ORIGINAL ARTICLE - ELECTRONICS, MAGNETICS AND PHOTONICS

Effect of Structural Control on the Magnetoelectric Characteristics of Piezoelectric–Magnetostrictive Laminate Composite in Resonance and Off-Resonance Modes

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Abstract

In this study, it is reported that the magnetoelectric (ME) performance in the resonance mode and that in the off-resonance mode are reversed depending on the thickness fraction of the piezoelectric layer in a ME laminate composite composed of piezoelectric and magnetostrictive layers. The ME performance in the resonance mode increased as the thickness fraction of the piezoelectric layer was increased, while the ME performance in the off-resonance mode decreased. Based on the impedance spectrum analysis of fabricated ME laminate samples, it was confirmed that the thickness fraction of the piezoelectric layer had a significant influence on the mechanical loss of the device. The phase angle change and the mechanical quality factor at the anti-resonance frequency of the ME laminate were found to be useful for predicting the ME performance in the resonance mode. High aspect ratio of the laminate was favorable in both resonance and off-resonance modes.

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Graphical Abstract
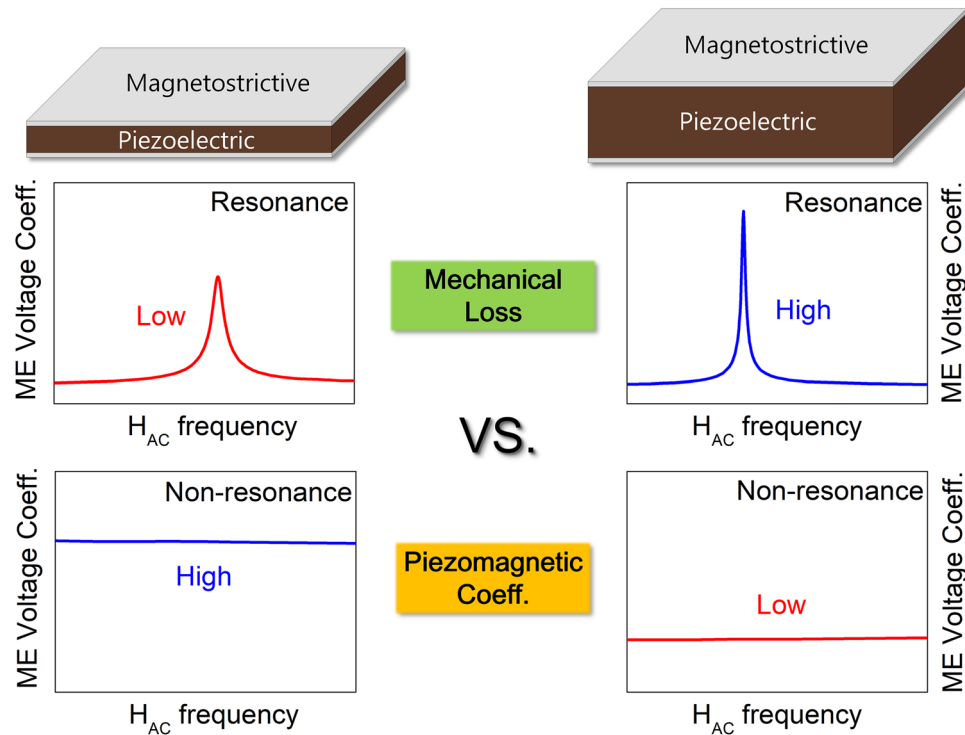
1 Introduction

Magnetoelectric (ME) composite materials composed of magnetostrictive metal layers and a piezoelectric ceramic layer are smart materials capable of mutual conversion between a magnetic field and an electric field by strain-coupling between a magnetostrictive layer and a piezoelectric layer. Studies on various applications using ME effects are actively being carried out [1–10].

ME voltage coefficient ($\alpha_{ME} = dE_{AC}/dH_{AC}$, E_{AC}: AC electric field, H_{AC}: AC magnetic field) is used as a representative index of the ME performance for converting a magnetic field into an electric field. Since the α_{ME} is a frequency dependent coefficient, its value may show a large difference in the resonance frequency regime and the off-resonance frequency regime of the piezoelectric layer constituting the ME composite. It is generally known that the α_{ME} is much larger in the resonance frequency regime than in the off-resonance frequency regime of the piezoelectric layer [1, 2], and it has been proven that the direct ME effect (dE_{AC}/dH_{AC}) is enhanced at the anti-resonance frequency (f_a) and the converse ME effect (dB_{AC}/dE_{AC}, B_{AC}: AC magnetic induction) is enhanced at the resonance frequency (f_r) [9, 11].

