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FAST TRACK COMMUNICATION

Shape and size effects on layered Ni/PZT/Ni composites magnetoelectric performance

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Abstract

This paper presents the magnetoelectric (ME) effect in trilayered Ni/PZT/Ni composites which is related to their size and shape. The ME composites with the same interfacial areas but different geometrical shapes have different ME voltage coefficients. Longitudinal resonant modes in the rectangular and triangular trilayered ME composites were studied. One should choose optimized size, shape and working frequency of the ME composites in order to gain the maximum ME effect. This study plays a guiding role for trilayered ME composites design for real applications.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Multiferroic materials have either ferromagnetic or anti-ferromagnetic, ferroelectric or anti-ferroelectric, ferroelastic properties or a combination thereof. They have promising applications due to unique coupling, which provides opportunities for energy transformation [1]. For example, some multiferroic materials exhibit coupling between ferroelectric and ferromagnetic phases and produce new magnetoelectric (ME) or magnetodielectric effects [2, 3]. The ME response, characterized by the appearance of electric polarization upon the application of a magnetic field and/or magnetization upon the application of an electric field, has been observed as an intrinsic effect in some single phase materials [4, 5], including $BiFeO_3$ and $BaMnF_4$ [6, 7]. Such ME effects originate from the local exchange between the internal magnetic structure and the ferroelectric sub-lattice [8]. While being interesting, most single phase materials are difficult to work with due to their low Néel and Curie temperatures and a relatively weak ME response.

To address these issues, researchers began to develop layered heterogeneous magnetoelectric composites containing ferromagnetic and ferroelectric phases. The ME effects in these heterogeneous composites are produced by the mechanical coupling between the ferroelectric and ferromagnetic materials through the piezoelectric and piezomagnetic effects [9]. Researchers quickly discovered that the ME composites exhibit an extremely high ME effect compared with single phase materials and can be operated over a fairly broad range of temperatures. Therefore, the ME layered composites overcome disadvantages associated with single phase ME materials.

Previous studies of ME composites focused on selecting suitable material combinations [10–14] and corresponding layer thicknesses to maximize the ME effect [15–20]. Typically, in these studies the in-plane dimensions were not taken into account. In a previous work, we reported that the ME voltage coefficient depends on relative in-plane sample dimensions due to the shape demagnetization effect on magnetostriction, which increases with the in-plane size due to reduction in the effective volume of the demagnetization region [21]. In this paper, we will further study the effect of the sample size and shape on the ME effect in trilayered

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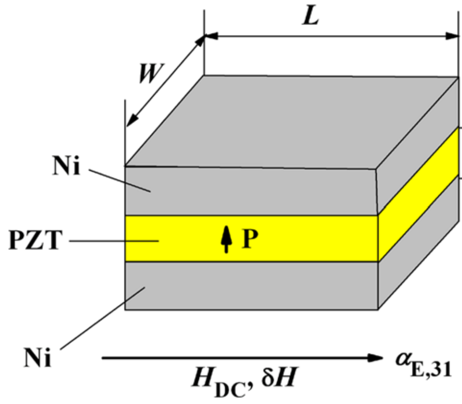

Figure 1. Schematic of the trilayered composite structure geometry and the applied magnetic field. Vector *P* shows the PZT polarization direction.

symmetrical composites, which strongly depends on their in-plane dimensions.

