temperature as that reported in the literature (shown in the inset of the figure). The magnetostriction λ is an important magnetic parameter for ME coupling. The dynamic ME coupling is directly proportional to the piezomagnetic coefficient defined by $q = \delta\lambda/\delta H$. The measurement of magnetostriction was made for the field parallel to the plane of the disc-shaped sample. The magnetic field dependence of the magnetostriction has shown a similar characteristic of trend to saturation with that of magnetization and also increases with increasing T_s at room temperature. The latter in fact reflects that the compactness of the ferromagnetic layer is one of the key factors influencing the machine-transmitting efficiency of the magnetostriction, and accordingly leads to a different ME coupling in the ME bilayers (to be discussed later).

Bilayers were made by bonding the thick discs of ferrite and PZT. PZT was first poled by heating to 425 K and cooling back to room temperature in an electric field of 30–50 kV cm^{-1} perpendicular to the sample plane. It was then bonded to NZFO with a type of slow-dried epoxy resin.

For ME characterization, we measured the electric field δE ($=\delta V/t$, where t is the thickness of PZT, δV is the voltage across the sample), which was produced by an alternating magnetic field applied to the composite and oriented to direction 3. The samples were positioned in a measurement cell and subjected to a bias magnetic field H and an ac magnetic field δH, which was oriented to direction 1, with a frequency of 100 Hz at room temperature. The voltage was amplified and measured with an oscilloscope or a lock-in-amplifier. The ME voltage coefficient was estimated from $\alpha_E = \delta E/\delta H = \delta V/t\delta H$. The measurements were done for the magnetic fields H and δH along the direction parallel and perpendicular to the sample plane, respectively. Then the ME voltage coefficients transverse and longitudinal to the sample plane $\alpha_{E,31}$ and $\alpha_{E,33}$, respectively, were obtained. Theory and experiments have shown that $\alpha_{E,33}$ is very small for all materials, so $\alpha_{E,31}$ is usually the main parameter investigated.

Figure 3 shows the bias magnetic field dependence of $\alpha_{E,31}$ for NZFO–PZT bilayers made up of NZFO sintered at different temperatures. It shows that the ME coupling is enhanced in the mass as the T_s of the NZFO increases. The peak value of the ME voltage coefficient

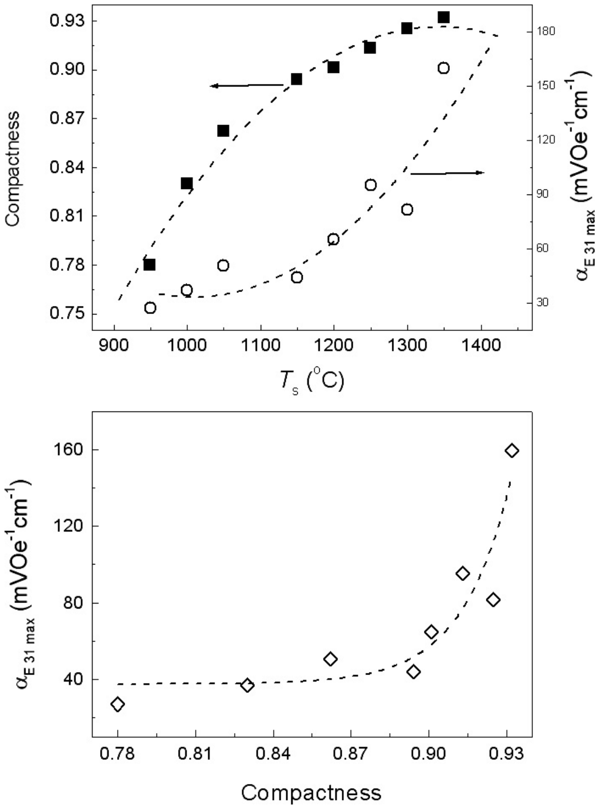

Figure 4. Upper panel: peak value of ME voltage coefficient $\alpha_{E,31\,\mathrm{max}}$ and compactness of the hot-pressed NZFO and as functions of the sintering temperature of the NZFO in the bilayers NZFO–PZT. Lower panel: compactness dependence of the peak value of the ME voltage coefficient $\alpha_{E,31\,\mathrm{max}}$.

$\alpha_{E,31\,\mathrm{max}}$ versus T_s of the NZFO is shown in the upper panel of figure 4. Meanwhile, the compactness of the NZFO is also observed to increase with increasing T_s, as also shown in the upper panel of figure 4. Thus we can infer $\alpha_{E,31\,\mathrm{max}}$, as the ME coupling increases with increasing compactness, as shown in the lower panel of figure 4. Evidently, the present result cannot be explained as being due to interface diffusion since the ME voltage usually decreases with increasing T_s if there is any interface diffusion [14]. We believe that the lower compactness of a magnet can decrease the machine-transmitting efficiency of its magnetostriction, and result in a loss of the magnetostriction property, as seen in figure 2. The ME coupling directly depends on magnetostriction, and thus on the compactness of the included ferromagnetic layer. From figure 4, we also notice that the role of increasing compactness does not completely agree with that of $\alpha_{E,31}$. The former shows a tendency to saturation, while the rate of increase in $\alpha_{E,31}$ increases as T_s increases. This suggests that there are other factors that affect the ME coupling besides the compactness, such as interface coupling.

