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The Influence of Flexural Deformation on the Static Magnetoelectric Coefficient of a Bilayered Magnetoelectric Composite

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In the bilayered Ni/Pb(Zr, Ti)O₃ (PZT) composite, upon the application of a magnetic field, a strong flexural deformation was observed. Using an analogous scenario involving a known thermo bimetal, a simple formula for the curvature in the bilayered Ni/PZT composite was obtained. This result permitted the strain distribution and the static magnetoelectric (ME) coefficient in the bilayer composite to be easily solved. Experimental and theoretical results quantitatively explained how the flexural deformation weakened the ME effect in the bilayer composite, which agreed well with the experimental results from many previous experiments.

Keywords: Layered Structures, Flexural Deformation, Magnetoelectric Composite

Introduction Multiferroic materials have been drawn increasing interest due to the physics inherent in the coexistence of two or more states of ferocity and their significant multi-field coupling effect [1], e.g. the magnetoelectric (ME) effect. Multiferroic composites constructed by combining magnetostrictive (MS) and piezoelectric materials have exhibited very large ME coefficients and are already employed in many prototype device applications such as magnetic field sensors [2], multi-state storage cells [3], and for use as miniature antennae [4]. In ME composites, the ME effect is a strain-media product effect between the piezoelectric effect and the MS effect. The stress/strain transfer between the two phases plays a key role during the ME coupling, which is significantly affected by the structure of the composite. Until now, various topological structures have been designed [5] and used to prepare ME composites, such as 0–3 [6,7], 1–3 [8–10], 2–2 [11–13], and 2–1 [14] connectivity structures. To ensure simple fabrication and large resulting ME coefficients, laminated (2–2) ME composites containing bilayer, trilayer, or multilayer structures are the most widely studied [15–18].

The laminated bilayer ME composites have shown very high ME coefficients and relatively low resonance frequencies when loaded in flexural modes. Their flexural

deformation at various resonance frequencies has been observed in detail and confirmed experimentally [15,19]. However, in the low-frequency (static) range, few studies have been carried out to observe the flexural deformation. Moreover, it remains problematic that the static ME coefficient in a bilayer composite is only approximately one-third of that in a trilayer composite with same constituents. At one point, this reduction was considered to be the result of the weaker interface inside the bilayer structure. An interface factor, k , was introduced to estimate the contribution of the weak interface in the model using the average field method [20], in which the flexural deformation in the bilayer structure was ignored. Recently, researchers designed a similar interface structure in bilayer and trilayer Pb(Zr, Ti)O₃ (PZT)/Ni_{0.8}Zn_{0.2}Fe₂O₃ (NZFO) composites [21]. This structure raised the concern that not only the weaker interface but also that the presence of non-linear flexural deformation reduced the ME effect in the bilayer ME composite. Similar results have been seen in other experiments [22]. Therefore, a quantitative explanation regarding the different ME behaviors between bilayer ME composites and trilayer ME composites is quite necessary. In this paper, bilayer and trilayer Ni/PZT ME composites were prepared using identical techniques and

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a detailed comparison of their MS and ME behavior was carried out. Theoretically, the strain distribution in the trilayer and bilayer structures was solved directly, and the different results between these two well-known laminated structures were analyzed.

Experimental Details The bilayer and trilayer ME composites were both prepared by gluing the Ni plate and the PZT-5A plate together with epoxy adhesive, as shown in Figure 1. The PZT plate was coated with the silver film to act as an electrode and poled along the thickness direction. The Ni plate was prepared by wire-electrode cutting and polished afterwards. The thickness of the composite is denoted as t , and the volume fraction of the Ni plate is denoted as n . The size of all samples was fixed at $4 \times 9 \times 1 \text{ mm}^3$. To perform the magnetostriction measurement, two strain gauges were glued parallel to the length direction on each sample's top and bottom surfaces. For the ME measurement, the samples were put into a bias DC magnetic field, H_{bias} , which superimposed a magnetic field, dH , from an alternating current. The induced ME voltage, dV , across the PZT plate was then measured using a lock-in amplifier. In this way, the ME coefficient dV/dH was obtained. Because the input impedance of the lock-in amplifier is very high, the PZT plate can be seen under an open circuit boundary condition. Furthermore, because the dV is also an alternating signal, the sign of the ME coefficient is defined according to the phase drift between dV and dH . If there is no phase drift between dV and dH , the sign of dV/dH is positive. If there is a phase drift of 180° , the sign of dV/dH becomes negative. Therefore, by applying the bias magnetic field in a range between -2 and $+2 \text{ kOe}$, the variation of the ME coefficient will construct an ME loop analogous to a ferromagnetic $M-H$ loop (or ferroelectric $P-E$ loop). In this experiment, all of the magnetic field was applied along the 9-mm length direction of the sample. The ME loop was measured at 1 kHz .

