

# Height-related magnetoelectric performance of PZT/Ni layered composites

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**Abstract** The effect of height on performance of the PZT/Ni cylindrical bilayered magnetoelectric (ME) composites was studied in situ in this paper. Multiple resonant peaks appear between 1 and 300 kHz frequency for cylinders of different heights. The first resonance frequency does not change with the cylinder height decreasing, but the second and the third resonant frequencies increase. The first three resonant modes are attributed to the cylinder radial, first-order height resonance, and second-order height resonance, respectively. The appropriate size and resonance frequency were chosen to obtain the highest ME voltage coefficient when designing cylindrical bilayered magnetoelectric devices. This article provides reference to design cylindrical magnetoelectric devices.

**Keywords** Cylindrical layered composites; Height; Resonant mode; Demagnetization factor

## 1 Introduction

Multifunctional materials draw much attention due to their integrated properties, such as magnetoelectric (ME) [1, 2], magnetocaloric, magnetoelastic, magneto-optical, magneto-semiconducting effect, etc. Numerous studies related to these materials were reported. Wang et al. [3] investigated the electric

and magnetic fields effects on the current spin polarization and magnetoresistance in a ferromagnetic/organic semiconductor/ferromagnetic system. Li et al. [4] studied the temperature stability of magnetoresistance in  $(1-x)\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3/x\text{Ag}$ . Magnetocaloric effect of  $\text{La}_{0.6}\text{Pr}_{0.4}\text{Fe}_{11.4}\text{Si}_{1.6}\text{B}_{0.2}$  alloy and its hydride were investigated by Ge et al. [5]. Magnetocaloric and barocaloric effects in a  $\text{Gd}_5\text{Si}_2\text{Ge}_2$  compound were studied [6].

Among the multifunctional materials, the ME materials provide significant potential for applications in devices operating at low frequencies, such as a pico-Tesla sensitivity magnetometer [7, 8], and at microwave frequencies, such as electrostatically tunable band-pass filters [9], band-stop filters [10], resonators [11], phase shifters [12], etc. Most of the devices are based on the layered ME composites, which exhibit stronger ME effect compared with the particulate ME materials [13].

Electro-deposited plate layered ME composites have comparable ME performance to the  $\text{Tb}_{1-x}\text{Dy}_x\text{Fe}_{2-y}$  (Terfenol-D)/PZT prepared by the gluing method [14, 15]. The electrodeposition becomes a major method of preparing the ME composites. In previous studies, plate and cylindrical layered ME composites made by electro-deposition were considered [14–20]. Cylindrical layered ME composites exhibit large gain ME effect at resonance frequency under high magnetic field induced by the volume magnetostriction [16–20], which provides significant potential for applications in high magnetic field detection. Recently, it is proposed that the cylindrical ME composites exhibit more superior performance than the plate composites [16]. The ME coefficient between the PZT/Ni bilayered and Ni/PZT/Ni trilayered structures was studied in detail. There are different superior performances between them at different resonant frequencies [19]. In the present work, the effect of height on ME performance of the cylindrical ME composites with PZT/Ni bilayered structure was studied.

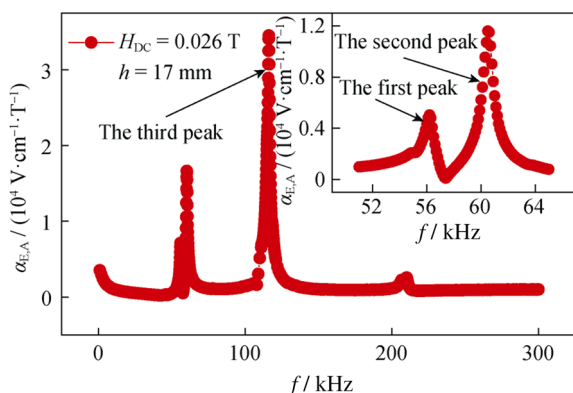
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## 2 Experimental

A commercial hollow  $\text{Pb}(\text{Zr}_{0.52}\text{Ti}_{0.48})\text{O}_3$  (PZT-5H, supplied by the Institute of Acoustics, Chinese Academy of Sciences) cylinder with a height of 17 mm, an inner diameter of 18 mm, and an outer diameter of 20 mm was electro-deposited with nickel on the outer surface to make a PZT/Ni cylinder ME composition. The composition of the plating solution and electro-deposition was described in detail elsewhere [14–17]. The total thickness of Ni was about 0.4 mm.