The ME effect in the off-resonance mode can be applied to sensor devices that detect tiny magnetic fields whose frequency is not specified, and many related experiments have been conducted [12–16]. In this case, it is important to obtain stable and high α_{ME} values over a wide frequency range. Meanwhile, researches have recently been performed on harvesting magnetic field energy having a specific frequency using the phenomenon that the α_{ME} value is significantly amplified in the resonance mode [17, 18]. In this case, it is advantageous to obtain a high α_{ME} and a low impedance at a specific frequency for high power generation. Therefore, fundamental study of device design for obtaining high performance in each device application using the ME effect is important.

In this study, the effect of aspect ratio and thickness control of 3-layered ME laminate composites with magnetostrictive/piezoelectric/magnetostrictive structure on ME performance in resonance and off-resonance mode is investigated. By comparison analysis between impedance and phase angle spectra and α_{ME} spectra of the ME laminate composite, it is demonstrated that the ME characteristics in resonance mode depend on the mechanical loss of the composite, and the magnitude of the phase angle change in the resonance frequency regime can represent the degree of amplification

of the α_{ME}. More importantly, it is demonstrated that the composite structure with a higher α_{ME} value in resonance mode exhibits a lower α_{ME} value in off-resonance mode than that of the other composite structures. Based on the findings, it is proposed that the composite structure for maximizing ME performance should be carefully designed according to the frequency mode to be used.
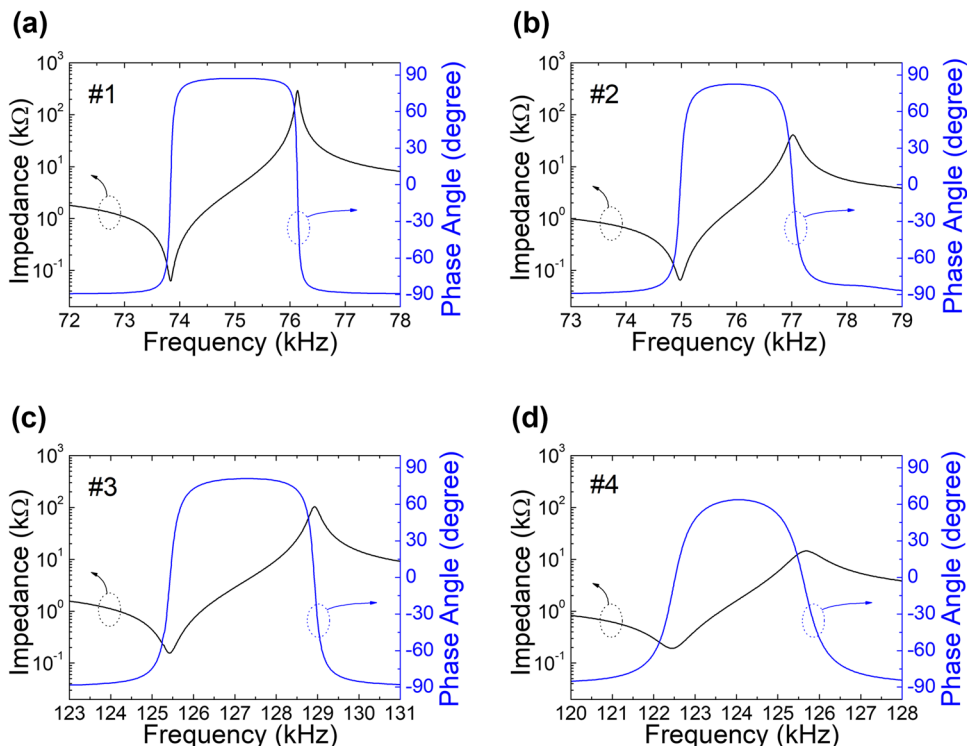
2 Experimental

