

# Resonant modes and magnetoelectric performance of PZT/Ni cylindrical layered composites

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**Abstract** Resonant modes and magnetoelectric performance of layered PZT/Ni and Ni/PZT/Ni cylindrical composites are considered. The first and the second resonant frequencies in the 1–150 kHz range correspond to the axial and the radial modes. Experimental results and theoretical analysis indicate that one should choose the trilayered structure and the first resonant frequency as the working frequency. This study is helpful in design and applications of magnetoelectric devices.

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## 1 Introduction

Bulk magnetoelectric (ME) materials can be utilized in devices operating at low frequencies (pico-Tesla sensitivity magnetometer [1, 2]) and at microwave frequencies (electrostatically tunable band-pass filters [3], band-stop filters [4], resonators [5], phase shifters [6]). Most of the devices are based on the layered ME composites, which exhibit stronger ME effect compared with the bulk particulate ME materials [7].

In previous studies plate and cylindrical layered ME composites made by electro-deposition were considered [8–11]. Electrodeposited plate layered ME composites have comparable ME performance to the Terfenol-D/PZT prepared by the gluing method [8, 9]. Nickel (Ni)/lead zirconate titanate (PZT)/Ni trilayered composites also have some novel ME properties [10], while cylindrical Ni/PZT bilayered composites exhibit high gain ME effect at resonant frequency under high magnetic field induced by the volume magnetostriction and possibly induction effects [11]. Cylindrical layered ME composites have a significant potential for applications in high magnetic field detection. In this paper, the resonant modes and magnetoelectric performance of the cylindrical magnetoelectric composites with PZT/Ni bilayered and Ni/PZT/Ni trilayered structure are studied.

## 2 Experimental

A hollow  $\text{Pb}(\text{Zr}_{0.52}\text{Ti}_{0.48})\text{O}_3$  (PZT-5H) cylinder (supplied by Institute of Acoustics, Chinese Academy of Sciences) was cut with a height of 8 mm, an inner diameter of 18 mm, and an outer diameter of 20 mm (1-mm PZT cylinder wall thickness). PZT was polarized at 425 K in a 50 kV/cm electric field along the radial directions after being coated with conductive plastic. The poled PZT cylinder was cleaned from the plastic prior to Ni deposition on its surface. Then, the PZT cylinder was bathed in nickel aminosulfonate plating solution to electro-deposit the Ni layer. The composition of the plating solution is described in detail elsewhere [8–11]. For the PZT/Ni bilayered structure, after the cylinder inner wall was protected by the silicone rubber, only the outer surface Ni electrode was deposited. For the Ni/PZT/Ni trilayered structure, Ni was electro-deposited on

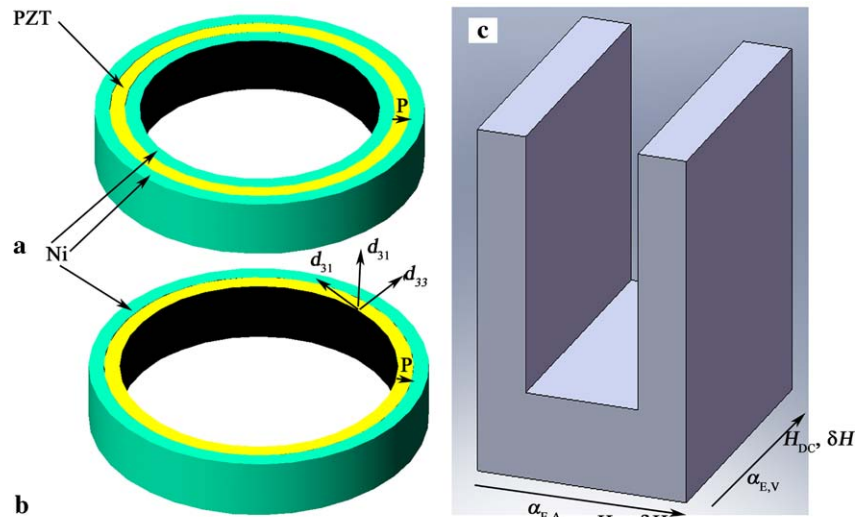
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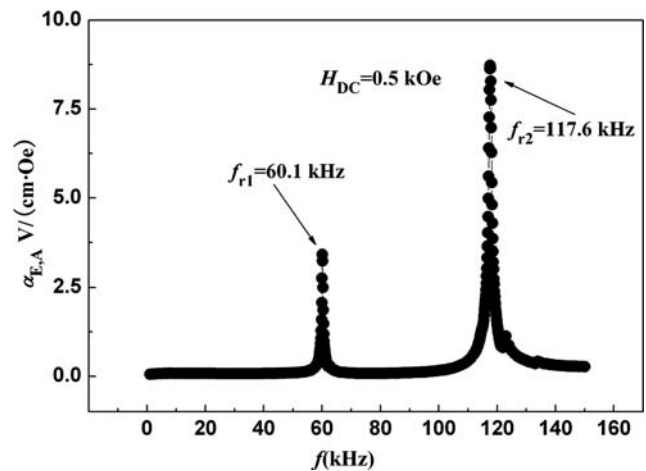
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**Fig. 1** A schematic diagram for (a) trilayered and (b) bilayered hollow cylindrical layered composite structures and (c) clamp [10]. Here,  $d_{31}$  and  $d_{33}$  are the piezoelectric constants of the PZT cylinder