For ME measurements, samples were subjected to a bias magnetic field,  $H_{\text{DC}}$ , superimposed with a sinusoidal field  $\delta H$  (frequency range from 1 to 300 kHz). A voltage,  $\delta V$ , generated across the PZT cylinder thickness was amplified and measured with an oscilloscope. The ME voltage coefficient was calculated as  $\alpha_E = \delta V \cdot (t_{\text{PZT}} \cdot \delta H)^{-1}$ , where  $t_{\text{PZT}}$  is the PZT thickness and  $\delta H$  is the change of the applied magnetic field.  $\alpha_{E,A}$  can be obtained with  $H_{\text{DC}}$  and  $\delta H$  applied along the cylinder's vertical axis [16].



**Fig. 1** Dependence of magnetoelectric voltage coefficient ( $\alpha_{E,A}$ ) on applied magnetic field frequency for PZT/Ni bilayered cylindrical ME composite

## 3 Results and discussion

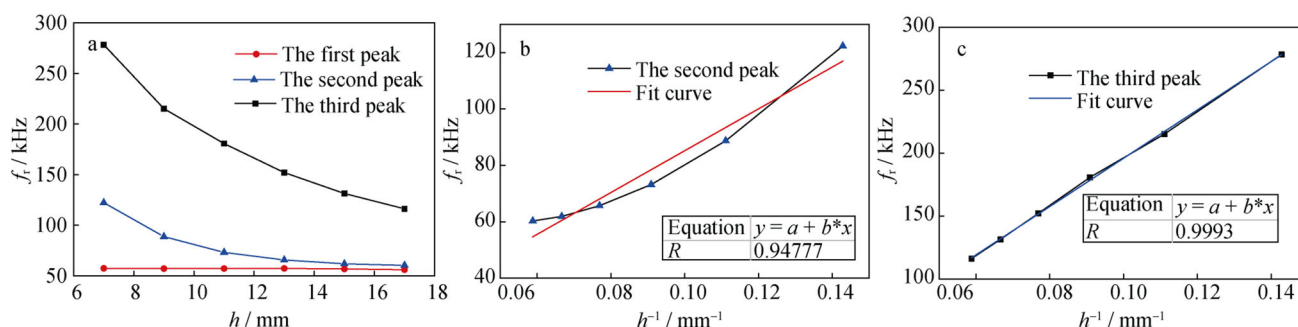
The dependence of magnetoelectric voltage coefficient  $\alpha_{E,A}$  on the applied magnetic field frequency for the PZT/Ni bilayered cylindrical ME composite with the height of 17 mm and  $H_{\text{DC}} = H_m = 0.026$  T is shown in Fig. 1, where  $H_m$  is the applied magnetic field when  $\alpha_{E,A}$  reaches the peak value. There are multiple resonant peaks in the ME voltage coefficient curve as a function of the frequency of the applied magnetic field varying from 1 to 300 kHz. Figure 1 shows that two resonant peaks appear at about 56 and 60 kHz, which look like twin peaks. A strong resonant peak also appears at about 116 kHz. All of the resonant frequencies are denoted as  $f_r$ . The ME coefficient decreases with the increase of frequency in the range of 0–20 kHz according to the reported studies [21, 22].