2. Experimental details

Experiments presented in this paper are based on PZT-5H ($PbZr_{0.52}Ti_{0.48}O_3$, supplied by the Institute of Acoustics, Chinese Academy of Sciences, the d_{33} piezoelectric coefficient is about 450 pm V^{-1}) as the piezoelectric phase and electrodeposited Ni layers as the piezomagnetic phase. A large PZT ceramic piece was cut into rectangular smaller pieces 0.8 mm thick and polarized along the through-thickness direction at 425 K in an electric field of 30–50 kV cm^{-1}. Following poling, PZT samples were bathed in nickel aminosulfonate plating solution, and Ni was electrodeposited on both sides of the PZT samples for 8 h. The resulting total thickness of the two Ni layers was approximately 0.8 mm (0.4 mm on each side of the PZT layer). The Ni electrodeposition process is described in detail elsewhere [21–25]. The ME voltage coefficient was calculated as $\alpha_E = \delta V/(t_{PZT} \cdot \delta H)$, where t_{PZT} is the PZT thickness, δV is the voltage measured across the PZT thickness and δH is the change in the applied magnetic field. The ME coefficient $\alpha_{E,31}$ was measured, with H and δH applied parallel to the sample length (direction 1) and perpendicular to δE (direction 3). The sample schematic is presented in figure 1. The ME voltage coefficient was measured in the system discussed in a previous publication [26].

3. Results and discussion

Figure 2 plots the ME voltage coefficient as a function of the applied magnetic field frequency (f) for an Ni/PZT/Ni trilayered composite with dimensions of 16 × 25 × 1.6 mm^3. There are three resonant peaks at 67.2, 102 and 125 kHz. Because the second resonant peak represents a mode that was not clearly identified in all the tested samples, we focus exclusively on the first and the third resonant frequencies in the rest of this paper.

Figure 3 plots ME voltage coefficients corresponding to the first and the third resonance peaks as a function of the sample width W with the constraint that the sample area remains constant, i.e. $W \times L = 400\,\text{mm}^2$. That is, the sample length was varied to keep the in-plane sample area and thus the interfacial area constant. The ME voltage coefficient decreases with increasing width W for the first resonant peak. At a large width, i.e. $W = L = 20$ mm, the first resonant frequency vanishes. The possible reason for this will be discussed further. On the other hand, the ME voltage coefficient increases with the sample width for the third resonant frequency. While the first peak vanishes, the largest amplitude value for the third resonant frequency occurs when $W = L = 20$ mm. These results clearly show that the magnetoelectric coefficient depends on the in-plane sample dimensions rather than on the interfacial area, i.e. the interfacial area is kept constant in these experiments.

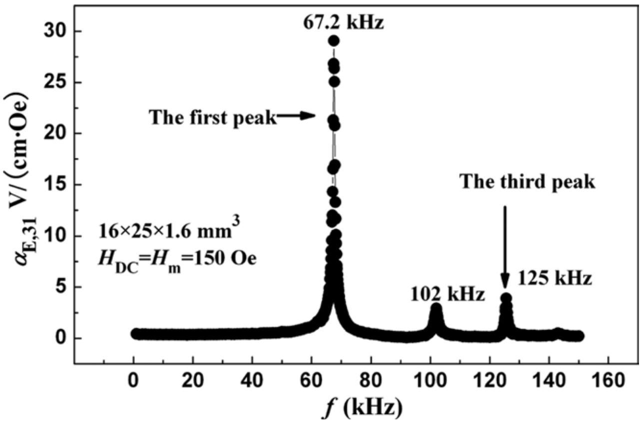

Figure 2. The dependence of the ME voltage coefficient $\alpha_{E,31}$ on the frequency for the ME trilayered composite with the dimensions of 16 × 25 × 1.6 mm^3 (where H_{dc} is the bias magnetic field and H_m is the applied bias magnetic field when the ME effect reaches a maximum in the H_{dc} curve [25]).

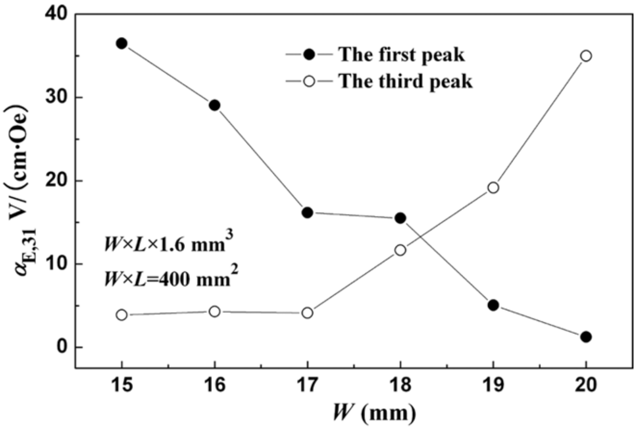

Figure 3. The dependence of the ME voltage coefficient on the width W of layered ME composites with the same interfacial area.