both hollow cylinder sides. During electro-deposition, a cathodic current density of  $5 \text{ mA/cm}^2$  was applied to both samples (a cathodic current of 25 mA and 48 mA was applied to PZT/Ni and Ni/PZT/Ni structures, respectively) [12]. In order to obtain the same mass of the Ni layers, electro-deposition time of about 100 and 190 hours was used for trilayered and bilayered structures, respectively. The product of cathodic current and electro-deposition time was constant for the two samples. The electro-deposition rates were about 0.01 mm/h and 0.005 mm/h for trilayered and bilayered structures, respectively. The resulting total Ni thickness was about 1 mm for both samples, with 1-mm outer Ni layer for the bilayered sample, and 0.5-mm outer and inner Ni layers for the trilayered sample. Samples are shown schematically in Fig. 1 (a and b).

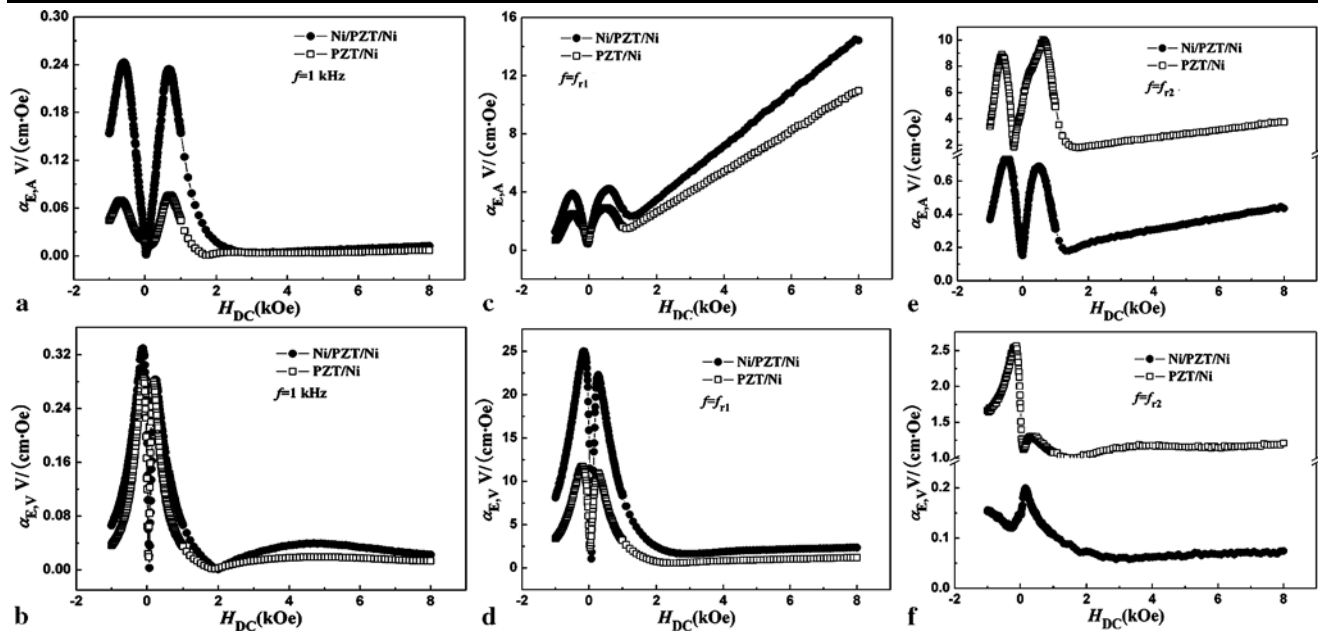
During the ME measurements, samples were subjected to a bias magnetic field  $H_{DC}$  superimposed with a sinusoidal field  $\delta H$  (1–150 kHz frequency range). The sinusoidal magnetic field with the amplitude  $\delta H = 22 \text{ Oe}$  was generated by the Helmholtz coils when AC current with the 1-A amplitude was passed through the coils. Voltage,  $\delta V$ , generated across the PZT cylinder thickness was amplified and measured with an oscilloscope. The ME voltage coefficient was calculated according to  $\alpha_E = \delta V / (t_{PZT} \cdot \delta H)$ , where  $t_{PZT}$  is the PZT thickness, and  $\delta H$  is the change in the applied magnetic field. Details about the measurement system have been reported in our previous paper [13]. Each sample was placed in a nylon clamp shown schematically in Fig. 1(c). The groove in the clamp is 8.5-mm wide, 15-mm deep, and 20-mm high. The clamp was fixed at the bottom, and the sample was hanging from its top supported by the voltage measuring wires. Two ME voltage coefficients,  $\alpha_{E,V}$  and  $\alpha_{E,A}$ , were obtained, corresponding to two conditions, where  $H_{DC}$  and  $\delta H$  were applied either along or perpendicular to the groove, as shown in Fig. 1(c).



