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Ni–PZT–Ni trilayered magnetoelectric composites synthesized by electro-deposition

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Abstract

We reported the high strength of magnetoelectric (ME) coupling of trilayered composites prepared by electro-deposition. The ME coupling of Ni–lead zirconate titanate–Ni trilayered structure was measured in the range of 1–120 kHz. The trilayered composites exhibit high ME voltage coefficient because of good interfacial bonding between piezoelectric and magnetostrictive layers. The peak value of the ME voltage coefficient can be up to $33 \text{ V cm}^{-1} \text{ Oe}^{-1}$ at the electromechanical resonance frequency. This magnetoelectric effect shows promise for application in transducers for magnetoelectric energy conversion.

1. Introduction

Multiferroic materials have drawn increasing interest due to their multifunctionality, which provides significant potential for applications in the next-generation multifunctional devices [1]. In the multiferroic materials, the coupling interaction between multiferroic orders could produce many absorbing effects, such as the magnetoelectric (ME) effect and the magnetodielectric effect [2]. The ME response, characterized by the appearance of an electric polarization upon applying a magnetic field and/or the appearance of magnetization upon applying an electric field, has been observed as an intrinsic effect in some single-phase materials (e.g. HoMnO_3 at low temperature and in high magnetic field) [3]. Alternatively, multiferroic composites made by combination of ferroelectric and ferromagnetic substances, such as piezoelectric ceramics (e.g. BaTiO_3 and lead zirconate titanate (PZT)) and ferrites, were found to exhibit large room temperature extrinsic ME effects [4–9]. This ME effect can be defined as a coupling of magnetic–mechanical–dielectric behaviors. That is, when a magnetic field is applied to the composites, the magnetostrictive phase changes the shape, the strain is passed along to the piezoelectric phase, and then an electric polarization happens [10].

In the last few years, a variety of layered ME composites were investigated. Lalein *et al* reported the ME interactions in

layered transition metal/PZT samples synthesized by bonding slices of PZT and Fe, Co or Ni [11]. To achieve better ME properties, giant magnetostrictive material $\text{Tb}_{1-x}\text{Dy}_x\text{Fe}_{2-y}$ (Terfenol-D) was used to combine with piezoelectric materials, such as PZT and polyvinylidene fluoride (PVDF), in a laminate structure [12–18]. The ME voltage coefficient was around $5 \text{ V cm}^{-1} \text{ Oe}^{-1}$. Latterly, much higher ME coefficients at the frequency of electromechanical resonance (EMR) have been achieved (from 30.8 to $238 \text{ V cm}^{-1} \text{ Oe}^{-1}$ for different multilayer structures) [19–23]. But the bonding plastic has the disadvantage of fatigue.

Electro-deposition has been widely used in preparing composite functional materials, composed of metal and alloys with good adhesion [24–26]. Electro-deposition has many other outstanding merits; for example it enables us to make materials with complex structure, to control the thickness of the plating, and to avoid the bonding plastic.

2. Experimental details

PZT slabs were first sliced as $10 \text{ mm} \times 20 \text{ mm} \times 0.25 \text{ mm}$, materialized using sinter sliver paint, and joined to an electrode on both sides. PZT slabs were polarized at 425 K in an electric field of $30\text{--}50 \text{ kV cm}^{-1}$ perpendicular to the sample plane. Then PZT slabs were bathed in nickel aminosulfonate plating solution, and 0.05 A cm^{-2} cathodic current density was

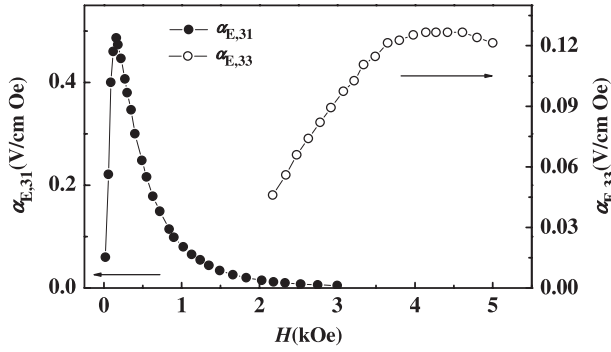


Figure 1. Magneto-electric voltage coefficient $\alpha_{E,31}$ and $\alpha_{E,33}$ at room temperature for Ni-PZT-Ni trilayered composites with total thickness of Ni of about 0.4 mm.