In order to study the height effect on the ME performance of the cylinder, initial ME measurements were performed with the original sample, and then its height was sequentially reduced by sanding, while all other dimensions remained the same. Figure 2a shows the resonance frequency ( $f_r$ ) dependence on the height ( $h$ ) of the cylinder ranged from 17 to 7 mm for the first three peaks. Figure 2a indicates that  $f_r$  increases with the decrease of  $h$  for the second and third peaks. However, for the first peak,  $f_r$  does not change with the decrease of  $h$ . For the second resonant peak, the resonance frequency increases linearly with  $h^{-1}$  ( $R = 0.94777$ ) decreasing as shown in Fig. 2b. Figure 2c shows that the third resonance frequency increases linearly with  $h^{-1}$  ( $R = 0.9993$ ) decreasing. The resonance frequencies are calculated by corresponding formulas as follows [23]:

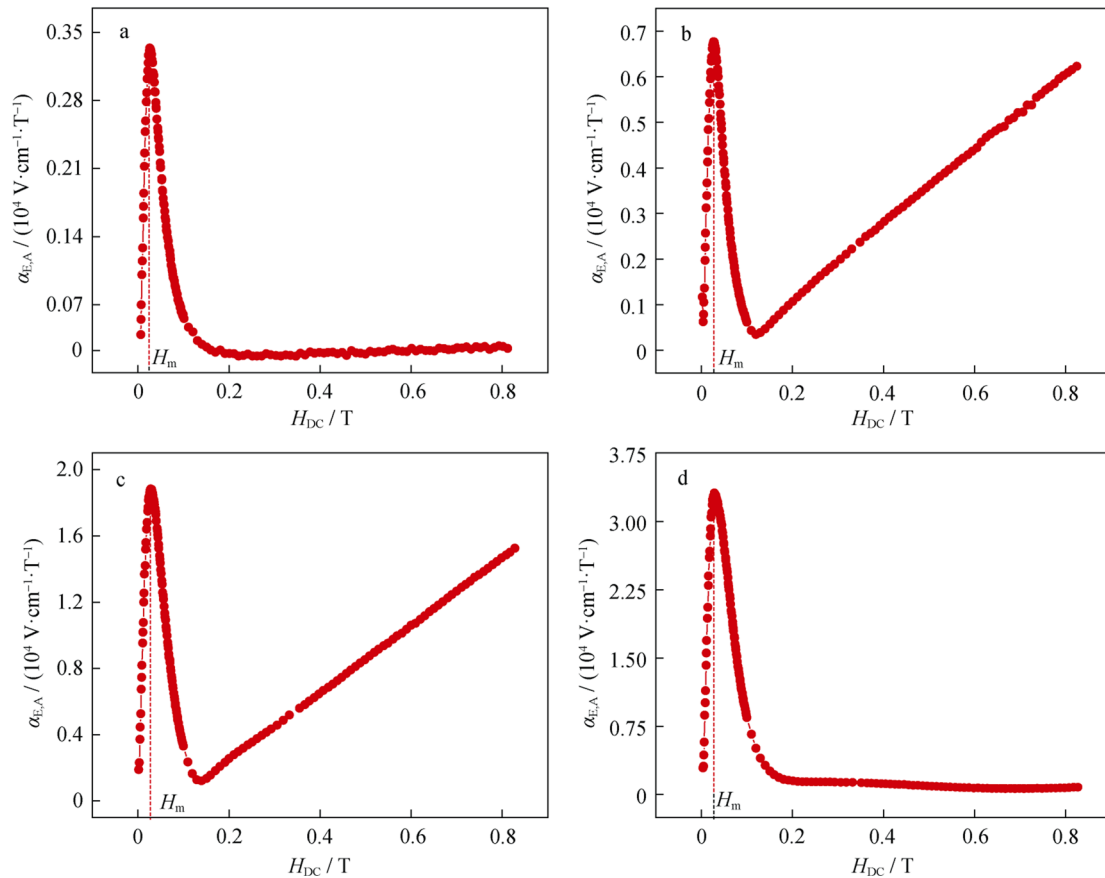
Radial mode resonance frequency:

$$f_r^d = \frac{1}{\pi d} \sqrt{\frac{1}{\rho s_{33}}} \quad (1)$$

Height mode resonance frequency:



**Fig. 2** Dependence of resonance frequency ( $f_r$ ) on height ( $h$ ) of cylinder reducing from 17 to 7 mm of the three peaks **a**, fit curve of the second peak **b**, and fit curve of the third peak **c**