**Fig. 2** The dependence of the magnetoelectric voltage coefficient  $\alpha_{E,A}$  on the applied magnetic field frequency for PZT/Ni bilayered cylindrical ME composites at the applied bias magnetic field  $H_{DC}$  of 0.5 Oe.  $f_{r1}$  and  $f_{r2}$  are the first and second resonant frequencies

### 3 Results and discussion

The magnetoelectric voltage coefficient  $\alpha_{E,A}$  dependence on the applied magnetic field frequency for the PZT/Ni bilayered cylindrical ME composite with  $H_{DC} = 0.5 \text{ kOe}$  is shown in Fig. 2. There are two resonant peaks in the ME voltage coefficient curve as a function of the frequency of the applied magnetic field in the 1–150 kHz range. We denote these frequencies as  $f_{r1}$  and  $f_{r2}$ , corresponding to the first and the second resonant peaks, respectively. The trilayered structure also exhibits two resonant peaks in the  $H_{DC}$  curve for the same 1–150 kHz frequency range. In this study, we will emphasize the ME effect ( $\alpha_{E,A}$  and  $\alpha_{E,V}$ ) dependence on the bias magnetic field ( $H_{DC}$ ) and the frequency of the applied magnetic field ( $f$ ).



**Fig. 3** The dependence of the magnetoelectric voltage coefficients  $\alpha_{E,A}$  (a, c, and e) and  $\alpha_{E,V}$  (b, d, and f) on the bias magnetic field  $H_{DC}$  varied from  $-1$  to  $8$  kOe at  $1$  kHz and on the first resonance frequency

$f_{r1}$  and the second frequency  $f_{r2}$  for the Ni/PZT/Ni and PZT/Ni layered cylindrical composites, respectively

The magnetoelectric voltage coefficients dependence on the bias magnetic field  $H_{DC}$  varied from  $-1$  to  $8$  kOe at  $1$  kHz for the Ni/PZT/Ni and PZT/Ni cylindrical composites  $\alpha_{E,A}$  and  $\alpha_{E,V}$  is shown in Fig. 3 (a and b). Figure 3 indicates that  $\alpha_{E,A}$  and  $\alpha_{E,V}$  are different for the bilayered and the trilayered structures at  $1$  kHz. The ME voltage coefficients  $\alpha_E$  initially increased and then decreased subsequently with  $H_{DC}$  at negative magnetic field when the applied frequency was  $1$  kHz for bilayered and trilayered samples. With  $H_{DC}$  increasing continuously, the  $\alpha_E$  trend for both samples is the same as reported in the previous work [10]. However, there appear two peaks in the  $H_{DC}$  curve in the vertical mode at positive fields.