Wan *et al* studied the resonant modes in layered Terfenol-D/Epoxy/PZT ME composites [17]. Below 60 kHz three bending modes were identified, and the first longitudinal peak was found at 70 kHz. The magnitude of the resonant frequency was inversely proportional to the square of the composite length in the bending mode and to the composite length in the longitudinal mode. We intentionally tried to avoid

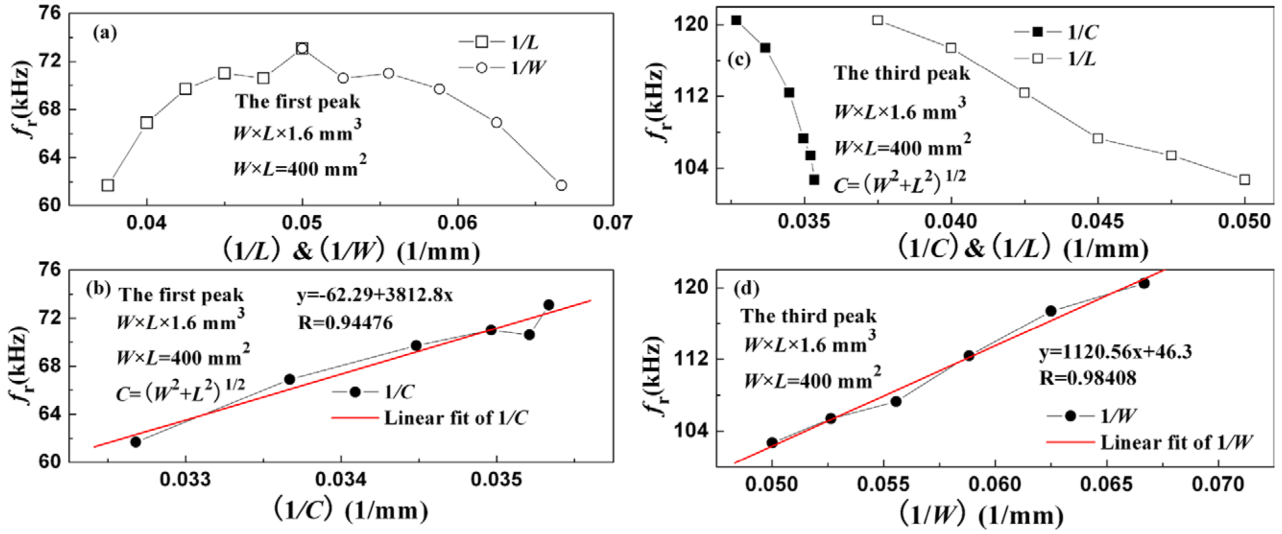

Figure 4. The dependence of resonant frequency on W, L and C of composites corresponding to the first (*a*) and (*b*) and the third (*c*) and (*d*) resonant peaks.

bending modes by building structures that are symmetric with respect to the neutral surface and focus on the longitudinal modes.