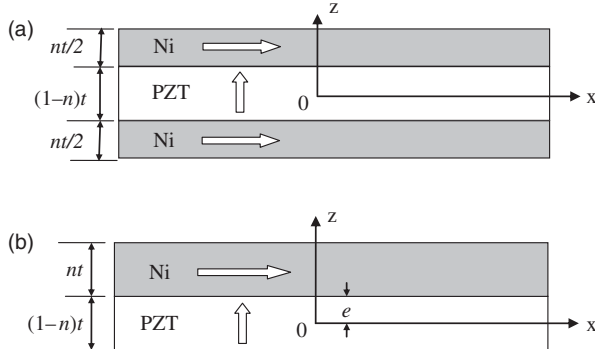


Figure 1. Schematic drafts of the prepared trilayer and bilayer Ni/PZT-laminated composites.

Results and Discussion Figure 2 shows the surface strain of the pure Ni plate, the bilayer composite, and the trilayer composite from applying the magnetic field. For the pure Ni plate, the MS curve shows a slight hysteresis, and the saturated MS strain is approximately -41 ppm , which is somewhat larger than the polycrystalline Ni saturated MS strain of -33 ppm . This strain is believed to originate from the slight textured structure of the Ni plate, which usually appears in the rolled Ni strip [23]. For the symmetric trilayer Ni/PZT/Ni composite, the surface strains on the top and the bottom surface are equal and are slightly smaller than that in the pure Ni plate. The reason for this result is that the trilayer Ni/PZT/Ni has a symmetric structure, and the inner PZT layer constrains the extensional deformation of the two Ni layers equally. The MS strain of the Ni is always negative, which is also seen in the pure Ni plate in Figure 2. Surprisingly, for the un-symmetric bilayer Ni/PZT composite, positive strain appears on the PZT surface. Moreover, the strain on the Ni surface in the bilayer composite is much larger than that of the pure Ni plate due to the application of the magnetic field. All of these ‘abnormal’ surface strains are quite different from that in the trilayer Ni/PZT composite, which suggests that a strong flexural deformation occurs in the Ni/PZT bilayer composite from applying a magnetic field.

The measured ME coefficient loops of the bilayer and trilayer ME composites are shown in Figure 3. Slight hysteresis is observed in both the bilayer Ni/PZT and the trilayer Ni/PZT/Ni in the low magnetic field range. The coercive field is only approximately 37 Oe , and a small remnant of the ME coefficient appears at zero magnetic field, which demonstrates the soft magnetic performance of Ni. The maximum ME coefficients in the bilayer composite and the trilayer composite are approximately 3.6 mV/Oe (72 mV/cmOe) and 10 mV/Oe

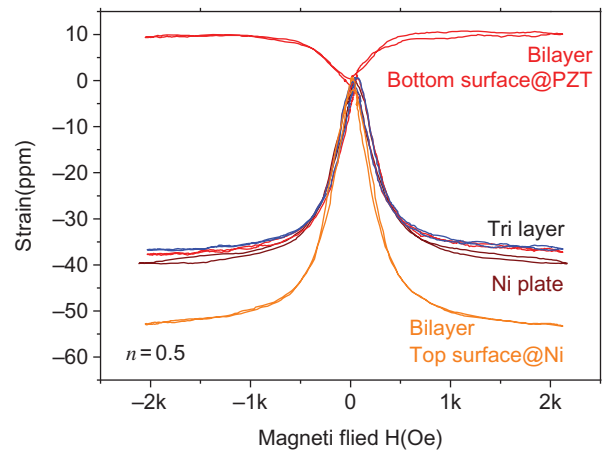


Figure 2. The measured surface strain of the pure Ni plate, trilayer Ni/PZT composite, and bilayer Ni/PZT composite on applying magnetic field.

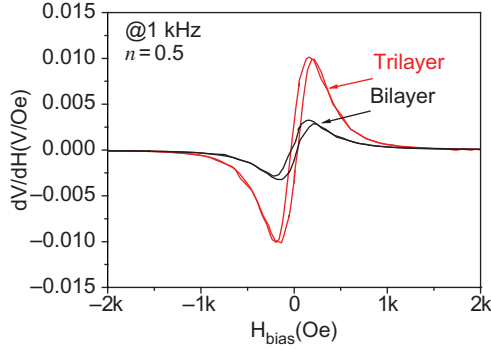


Figure 3. The measured ME loop of the prepared bilayer Ni/PZT and trilayer Ni/PZT/Ni.