This effect is induced by the difference in series of infinitesimal plate units and is discussed in the previous work [14]. The cylindrical layered ME composite axial coupling mode can be simplified as a plate trilayered ME composite longitudinal mode (Fig. 4). The dimensions of the simplified plate trilayered ME composite are  $h \times L^{\text{eff}} \times t$ , where  $h$  is the cylinder height,  $L^{\text{eff}}$  is the effective length of the simplified plate, and  $t$  is the total sample thickness corresponding to the cylinder wall thickness. After simplification, the effective magnetic fields  $H^{\text{eff}}$  and  $\delta H^{\text{eff}}$  are along the  $h$  direction, as shown in Fig. 4. The cylinder in the vertical mode also can be simplified as a plate trilayered ME composite with the effective magnetic fields acting along the thickness ( $t$ ) and the effective length ( $L^{\text{eff}}$ ) directions of the plate, simultaneously [14]. In the plate layered ME composites, there was a maximum at low magnetic field

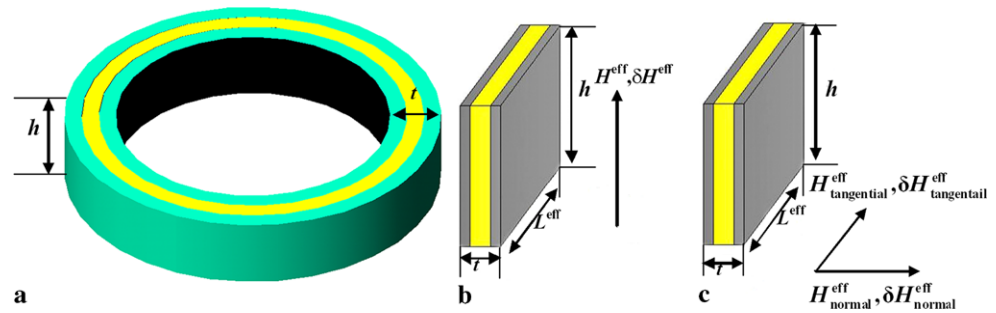
of the  $H_{DC}$  curves in  $\alpha_{E,31}$  (length direction,  $L^{\text{eff}}$ ) and  $\alpha_{E,32}$  (width direction,  $h$ ). However, there was a maximum at high magnetic field of the  $H_{DC}$  curves in  $\alpha_{E,33}$  (thickness direction,  $t$ ) [8, 15, 16]. That is why there appears one peak in the axial mode but two peaks in the vertical mode at positive fields.

There is a difference between the two samples in the axial mode. That is, at the same bias magnetic field below  $2$  kOe, the  $\alpha_E$  of the trilayered structure is larger than that of the bilayered sample, and passing  $2$  kOe, this difference becomes smaller. However, in the vertical mode, the ME voltage coefficients of both samples are similar for the same bias magnetic field below  $2$  kOe, and the ME effect in the trilayered sample is a little bit larger than in the bilayered sample above  $2$  kOe.

Figure 3(c, d) shows the dependence of the magnetoelectric voltage coefficients  $\alpha_{E,A}$  and  $\alpha_{E,V}$  on  $H_{DC}$  varied from  $-1$  kOe to  $8$  kOe at  $f_{r1}$  applied frequency for the Ni/PZT/Ni and PZT/Ni layered cylindrical composites. Figure 3(c) shows that the  $\alpha_{E,A}$  variation below  $1$  kOe is the same as in Fig. 3(a) for the two samples. However, with the increasing bias magnetic field,  $\alpha_{E,A}$  linearly increases. This novel ME effect is caused by the Ni volume magnetostriction under high magnetic field [11].

For ferromagnetic materials, like Ni, the line magnetostriction,  $\lambda = \Delta l/l$ , increases with bias magnetic field,  $H_{DC}$ , and then reaches a saturation value  $\lambda_s$  at  $H_s$ . When  $H_{DC} < H_s$ , the volume changes, while the volume magnetostriction  $\omega = \Delta V/V$  is too small to be measured [17].