Figure 1 shows a schematic diagram of the ME laminate composite devices fabricated in this study. The length/width aspect ratio of sample #1 is 5, and the thickness of the piezoelectric layer is 1 mm. Sample #2 has a thickness
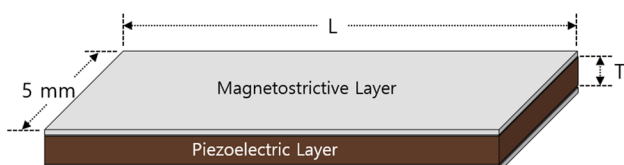
	Sample #1	Sample #2	Sample #3	Sample #4
L (mm)	25	25	15	15
T (mm)	1	0.45	1	0.45

Fig. 1 Schematic diagram and dimension information of magnetoelectric laminate composite fabricated in this study

of 0.45 mm with the same length/width aspect ratio as sample #1. Sample #3 has a length/width aspect ratio of 3 and a piezoelectric layer thickness of 1 mm. Sample #4 has a thickness of 0.45 mm with the same length/width aspect ratio as sample #3. The thickness of the magnetostrictive layer used for all samples is 50 μm. $0.95Pb(Zr_{0.52}Ti_{0.48})O_3–0.05Pb(Mn_{1/3}Sb_{2/3})O_3$ fabricated by conventional solid-state sintering method was used as the piezoelectric layer. An FeSiB-based amorphous alloy (Metglas, 2605SA1) was employed as the magnetostrictive material. The piezoelectric layer and the magnetostrictive layers were laminated using an epoxy adhesive (DP460, 3M). Impedance and phase angle spectra of the samples were measured using an impedance analyzer (IM3570, Hioki). To measure the ME voltage coefficients, the laminate composite was placed at the center of a Helmholtz coil located at the center of the electromagnet. The voltage induced on the laminate composite was monitored using a lock-in amplifier (SR860, Stanford Research System).

3 Results and Discussion

Figure 2 shows the impedance and phase angle spectra of the fabricated ME laminate samples. Samples #1 and #3 exhibited high overall impedance due to the thick piezoelectric layer thickness, and resonance peaks for 31-mode longitudinal vibration were observed at approximately 74–76 and 125–129 kHz, respectively. Samples #2 and #4 exhibited

Fig. 2 Impedance and phase angle spectra of magnetoelectric laminate composites: **a** sample #1, **b** sample #2, **c** sample #3, and **d** sample #4

lower overall impedance than that of samples #1 and #3 as the piezoelectric layer thickness decreased, and resonance peaks were observed at approximately 75–77 and 122–126 kHz,
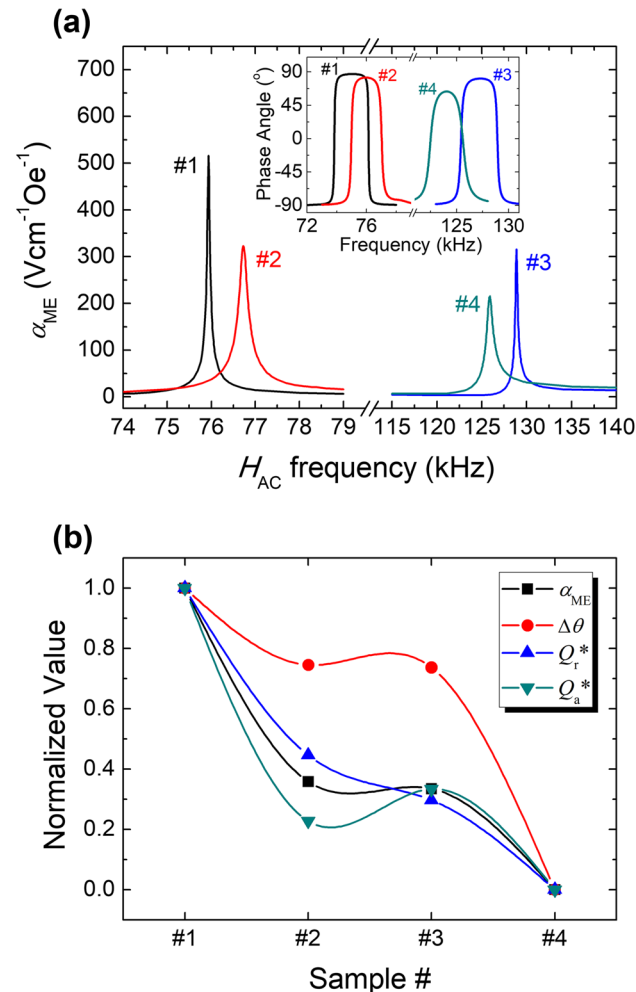