Table 1. Components and process parameters of the nickel electro-deposition.

Nickel aminosulfonate (g l^{-1})	600
Nickel chloride (g l^{-1})	20
Boric acid (g l^{-1})	20
Sodium lauryl sulfate (g l^{-1})	0.1
pH value	4
Temperature ($^{\circ}\text{C}$)	60
Cathodic current density (A dm^{-2})	5

applied to electroplate Ni on both sides of PZT slabs. The components of the plating solution and processing parameters are listed in table 1. Nickel aminosulfonate plating solution was used because of its advantages of solution stability, rapid plating speed and small internal stress. Nickel chloride could help the dissolution of the anode. Boric acid acted as a buffer to stabilize the pH of the plating solution. The pH value was adjusted to about 4 by using sulfamic acid and sodium hydroxide. The surfactant sodium lauryl sulfate was used to prevent pinholes on the surfaces of final products. The volume of the total solution was about 1 l. The plating's thickness could be controlled by the plating time. The thickness of the Ni layer was 0.1 mm for one hour's plating, and 0.4 mm for four hours' plating. Before the ME voltage coefficient was measured, the trilayered composites were dried for six days at 100°C in an oven.

During measuring the ME voltage coefficient, the samples were put in a bias magnetic field H and an AC field δH (20 Hz–100 kHz). Since the AC magnetic field δH was generated by a Helmholtz coil, the amplitude of the AC magnetic field δH equals 22 Oe when the amplitude of the AC current is 1 A through the coil. The generated voltage δV was amplified and measured using an oscilloscope. The ME voltage coefficient was calculated according to $\alpha_E = \delta V / (t_{\text{PZT}} \cdot \delta H)$, where t_{PZT} is the thickness of PZT. They were measured in two different field orientations. The transverse coefficient $\alpha_{E,31}$ was measured, with H and δH parallel to the length of the samples (direction 1) and perpendicular to δE (direction 3). The longitudinal coefficient $\alpha_{E,33}$ was measured with all the fields perpendicular to the sample plane (direction 3).

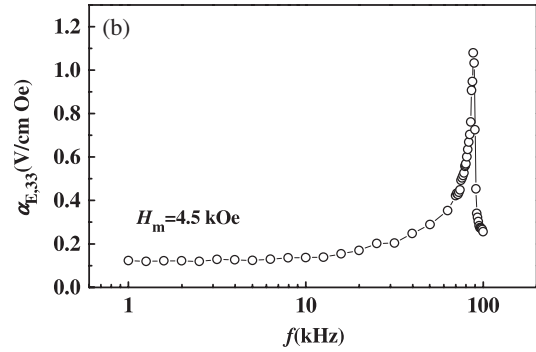
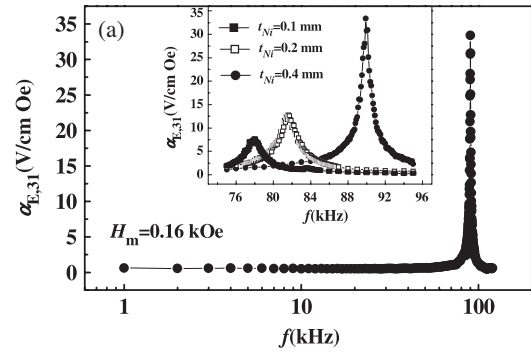


Figure 2. Frequency dependence of $\alpha_{E,31}$ (a) and $\alpha_{E,33}$ (b) for the Ni-PZT-Ni trilayered composites with total thickness of Ni about 0.4 mm at H_m corresponding to maximum ME coupling (see figure 1). The inset shows the ME voltage coefficient around the EMR frequency for the samples with different Ni thicknesses.