**Fig. 3** Dependence of magnetoelectric voltage coefficient ( $\alpha_{E,A}$ ) on bias magnetic field  $H_{DC}$  varying from 0 to 0.8 T: **a** 1.0 kHz, **b** 56.0 kHz, **c** 60.3 kHz, and **d** 116.2 kHz

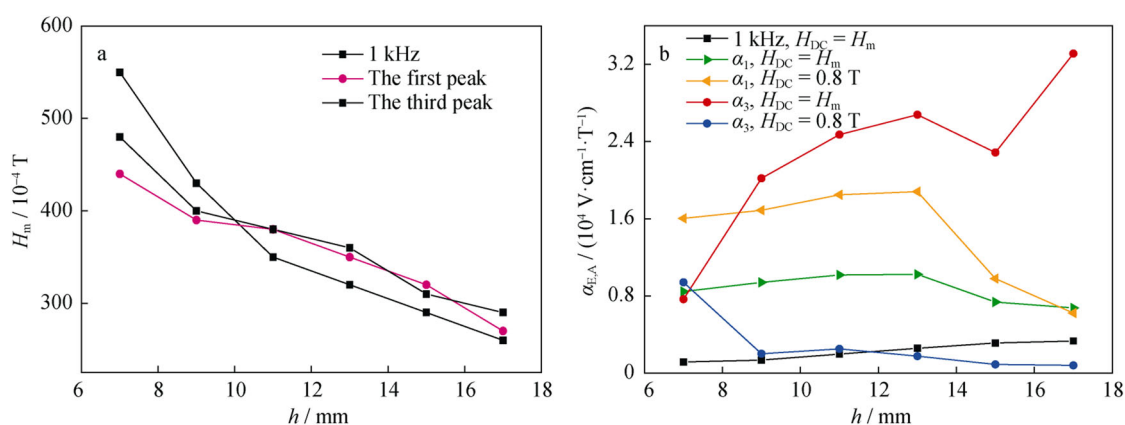
$$f_r^h = \frac{n}{\pi h} \sqrt{\frac{1}{\bar{\rho} \bar{s}_{33}}} \quad (2)$$

$$\bar{v} = \sqrt{\frac{1}{\bar{\rho} \bar{s}_{33}}} \quad (3)$$

where  $\bar{\rho}$  is the average density of the bilayered composite,  $\bar{s}_{33}$  is the equivalent elastic compliance,  $d$  is the diameter of the cylindrical ME composite,  $n$  is the order of resonance mode, and  $\bar{v}$  is the average acoustic velocity of the composite [24]. When  $\bar{v} = 3,500 \text{ m}\cdot\text{s}^{-1}$ ,  $f_r^d = 58.64 \text{ kHz}$  which is close to the first resonance peak frequency (56 kHz); when  $n = 1$ ,  $f_r^h = 65.53 \text{ kHz}$  which is close to the second resonance peak frequency (60.3 kHz); when  $n = 2$ ,  $f_r^h = 131.06 \text{ kHz}$  which is close to the third resonance peak frequency (116.2 kHz). The fitted curves are in good agreements with the experiment results. Therefore, the first three resonant modes are attributed to the cylinder radial, first-order height resonance, and second-order height resonance, respectively.

The dependence of magnetoelectric voltage coefficients  $\alpha_{E,A}$  on the bias magnetic field  $H_{DC}$  varying from 0 to 0.8 T