**Fig. 4** (a) Trilayered cylinder; (b) trilayered plate composite simple bound state applied for the cylindrical layered composite in the axial coupling mode; and (c) in the vertical coupling mode [14]



When  $H_{DC} > H_s$ , however, the volume magnetostriction increases with applied magnetic field,  $H_{DC}$  [18]. Under low magnetic field, i.e.,  $H_{DC} < H_s$ , the magnetic field dependence of the ME voltage coefficient  $\alpha_E$  is determined by the variation of the piezomagnetic coupling  $q$  with the magnetic field  $H_{DC}$  [19], and  $\alpha_E$  is proportional to  $q$ , i.e.,  $\delta\lambda/\delta H$ , where  $\delta\lambda$  is the differential magnetostriction. When  $H_{DC} = H_s$ ,  $\lambda = \lambda_s$ , and then  $\delta\lambda/\delta H = 0$ ; therefore, the  $\alpha_{E(\lambda)}$  caused by the line magnetostriction is equal to zero under high applied magnetic field. The volume magnetostriction of ferromagnetic Ni phase under high bias magnetic field of  $H_{DC} > H_s$  can also generate stress in the piezoelectric PZT phase, resulting in voltage increase across the PZT. According to the field dependence of the ME voltage coefficient at resonant and nonresonant states in our experiments, the line magnetostriction is saturated below a bias field of 1 kOe, and thus it does not contribute to the ME effect under higher magnetic field. The strong ME coefficient under high magnetic field may be induced by the volume magnetostriction.

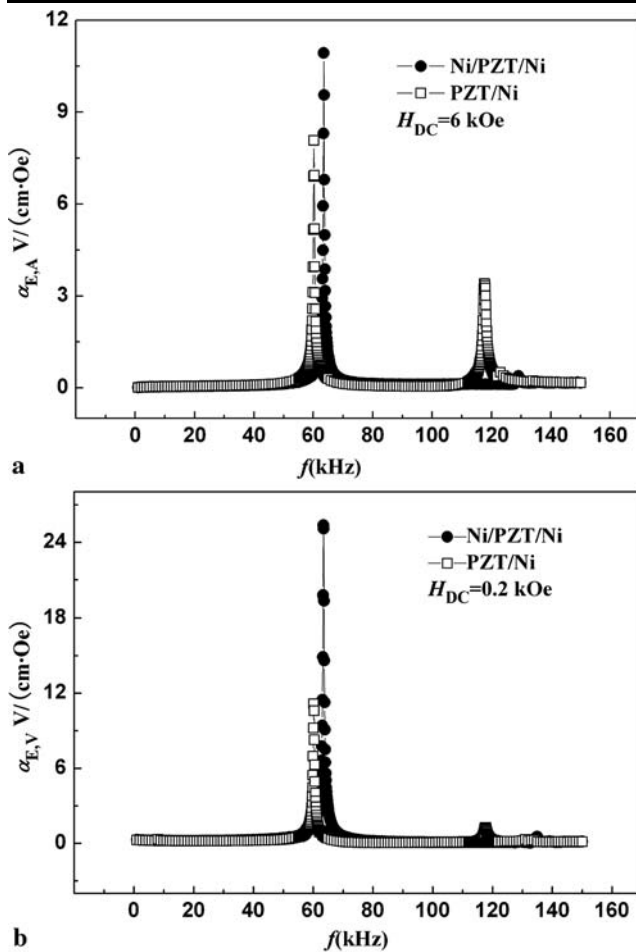
Comparing  $\alpha_{E,A}$  of the two samples at the same applied magnetic field, the trilayered sample has larger magnetoelectric voltage coefficient than the bilayered sample. At 8 kOe,  $\alpha_{E,A}$  of the trilayered sample is 14.5 V/(cm·Oe) compared to only 11 V/(cm·Oe) for the bilayered sample. The same behavior is observed in the vertical mode ( $\alpha_{E,V}$ ) (Fig. 3(d)). The trilayered sample has the magnetoelectric voltage coefficient larger than the bilayered sample at the same bias magnetic field. However, after 2 kOe, the second ME peak disappears, indicating that under high magnetic field, the effect of normal infinitesimal unit to the magnetic fields is weakened at this resonant frequency [10].