(a)

(b)

Fig. 3 **a** Magnetoelectric voltage coefficient of magnetoelectric laminate composites measured in the resonance frequency range (inset of **a** shows the phase angle spectra measured in the resonance frequency range), **b** normalized values of magnetoelectric voltage coefficient (α_{ME}), phase angle change ($\Delta\theta$), and effective mechanical quality factors at resonance frequency (Q_r^*) and at anti-resonance frequency (Q_a^*) of magnetoelectric laminate composites

respectively. For the phase angle change in the resonance frequency range, sample #1 exhibited the highest value of 176.5°; the other samples exhibited lower values.

Figure 3a shows the magnetoelectric voltage coefficient (α_{ME}) of the ME laminate samples as a function of the magnetic field (H_{AC}) frequency. The inset shows the phase angle spectra of the samples. For all four samples, a large amplification of α_{ME} was observed at the anti-resonance frequency of the piezoelectric layer [9]. Sample #1 exhibited a very high α_{ME} value of 515.7 V cm^{-1} Oe^{-1} in the resonance mode, while the other samples exhibited smaller α_{ME} values (Table 1). Note that the order of the α_{ME} magnitude in the resonance mode coincides with the order of the phase angle magnitude of the inset of Fig. 3a.

The fact that the phase angle change at the resonance frequency is less than 180° and the shape of the phase angle spectrum deviates from the rectangular shape can be related to the increase of the mechanical loss of the piezoelectric layer during the 31-mode operation. Sample #1 can effectively induce the 31-mode converse piezoelectric effect due to its high aspect ratio of 5 and high thickness fraction of the piezoelectric layer of 0.91. Conversely, for sample #2, the 31-mode motion of the piezoelectric layer is more disturbed by the magnetostrictive layer as the thickness fraction of the piezoelectric layer decreases to 0.82, so the mechanical loss can increase. In the case of sample #3, although the thickness fraction of the piezoelectric layer is the same as that of sample #1, the mechanical loss of the laminate can increase because the 31-mode vibration in the longitudinal direction is adversely affected by the vibration in the width direction as the aspect ratio is reduced to 3. Sample #4, which has the smallest aspect ratio and the smallest thickness fraction of the piezoelectric layer, exhibited the lowest phase angle change. Therefore, the highest mechanical loss of the device could be expected. From the resonance and anti-resonance peak information of the impedance spectra in Fig. 2, the expected mechanical loss (tan φ) for direct ME operation can be calculated by using the following equation [19]:

$$\tan \varphi = \Delta f / f, \tag{1}$$

Table 1 Length to width aspect ratio (L/W AR), thickness fraction of piezoelectric layer (T_{piezo}/T_{total}), phase angle change, magnetoelectric voltage coefficient (α_{ME}), mechanical loss (tan φ), and effective electromechanical coupling coefficient (k_{31}^*) of magnetoelectric laminate composites

	Sample #1	Sample #2	Sample #3	Sample #4
L/W AR	5	5	3	3
T_{piezo}/T_{total}	0.91	0.82	0.91	0.82
phase angle change (°)	176.5	169.2	169.0	148.0
α_{ME} (V cm^{-1} Oe^{-1}) (resonance)	515.7	322.9	315.7	215.1
α_{ME} (V cm^{-1} Oe^{-1}) (off-resonance)	0.22	0.61	0.19	0.48
tan φ (@ f_r)	0.00099	0.00181	0.00233	0.00550
tan φ (@ f_a)	0.00076	0.00236	0.00183	0.00610
k_{31}^*	0.244	0.229	0.232	0.226