at 1 kHz and the first three resonant frequencies for the PZT/Ni cylindrical composites is shown in Fig. 3. With the rise of  $H_{DC}$ , there is a sharp peak in the  $\alpha_{E,A}$  field dependence at 1 kHz and resonant frequencies under  $H_m$ . However, with  $H_{DC}$  increasing continuously,  $\alpha_{E,A}$  increases slowly at 1 kHz (Fig. 3a), and quickly increases linearly at the first (Fig. 3b) and the second (Fig. 3c) resonant frequencies, but tends to 0 at the third resonance frequency (Fig. 3d). In the previous study, it was proposed that the total ME effect was the sum of  $\alpha_{E(\lambda)}$  caused by line magnetostriction under low fields and  $\alpha_{E(\omega)}$  induced by volume magnetostriction under high fields [15]. The linear increase of  $\alpha_{E,A}$  with high applied magnetic field is associated with the volume magnetostriction. Therefore, the giant ME voltage coefficients under the high magnetic field at the first and the second peaks are induced by volume magnetostriction. However, the giant ME voltage coefficient is not obtained at the third peak. The third peak corresponds to the second-order height resonance mode, as previously discussed. The second-order height resonance may be suppressed under high applied magnetic field, unlike the first-order cylinder radial and first-order height resonance.



**Fig. 4** Dependence of  $H_m$  **a** and  $\alpha_{E,A}$  **b** on  $h$  varying from 17 to 7 mm

Figure 4a shows the dependence of  $H_m$  on  $h$  varying from 17 to 7 mm at 1 kHz of the first and the third resonant frequencies for the PZT/Ni layered cylindrical composites.  $H_m$  increases with the decrease of the cylinder height. The magnitude and field dependences of  $\alpha_E$  are related to many factors, such as the large magnetoelectric susceptibility of the composites and the demagnetization field variation [25]. For the ferromagnetic materials, the demagnetization field will increase with the size decreasing when keeping other dimensions the same [19, 26]. The higher applied magnetic field induces higher demagnetization field. It can be concluded that decreasing the height of the cylinder will increase the working magnetic field in order to obtain better ME performance at the same working frequency.

Figure 4b shows the dependence of magnetoelectric voltage coefficient  $\alpha_{E,A}$  on  $h$  varying from 17 to 7 mm at 1 kHz of the first and the third resonant frequencies for the PZT/Ni cylindrical composites with  $H_m$  and 0.8 T applied magnetic fields.  $\alpha_1$  and  $\alpha_3$  correspond to the first and the third resonant frequencies, respectively.  $\alpha_{E,A}$  decreases with the decrease of  $h$  when  $H_{DC} = H_m$  under non-resonance frequency of 1 kHz. For the applied first resonance frequency,  $\alpha_{E,A}$  initially increases and then decreases subsequently with  $h$  decreasing at  $H_m$  and 0.8 T applied magnetic fields.  $\alpha_{E,A}$  reaches the maximum value at  $H_m$  and 0.8 T when  $h$  is sanded to 13 mm. The ME performance at 0.8 T applied magnetic field is better than that at  $H_m$ . The difference between the measured  $\alpha_{E,A}$  of the sample with specific  $h$  under applied magnetic fields  $H_m$  and 0.8 T becomes larger with the decrease of  $h$ . Under the cylinder radial resonance mode, there exists an optimal aspect ratio for a cylindrical ME composite with specific diameter when the composite resonates under cylinder radial mode. However, the changes in  $\alpha_{E,A}$  are adverse at  $H_m$  and 0.8 T for the third applied resonance frequency.  $\alpha_{E,A}$  decreases at  $H_m$ , and mildly changes at 0.8 T. The giant ME voltage coefficient is not obtained at the second-

order height resonance mode due to the suppression effect of high applied magnetic field as mentioned above. On the contrary, the difference becomes smaller with the decrease of  $h$ . Figure 4b indicates that  $\alpha_1$  is smaller than  $\alpha_3$  when  $h$  is not less than 9 mm, but is larger when  $h$  is less than 9 mm. These results indicate that the appropriate size and applied resonance frequency should be chosen to obtain the highest ME voltage coefficient when designing the ME devices.

## 4 Conclusion

In this work, the effect of height on the ME performance of PZT/Ni cylindrical ME composites was analyzed. Multiple resonant peaks appear between 1 and 300 kHz. The first three resonant modes are attributed to the cylinder radial, first-order height resonance, and second-order height resonance, respectively. The size and applied resonance frequency should be chosen appropriately to obtain the optimal ME voltage coefficient when designing ME devices.

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