Figure 4 plots the value of the first and third resonant frequencies as functions of the composite reciprocal dimensions, $1/W$, $1/L$ and $1/C$. Here, C is defined as the diagonal length of the rectangular sample, i.e. $C = \sqrt{W^2 + L^2}$. The results shown in these figures are obtained from the same samples as in figure 3, where $W \times L = 400\,\mathrm{mm}^2$. In figure 4(*a*) the first resonant frequency increases as L increases or W decreases. Figure 4(*b*) shows that the first resonant frequency increases linearly with decreasing C ($R = 0.944\,76$). On the other hand, in figure 4(*c*) the third resonant frequency decreases as either C or L increases. Figure 4(*d*) shows that the third resonant frequency increases linearly with decreasing W ($R = 0.984\,08$). That is, the first resonant peak is proportional to $1/C$, while the third resonant frequency is proportional to $1/W$. Therefore, we think that the first and third resonant modes are attributed to the longitudinal resonance along the diagonal and width directions in the rectangular sample, respectively. We think the reason for other studies observing only one resonant peak in the longitudinal mode is because the ME samples in [10–20] have different sizes or shapes compared with the sample reported here. For example, in [10] a cylindrical ∅9 mm sample is considered, [11] considers a bilayered structure and [17] studied a flexural bilayered sample and finally [18] considered a rectangular $12.7 \times 6 \times 4.8\,\mathrm{mm}^3$ sample. These dimensions are smaller than our samples; therefore, below 100 kHz, there is the appearance of only one longitudinal mode. In this paper we report that the resonant frequency increased with the decrease in dimensions. Perhaps the second longitudinal peak appears after 100 kHz and was not obtained in other studies.

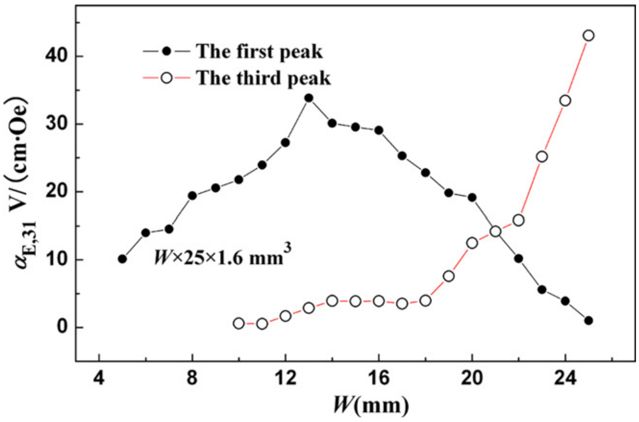

Figure 5. The dependence of the ME voltage coefficient on the width W of layered ME composites with the dimension of $W \times 25 \times 1.6\,\mathrm{mm}^3$.

In order to better understand these results, a larger square $25 \times 25 \times 1.6\,\mathrm{mm}^3$ sample was fabricated. Initial ME measurements were made from this sample and then its side was sequentially sanded to reduce the sample width while all other dimensions remained the same. The results of this study are presented in figure 5, which plots the ME voltage coefficient measured at the first and third resonance frequencies as a function of W. The dependence of the third resonant frequency is very similar to that observed for a sample with a constant interfacial area (see figure 3). However, the results in figure 5 for the first resonant peak are distinctly different from those shown in figure 3. That is, the ME voltage coefficient corresponding to the first resonant frequency does not monotonically decrease with increasing W. The ME voltage coefficient has a maximum of $35\,\mathrm{V\,cm^{-1}\,Oe^{-1}}$ when the sample width is decreased to 13 mm. These results indicate that the size and the working frequency of ME composites should be optimized for making efficient ME devices. For example, for a $25 \times 25 \times 1.6\,\mathrm{mm}^3$ ME device, in order to gain the maximum ME effect, one should choose the third but not the first resonant frequency as the working frequency. In contrast, for a $13 \times 25 \times 1.6\,\mathrm{mm}^3$ ME device, one should choose the first but not the third resonant frequency as the working frequency.