where, f is the resonance frequency (f_r) or anti-resonance frequency (f_a), and Δf is the peak width at which the impedance value is $2^{0.5}$ times or $2^{-0.5}$ times the impedance value at the resonance or anti-resonance frequency, respectively. The tan φ values of the samples calculated from Eq. (1) are shown in Table 1. The inverse of these tan φ values are the effective mechanical quality factors of the ME laminate samples. Figure 3b shows the normalized values of α_{ME}, phase angle change ($\Delta\theta$), mechanical quality factor at resonance frequency (Q_r^*) and mechanical quality factor at anti-resonance frequency (Q_a^*) obtained in the resonance frequency range of the samples. The trend of the phase angle is useful for quickly predicting the α_{ME} trend in the resonance mode. However, as can be clearly seen in Fig. 3b, the tendency of Q_r^* or Q_a^* is more advantageous in accurately predicting the α_{ME} trend. In general, the mechanical quality factor of the piezoelectric device is obtained from the resonance frequency. However, because the direct ME effect is maximized at the anti-resonance frequency, the Q_a^* has a decisive influence on the α_{ME}. Another interesting point is that, as can be seen in Table 1, high thickness fraction of the piezoelectric layer (samples #1 and #3) resulted in lower tan φ at the f_a than at the f_r, whereas decreased thickness fraction of the piezoelectric layer (samples #2 and #4) resulted in higher tan φ at the f_a than at the f_r. This suggests that the higher the thickness fraction of the piezoelectric layer in a device using the direct piezoelectric effect, the more the mechanical loss can be minimized.

Another factor that can affect the mechanical loss of the ME laminate composite is the bonding state between the piezoelectric layer and the magnetostrictive layer [20, 21]. In this study, because all four samples were fabricated by the same process using the same epoxy bonding material, the bonding state of the four samples is not considered to be different. Therefore, the mechanical loss of the samples depends on the aspect ratio of the laminate and the thickness fraction of the piezoelectric layer, and directly affects the ME amplification in the resonance mode.

Conversely, in the off-resonance frequency range, samples #2 and #4 showed higher α_{ME} values than those of samples #1 and #3 as shown in Fig. 4a and Table 1, unlike in the resonance mode. The α_{ME} of the ME laminate composite composed of the 31-mode piezoelectric layer is expressed as follows [1]:

$$\alpha_{ME} = \frac{nq_{11}g_{31}}{nS_{11}^E\left(1 - k_{31}^2\right) + (1 - n)S_{11}^H},\qquad(2)$$

where n is the volume fraction of the magnetostrictive layer, q_{11} is the piezomagnetic coefficient, g_{31} is the piezoelectric voltage coefficient, k_{31} is the electromechanical coupling coefficient, and S_{11}^E and S_{11}^H represent the elastic compliance of the piezoelectric layer and the magnetostrictive
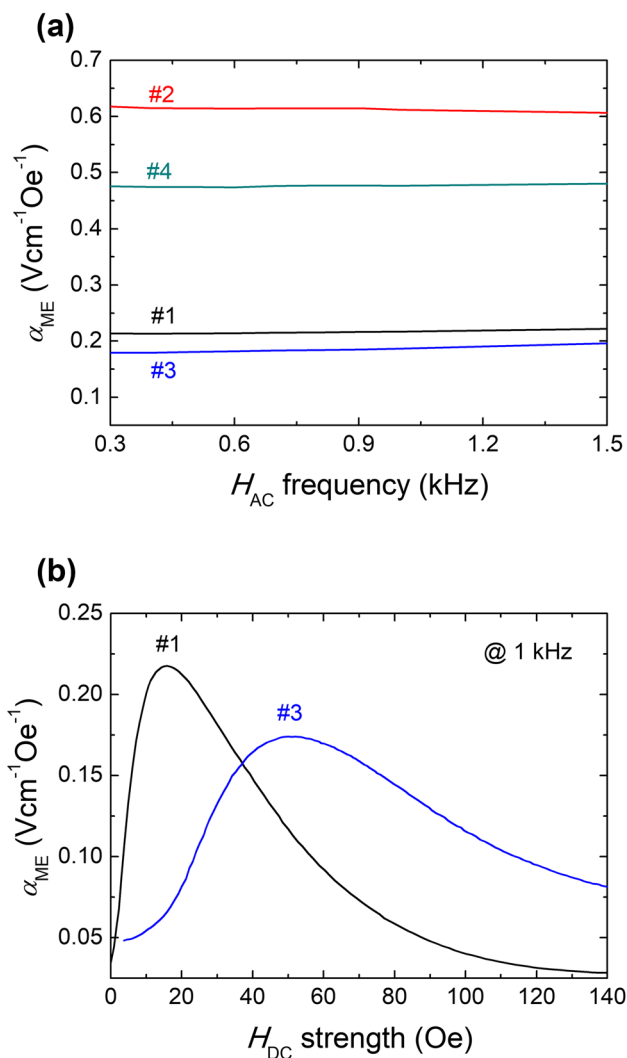