(200 mV/cmOe), respectively. The ME effect in the bilayer structure is approximately one-third of that seen in the trilayer structure in the Ni/PZT ME composites. This result agrees with those from previous experiments conducted by Zhang [21].

Theory To explain the MS and ME behavior, a simple mechanical analysis was carried out. For convenience, the sample was simplified to a one-dimensional composite beam, so that only the strain along the length direction needs to be considered. The strain was assumed to be uniform along the length direction, and the interface was assumed to be perfect.

For the trilayer composite, because there is no extrinsic force, by solving the force balance equation in the two phases, the strain of the composite $\bar{S}$ can be easily obtained as

$$\bar{S} = \frac{nS_{11}^{ms}S_{11}^D}{(1-n)S_{11}^H + nS_{11}^D}, \quad (1)$$

where S_{11}^{ms} is the MS strain of the pure Ni plate before combination, S_{11}^D is the compliance of the PZT phase along the direction of the applied magnetic field under the open circuit condition, S_{11}^H is the compliance of the Ni phase along the direction of the applied magnetic field in a constant magnetic field, and n is the volume fraction of the Ni layer.

For the bilayer structure, the MS extension of the Ni phase induces not only the uniform elongation along the direction of the applied magnetic field (as described in Equation (1)), but also a flexural deformation from the asymmetric stress mismatch, similar to the well-known occurrence of bending in thermo bimetals. By substituting the thermo strain mismatch with the MS strain mismatch, a simple curvature formula [24] for the bilayer ME composite can be written as

$$\frac{1}{\rho} = -\frac{6S_{11}^{ms}(1+m)^2}{t[3(1+m)^2 + (1+m\delta)(m^2 + 1/m\delta)]}, \quad (2)$$

where $1/\rho$ is the curvature, $m = n/(1-n)$ is the relative thickness ratio, $\delta = S_{11}^D/S_{11}^H$ is the ratio of Young's

modulus, and t is the total thickness of the bilayer composite. For the bending of the sample, the strain in the bilayer structure is the sum of the flexural strain and the extensional strain:

$$S_1 = -\frac{z}{\rho} + \bar{S}, \quad (3)$$

where z is the coordinate along the thickness direction, as shown in Figure 1(b), i.e. the plane $z = 0$ is the neutral plane, where the strain is defined as only $\bar{S}$ (Equation (1)). The position of the neutral plane, e (Figure 1), is determined by solving the moment balance equation

$$e = \frac{(1-n)^2/s_{11}^D - n^2/s_{11}^H}{2(1-n)/s_{11}^D + 2n/s_{11}^H} \cdot t. \quad (4)$$

In this calculation, $s_{11}^D = 1.46 \times 10^{-11} \text{ m}^2/\text{N}$, $s_{11}^H = 6.62 \times 10^{-12} \text{ m}^2/\text{N}$, and $S_{11}^{ms} = -41 \text{ ppm}$. The elastic coefficients were measured by the standard method using long-beam vibrators made by the individual constitute phase. Using Equations (1) and (3), the surface strain is calculated as a function of the Ni volume fraction and is shown in Figure 4. The measured surface strain is also plotted. The calculated results are found to be in good agreement with the experimental results (Figure 4). For the trilayer composite, when $n = 0.5$, the calculated value is slightly smaller than the measured saturated strain, -37 ppm . This discrepancy results from the strain transfer lag from the Ni layer to the PZT layer because the outer surface of the Ni layer is less constrained by the inner PZT layer. For the bilayer composite, when the thickness of the Ni layer is small, there is actually positive strain that appears on the PZT surface. In middle range, n , the calculated strain on the Ni surface is actually larger than the strain of the pure Ni plate, -41 ppm . This finding provides a good explanation for the surprisingly large measured results described above.