Figure 6(*a*) shows the ME voltage coefficient frequency dependence for the square sample with $20 \times 20 \times 1.6\,\mathrm{mm}^3$ dimensions and figure 6(*b*) shows the isosceles right triangular

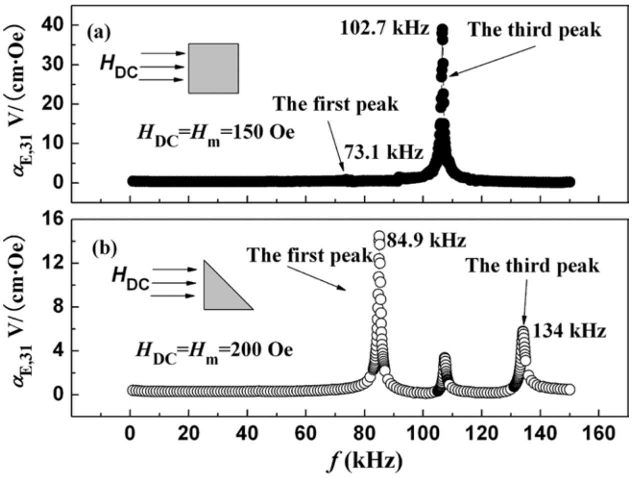

Figure 6. The dependence of frequency on the ME voltage coefficient for square (*a*) and triangular (*b*) samples with the dimension of 20 × 20 × 1.6 mm^3.

sample with the 20×20×1.6 mm^3 dimensions (where 20 mm is the side length and 1.6 mm is the total thickness of Ni and PZT layers combined). There are three resonant peaks obtained from the square and triangular samples. However, there are distinct differences between them: the relative heights of the first and third peaks (the value of the magnetoelectric voltage coefficient) are obviously different. In the square sample, the value of the first peak is extremely small, while the third peak is relatively large (40 $\text{V cm}^{-1}\,\text{Oe}^{-1}$). In contrast, the first peak height in the triangular sample is greater than that of the third peak: the first peak is 15 $\text{V cm}^{-1}\,\text{Oe}^{-1}$, and the third peak only reaches 7 $\text{V cm}^{-1}\,\text{Oe}^{-1}$. We believe that the differences between the first and the third peaks of the triangular and the square samples are related to their shape. As mentioned earlier, the first resonant longitudinal mode is along the diagonal direction in the rectangular layered sample. In the square sample, there are two diagonals, and the angle between them is 90°. There may be mutual weakening effects in these two diagonal directions, which reduce the ME response. However, there is only one long side (hypotenuse) in the right triangle; therefore, there is no mutual weakening effect, and the ME response is relatively high. The mutual weakening role of the two diagonals in the square samples maybe the reason for the suppressed ME effect at the first resonant frequency (sample dimensions of 20 × 20 × 1.6 mm^3 and 25 × 25 × 1.6 mm^3).

There is another obvious difference between the frequency curves of the square versus triangular samples: the three resonant frequencies are different. For the triangular sample, all resonant peaks move to the higher frequency compared with the square sample. We think the main reason for the difference is due to the masses of the samples. The mass of the triangular sample is smaller than that of the square sample; therefore, all resonant peaks shift to a higher frequency and the maximum magnetoelectric voltage coefficient is not the same. In the square sample, the maximum ME voltage coefficient is 40 $\text{V cm}^{-1}\,\text{Oe}^{-1}$ at the third resonance frequency. However, it is only 15 $\text{V cm}^{-1}\,\text{Oe}^{-1}$ for the triangular sample at the first frequency, and is associated with the sample demagnetization factor. The triangular sample demagnetization factor is larger than that of the square sample; therefore, the ME voltage coefficient of the square sample is larger than that of the triangular sample [21].

4. Summary

In summary, there are three resonant modes in rectangular symmetric trilayered ME composites. The first resonance peak corresponds to the longitudinal mode along the diagonal direction in a rectangle and not to the bending resonant mode traditionally considered for bilayered structures. The diagonal resonant mode causes mutual weakening of the ME effect down to zero at the first resonant frequency for a square sample. The ME voltage coefficient of layered ME composites scales with its in-plane dimensions. This study plays a guiding role for the design of ME devices for real applications.

Acknowledgments

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