(a)

(b)

Fig. 4 Magnetoelectric voltage coefficient of magnetoelectric laminate composites: **a** measured in off-resonance frequency range and **b** measured as a function of magnetic bias field strength

layer, respectively. This equation does not include the parameters directly related to the resonance mode and thus can be considered to reflect the ME characteristics in the off-resonance mode. In the direct ME effect, the part that primarily responds to external stimuli is the magnetostrictive layer. If the deformation of the magnetostrictive layer is difficult, the direct ME effect will inevitably decrease. Therefore, in the interpretation of the results in Fig. 4a, the first factors to be noted in Eq. (2) are n and q_{11}. As n is increased, i.e., as the thickness fraction of the piezoelectric layer decreases, the magnetostrictive layer effectively deforms, increasing the q_{11} value. Thus, it is considered that n and q_{11} in Eq. (2) together contribute to the increase in the α_{ME}. When n is doubled from 0.09 (sample #1) to 0.18 (sample #2), the α_{ME} at 1 kHz increases more sharply from 0.22 to 0.61 V cm^{-1} Oe^{-1} (2.8 times increase), supporting

the joint contribution of n and q_{11} to the α_{ME}. As can be clearly seen in Fig. 4a and Eq. (2), the α_{ME} increases with increasing n in the off-resonance mode, but the α_{ME} varies if the aspect ratio of the ME laminate is changed even if the n values are the same (Table 1). The larger the α_{ME} value of the sample with the higher aspect ratio means the aspect ratio has a direct impact on q_{11} [8].

The electromechanical coupling coefficient is an important factor in determining the efficiency of the piezoelectric device. From the impedance spectra of Fig. 2, the effective electromechanical coupling coefficient ($k_{31}*$) of the laminate composite samples was obtained from the following equation (Table 1) [21].

$$k_{31}^* = \sqrt{(f_a^2 - f_r^2)/f_a^2} \qquad (3)$$

When $k_{31}*$ values alone are compared, sample #1 has the highest piezoelectric conversion efficiency. However, if we compare the degree of contribution to the α_{ME} value $(1 - k^2)$ of Eq. (2), there is no significant difference between the four samples. Therefore, the characteristics of the magnetostrictive layer mainly affect the ME performance in the off-resonance mode. However, in the resonance mode, the $k_{31}*$ value is also important for enhancing the ME performance along with the mechanical quality factor.

The fact that the resonance mode ME characteristic and the off-resonance mode ME characteristic tend to be opposite to each other in the laminate samples made from the same materials is remarkable in designing a high performance ME device. In order to utilize the off-resonance mode, the structure in which the magnetostrictive layer effectively deforms is advantageous. However, in order to utilize the amplified ME effect in the resonance mode, a structure capable of minimizing the mechanical loss of the piezoelectric layer is advantageous. From the viewpoint of thickness fraction, the structure with higher thickness fraction of the piezoelectric layer is more advantageous in the resonance mode, and the structure with higher thickness fraction of the magnetostrictive layer is more advantageous in the off-resonance mode. High aspect ratio of the laminate is appropriate for both resonance and off-resonance mode because it is advantageous for both low mechanical loss and high piezomagnetic coefficient. Furthermore, as shown in Fig. 4b, the higher the aspect ratio, the smaller the magnetic bias field strength at which the maximum α_{ME} appears, which is more advantageous for miniaturization of the device.

4 Conclusions

In this study, the ME laminate samples were fabricated by varying the aspect ratio of the laminate and the thickness fraction of the piezoelectric layer, and the ME characteristics in the resonance and off-resonance modes were compared and analyzed. A strong amplification of the ME effect was observed at the anti-resonance frequency of the samples, and the degree of amplification was consistent with the magnitude of the phase angle change, which was affected by the mechanical loss in the resonance mode. The smaller the thickness fraction of the piezoelectric layer and the lower the aspect ratio of the laminate, the larger the mechanical loss and the smaller the ME amplification in the resonance mode. However, the α_{ME} in the off-resonance mode was higher as the thickness fraction of the piezoelectric layer was lower, demonstrating that the device design for obtaining high ME performance in the resonance mode and the off-resonance mode is different from each other.

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