3. Results and discussion

First, the dependence of α_E on H was measured at the frequency of the AC magnetic field δH of 1 kHz in transverse (in-plane) and longitudinal (out-of-plane) magnetic fields respectively (figure 1). With the rise of H , there is a sharp peak in the field dependence of $\alpha_{E,31}$ at $H_m = 0.16$ kOe, while a flat peak at $H_m = 4.5$ kOe appears in the field dependence of $\alpha_{E,33}$. The magnitude and field dependences of α_E are related to many factors, such as the large magneto-electric susceptibility of the composites and the variation of the demagnetization field [27]. The ME coefficients are directly proportional to the magnetostrictive coefficient q of the ferromagnetic layer, which will approach zero at saturation magnetization. So both $\alpha_{E,31}$ and $\alpha_{E,33}$ will tend to zero in high field. On account of the shape demagnetization effect, H_m of $\alpha_{E,31}$ is much lower than that of $\alpha_{E,33}$. The trend of $\alpha_{E,31}$ to zero in high field can be observed in the experimental scope of intensity of the magnetic field, while that is not obvious for $\alpha_{E,33}$.

Second, α_E was measured in the bias field of H_m when the frequency of the AC magnetic field (f) varied from 1 kHz to 120 kHz (figure 2). A typical α_E versus f profile for transverse fields is shown in figure 2(a). For both $\alpha_{E,31}$ and $\alpha_{E,33}$, there is a sharp peak at about 88 kHz. However, the peak value of $\alpha_{E,33}$, about $1.08 \text{ V cm}^{-1} \text{ Oe}^{-1}$, is an order of magnitude smaller than that of $\alpha_{E,31}$, $33 \text{ V cm}^{-1} \text{ Oe}^{-1}$.

Figure 3 shows the frequency dependence of the dielectric constant and dielectric loss of the trilayered Ni-PZT-Ni composite. There is a resonance peak at about 89.9 kHz which

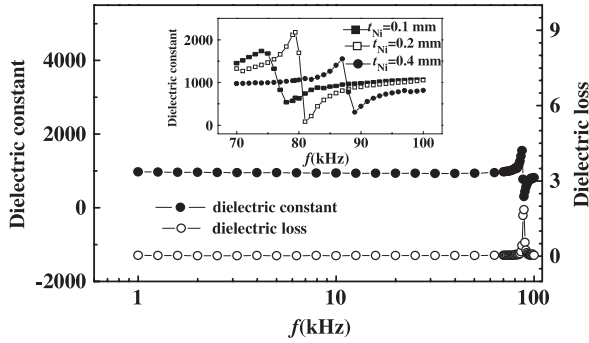


Figure 3. Frequency dependence of the dielectric constant and dielectric loss for the Ni–PZT–Ni trilayered composites with total thickness of Ni about 0.4 mm. The inset shows the dielectric constant around the EMR frequency for the samples with different Ni thicknesses.

is associated with the electromechanical resonance [28, 29]. The consistency of the resonant frequencies of the dielectric constant and α_E indicates that the high ME effect is associated with the EMR.

Figure 2 inset shows the frequency dependence of $\alpha_{E,31}$ for the samples with different thicknesses of the Ni layer around the frequency of EMR. The frequency of the EMR shifts towards high frequency when Ni layers get thicker, because it is directly proportional to the thickness of the piezoelectric and piezomagnetic layers [30]. Figure 3 inset also shows the frequency dependence of the dielectric constant for the samples with different Ni layer thicknesses, which is consistent with the frequency dependence of the ME coefficient. The resonant frequency of the ME voltage coefficient corresponds to that of dielectric constant for the sample with the same Ni thickness.