According to the literature, the highest value of $\alpha_{E,31}$ is 420 mV Oe^{-1} cm^{-1} for nickel ferrite–PZT bilayers [9, 12, 13]. Compared with the values above, the results in figure 3 are not impressive. However, our observations make it evident that weakening of the interfacial coupling is not the only factor decreasing the ME effect; the compactness of the magnet used also affects the ME coupling. Additionally, the compactness of the ferrite can also influence the

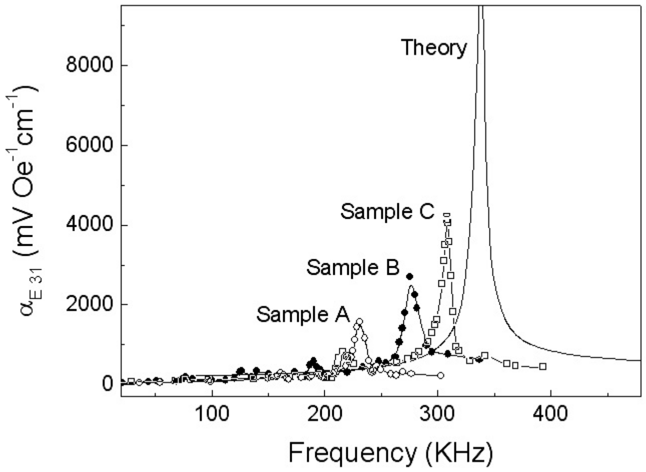

Figure 5. Frequency dependence of transverse ME voltage coefficients for the NZFO–PZT bilayers composed of NZFO sintered at 1000 and 1300 °C, respectively, and the theoretical values of $\alpha_{E,31}$ for a NFO–PZT bilayer from [18]. The bias field H was set for maximum ME coupling (figure 3). The dots are measured data and the lines are guides to the eye.

ME resonance in the bilayers investigated. Figure 5 shows the frequency dependence of $\alpha_{E,31}$ for the bilayers composed of ferrite sintered at 1150, 1250 and 1380 °C and that of theoretical estimates taken from [18].

In the measurements, the bias field was set at the field corresponding to a maximum in α_E. The voltage coefficients were then measured as a function of the frequency f of the ac field. Upon increasing f, we observe that $\alpha_{E,31}$ increase gradually, with some minor peaks in the beginning, then with higher peaks at 231.4, 275.5 and 308.5 kHz for the bilayers composed of NZFO sintered at 1150 °C (sample A), 1250 °C (sample B) and 1380 °C (sample C), respectively. The peak values for these can reach about 1565, 2700 and 4200 mV Oe^{-1} cm^{-1}, respectively.

We identified the resonance at f_r as electromechanical resonance (EMR) in the bilayers, but not in PZT only. The resonance is characterized by a discontinuity in impedance versus f data [19, 20]. The resonance in the ME coefficient occurs when the ac field is tuned to EMR. We recently developed a model for magnetoelectric interactions at EMR. In a bilayer in the form of thin disc of radius R the ac magnetic field induces harmonic waves in the radial or thickness modes. The model considers radial modes for transverse or longitudinal fields. An averaging procedure was employed to obtain the composite parameters and the ME voltage coefficient α_E. The frequency dependence of α_E shows a resonance character at electromechanical resonance in the bilayer. The resonance frequency depends on R, mechanical compliances, density and the coefficient of electromechanical coupling for the radial mode. The peak value of α_E and the width of the peak are determined by effective composite parameters. It is quite clear that the samples composed of the ferrite with different values of compactness should display different resonance frequencies. That has been shown in figure 5.

The profile in figure 5 thus shows resonance with $f_r = 231.4$, 275.5 and 308.5 kHz for samples A, B and C, and a width $\Delta f = 10$ kHz. It is found that the resonance frequency and the height of the resonance peak all increase with increasing compactness, and gradually tend to the theoretical estimate. A similar resonance occurred for longitudinal fields. The resonance occurs at the same frequency as for the transverse fields, but with a much smaller maximum compared to the transverse fields.

4. Conclusion

We have investigated the nature of the magnetoelectric interactions in bilayers consisting of sol–gel prepared ferrite and piezoelectric PZT. Very strong ME interactions are measured for the bilayers. The ME coupling in the bilayer was found to increase with increasing compactness of the included ferrite. This is evidence that the compactness, and therefore the mechanical transport efficiency of the magnetostriction of the ferromagnetic layer, is one of the key factors in ME coupling and electromechanical resonance in laminate composites.

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