Figure 5 shows the calculated curvature (Equation (2)) and the neutral plane's position, e (Equation (4)), as

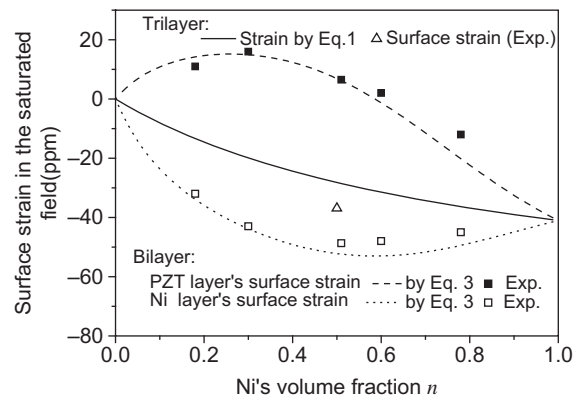


Figure 4. The dependence of the surface strain of bilayer and trilayer Ni/PZT-laminated composite on the Ni's volume fraction.

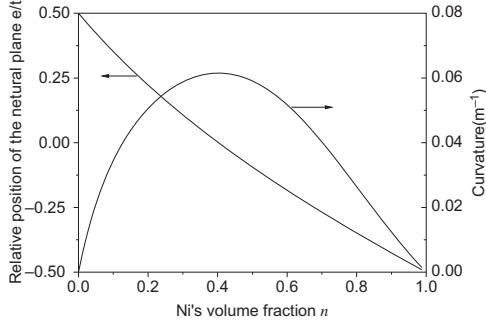


Figure 5. The dependence of the calculated curvature in the saturated field and the relative position of the neutral plane in the bilayer Ni/PZT composite on the Ni's volume fraction.

the function of the Ni volume fraction, n . As seen from Figure 5, the curvature is always positive because the MS strain of the Ni is negative, and the top layer is Ni (Figure 1). The maximum deflection occurs when n is approximately 0.40, where $e = 0$, i.e. where the neutral plane is the interface of the composite. When the volume fraction n approaches 0 or 1, i.e. when either the Ni layer or the PZT layer is very thin, the curvature is quite small; thus, the extensional strain will be dominant.

According to Equation (3), if the curvature is positive, then the strain above the neutral plane decreases and the strain below the neutral plane will increase. Therefore, if the flexural deformation is sufficiently great, its contribution will exceed that of the extensional deformation, inducing an 'abnormally' larger strain on the Ni surface and result in the positive strain on the PZT surface. Because none of the additional assumptions is used, Equations (1)–(4) are reasonable for all bilayer composites. In the bilayer Terfenol-D/ $\text{Bi}_{12}\text{GeO}_{20}$ (BGO) ME composite described by Zhu and co-workers [22], 'abnormal' negative strain was observed on the BGO surface in the low field range, whereas the MS strain in the Terfenol-D is always positive. The negative strain on the BGO surface may in fact result from the flexural deformation.

On the basis of the strain calculated above, the ME coefficient of dV_3/dH_1 is easily calculated using the piezoelectric equation of the PZT phase.

For the trilayer structure:

$$E_3 = -g_{31p}T_{1p} = -\frac{g_{31p}\bar{S}}{s_{11}^D} = -\frac{g_{31p}nS^{\text{ms}}}{(1-n)s_{11}^H + ns_{11}^D}, \quad (5)$$

where g_{31p} is the piezoelectric constant of the PZT and T_{1p} is the stress of the PZT along the length direction. In the low field range, we can assume $S^{\text{ms}} = d_{11m}H_1$, where d_{11m} is the linear piezomagnetic constant. The linear piezomagnetic constant for Ni from the MS curve in Figure 2 is approximately -1×10^{-10} m/A. For PZT-5A, the piezoelectric constant is measured as $g_{31p} = -0.0103$ Vm/N. Because $V_3 = E_3 \cdot (1-n)t$, the ME coefficient can be

written as

$$\frac{dV_3}{dH_1} = -\frac{n(1-n)g_{31p}d_{11m}t}{(1-n)s_{11}^H + ns_{11}^D}. \quad (6)$$

In the case of the bilayer structure, because the strain is not uniform along the thickness direction, the induced voltage must be integrated along the thickness direction in the PZT phase as

$$\begin{aligned} V_3 &= \int_{e-(1-n)t}^e E_3 dz = \int_{e-(1-n)t}^e -g_{31p}T_{1p} dz \\ &= -\int_{e-(1-n)t}^e \frac{g_{31p}S_1}{s_{11}^D} dz \\ &= -\frac{g_{31p}}{s_{11}^D} \int_{e-(1-n)t}^e \left(-\frac{z}{\rho} + \bar{S}\right) dz \\ &= \frac{g_{31p}}{s_{11}^D \rho} \cdot \frac{e^2 - [e - (1-n)t]^2}{2} - \frac{g_{31p}}{s_{11}^D} \bar{S} \cdot (1-n)t. \end{aligned} \quad (7)$$