Figure 4 plots the dependence of the ME voltage coefficient on $t_{Ni}/(t_{Ni} + t_{PZT})$ for trilayered Ni–PZT–Ni samples at the frequency of EMR. When $t_{Ni}/(t_{Ni} + t_{PZT})$ rises from 0.2 to 0.65, both $\alpha_{E,31}$ and $\alpha_{E,33}$ increase monotonically. When the total thickness of Ni is about 0.4 mm, $\alpha_{E,31}$ is as high as $33 \text{ V cm}^{-1} \text{ Oe}^{-1}$, and $\alpha_{E,33}$ is $1.08 \text{ V cm}^{-1} \text{ Oe}^{-1}$. According to Bichurin’s theoretical model on resonance magnetoelectric effects, the peak values of the ME coefficient are determined by the effective piezomagnetic (q_{31}) and piezoelectric (d_{31}) coefficients, compliances and permittivity [29]. The experimental result is qualitatively in agreement with the theoretical prediction.

For the magnetoelectric laminate structure, the interfacial bonding between the magnetostrictive layer and piezoelectric layer is important to the ME coefficient. Liu *et al* theoretically reported the influence of the interfacial binder layer’s thickness and shear modulus on the magnetoelectric effect [31]. While the interfacial binder layer’s thickness rises or its shear modulus reduces, the ME responses will decrease rapidly. It is worthwhile noting that although the magnetostriction of Ni is two orders of magnitude smaller than that of Terfenol-D, the coefficient α_E of the laminated Ni–PZT–Ni composite synthesized by electro-deposition is comparable with that of Terfenol-D/PZT/PVDF bulk samples [32, 33]. This is

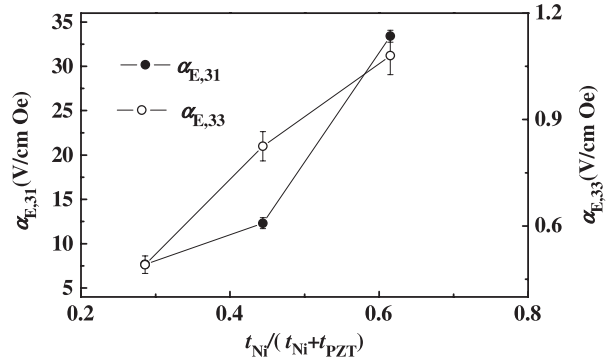


Figure 4. $t_{Ni}/(t_{Ni} + t_{PZT})$ dependence of the ME voltage coefficient for Ni–PZT–Ni trilayered composites at the frequency of the EMR.

because the plastic interfacial layer of PVDF in the Terfenol-D/PZT/PVDF system is replaced by an Ag metal interfacial layer in the electro-deposited Ni–PZT–Ni system. The shear modulus of Ag is much higher than that of PVDF, and the Ag layer is thinner than the PVDF binder layer. Hence, better interfacial coupling between PZT and Ni layers will fill the gap of Ni’s small magnetostriction. It is promising to enhance the ME coefficient more by improving electro-deposition technology, such as electro-depositing metal with a higher magnetostriction coefficient.

4. Summary

In summary, this paper presents the first report on ME interaction of Ni–PZT–Ni trilayered composites synthesized by electro-deposition. At the frequency of the EMR, the coefficient α_E has a peak value $\alpha_{E,31}$ up to $33 \text{ V cm}^{-1} \text{ Oe}^{-1}$ when the thickness of Ni is about 0.4 mm. Tight bonding between PZT and Ni layers makes these samples exhibit large ME voltage coefficients among bulk magnetoelectric composites. Electro-deposition provides an effective method for remarkably enhancing the ME coefficient of ME composites. In addition, the electro-deposition method makes easy the preparation of ME composites with complex structure, and can control the structural parameters effectively. It will promote a rapid development of magnetoelectric composites’ applications, such as in various magnetoelectric coupling devices.

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