From Equation (7), we can see that the contribution of the flexural strain above and below the neutral plane will be counteracted in the integration. Substituting Equation (2) into Equation (7) and simplifying yields:

$$\begin{aligned} \frac{dV_3}{dH_1} &= -\frac{(1+m\delta)(m^2 + 1/m\delta)}{3(1+m)^2 + (1+m\delta)(m^2 + 1/m\delta)} \\ &\quad \cdot \frac{n(1-n)t \cdot g_{31p}d_{11m}}{(1-n)s_{11}^H + ns_{11}^D}. \end{aligned} \quad (8)$$

Equation (8) is almost identical to Equation (6) except for a factor which is always smaller than 1, which explains why the static ME effect in the bilayer structure is always weaker than that in the trilayer structure. For mid-range thickness ratios, and for comparable thicknesses of PZT and Ni, when m approaches unity, the factor on the left-hand side of Equation (8) is approximately 1/3. This finding also agrees well with our own experimental results in addition to previous results [21]. For the equation $s_{11}^D = s_{11}^E(1 - k_{31p}^2)$, where s_{11}^E is the compliance of the PZT under short-circuit conditions and k_{31p} is the coupling factor of the PZT, Equations (6) and (8) are the same as those derived by the equivalent circuit method [25,26], but the formulas used here are much simpler.

Figure 6 shows the calculated ME coefficients from Equations (6) and (8). As shown in Figure 6, for bilayer ME composite, the function of the ME coefficient on n is a saddle-shaped curve with two maxima around $n = 0.2$ and 0.8 , which can be explained by Figure 5 and Equation (7). From Equation (7), it can be seen that the ME voltage is derived from two kinds of strain, the flexural deformation $-z/\rho$ and the longitudinal deformation $\bar{S}$. The flexural deformation weakens the ME effect. Hence,

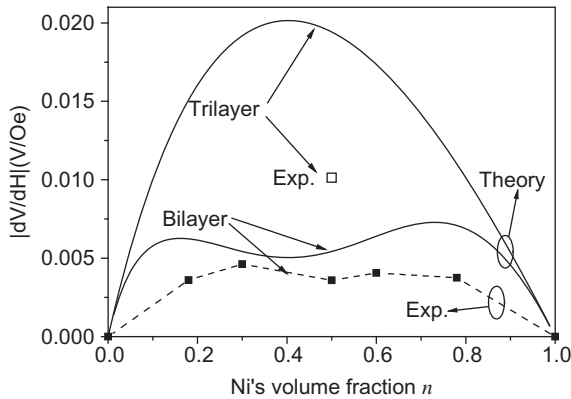


Figure 6. The comparison of the theoretical and experimental ME coefficients of the bilayer and trilayer Ni/PZT laminated composites.

when n approaches either 0 or 1 due to the flexural deformation being very small (Figure 5), the ME coefficient in the bilayer composite is similar to that seen in the trilayer composite. But for mid-range n , the contribution of the flexural deformation is substantial (see the maximum curvature around $n = 0.4$ in Figure 5) and correspondingly the ME effect is weakened strongly, resulting in a saddle-shaped ME coefficient $\sim n$ curve for the bilayer structure. The theoretical result is in good agreement with the experimental curve as seen from Figure 6. However, the experimental ME coefficients are much smaller than the calculated ones, which may result from a weaker interface bond from the epoxy glue. Another important reason for this finding is that we assume that the strain in the length direction is uniform. Actually, because of the ‘end effect’ of the composite, the end section of the beam has a much lower ME coefficient, which has been confirmed by the observation of the strain transfer lag in the trilayer structure [27] and the inhomogeneous distribution of the ME coefficient in the bilayer structure [28]. Further study can be carried out to consider the contribution of the ‘end effect’ in the theoretical model.

Summary In summary, flexural deformation in the bilayer structure has been observed and its influence on the ME effect has been further analyzed using a simple one-dimensional composite beam model. In the low-frequency range, the flexural deformation derived from the MS strain mismatch significantly weakens the contribution of the extensional deformation in the bilayer ME composite, resulting in a lower ME coefficient than is seen in the symmetric trilayer ME composite. Our analysis provides a simple and clear formula for the determination of the ME coefficient in the bilayer ME composite, quantitatively explaining how the flexural deformation occurs and weakens the low-frequency ME effect.

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