We also measured the dependence of the magnetoelectric voltage coefficients  $\alpha_{E,A}$  and  $\alpha_{E,V}$  on  $H_{DC}$  varied from -1 kOe to 8 kOe with the  $f_{r2}$  applied frequency for the Ni/PZT/Ni and PZT/Ni layered cylindrical composites, shown in Fig. 3 (e, f). As for the axial mode, the bilayered sample ME effect is larger than that of the trilayered sample (Fig. 3(e)). The maximum of  $\alpha_{E,A}$  is about 10 V/(cm·Oe) for the bilayered sample, but it is less than 1 V/(cm·Oe) for the trilayered sample. After 1 kOe,  $\alpha_{E,A}$  linearly increases with the bias magnetic field, which indicates that the ME effect

caused by the volume magnetostriction under high magnetic field is weaker than the one caused by the line magnetostriction under low magnetic field. This means that one cannot choose this resonant frequency as the working frequency for the high magnetic field detection devices. Figure 3(f) shows  $H_{DC}$  curve of  $\alpha_{E,V}$  in the vertical mode. Here,  $\alpha_{E,V}$  is very weak, and the bilayered sample ME effect is larger than for the trilayered sample. This also indicates that one cannot choose this resonant frequency as the working frequency.

From Fig. 3 it is obvious that the magnetoelectric voltage coefficient of the trilayered sample is larger than that of the bilayered one at low and at the first resonant frequencies. However, at the second resonance frequency, the situation is reversed. This indicates that the first and the second resonant peaks correspond to different resonant modes. In a cylinder, there are axial and radial vibration modes, and thus the first and the second resonant vibrations correspond to the axial and the radial modes, respectively [20]. In the axial resonant mode, the mechanical coupling-induced ME effect is along the cylinder axis, and the contact area in the trilayered sample is twice larger compared with the bilayered sample. In this mode, the strain transfer in the trilayered sample is more effective than in the bilayered sample, and thus the ME effect induced in the trilayered sample is larger than in the bilayered sample. However, in the radial resonant mode, the main contribution to the ME effect is along the radial cylinder direction. The Ni cylinder will shrink or expand in the magnetic field, depending in which direction the field is applied. In order to explain the state of stress in the radial mode, we assume that the Ni layer shrinks in the magnetic field and neglect the tangent force in the vertical mode. For the bilayered sample, because of the outer Ni layer shrinking, the PZT hollow cylinder will suffer compressive stress along the radial direction. However, for the trilayered sample, the stress state is more complex than that of the bilayered sample. The middle PZT layer will be under compressive stress in the radial direction induced by the outer Ni layer shrinking and under tensile stress induced by the inner Ni layer shrinking. The two stresses of the opposite sign act along the radial direction and as a result, the overall radial stress in the PZT is reduced. This is why the magnetoelectric voltage coefficient of bilayered sample is larger than that of





**Fig. 5** The dependence of the magnetoelectric voltage coefficients  $\alpha_{E,A}$  (a) and  $\alpha_{E,V}$  (b) on the applied magnetic field frequency for Ni/PZT/Ni and PZT/Ni layered cylindrical ME composites when the applied bias magnetic field  $H_{DC}$  is 6 kOe and 0.2 kOe, respectively

the trilayered sample as reported above at the second resonant frequency (radial mode).

Comparing Figs. 3(c) and 3(e), the maximum ME voltage coefficient is obtained at high magnetic field of 8 kOe at the first resonant frequency, but at low magnetic field of 0.5 kOe at the second resonant frequency for the bilayered sample. The ME maximum at the first and the second resonant frequencies is almost the same (about 11 V/(cm·Oe)). This indicates that the tangent force is the main cause of stress in the cylinder. That is to say, in the ME effect, the piezoelectric modes of  $d_{31}$  in PZT play the main role in the ME effect. Cylindrical PZT ceramic was polarized along the radial direction; the corresponding piezoelectric constants are denoted in Fig. 1(b). The axial and tangential shear stresses are contributing to deformation along  $d_{31}$ , and the normal compressive/tensile stresses are contributing to deformations along  $d_{33}$ . Moreover,  $d_{33}$  dielectric constant is 3–4 times larger than  $d_{31}$ .

Figure 5 shows the dependence of the ME voltage coefficients  $\alpha_{E,A}$  (a) and  $\alpha_{E,V}$  (b) on the applied magnetic field frequency for Ni/PZT/Ni and PZT/Ni layered cylindrical ME composites when  $H_{DC}$  is 0.2 kOe and 6 kOe. The trilayered sample has the largest ME voltage coefficient at the first resonant frequency for both 0.2 kOe and 6 kOe  $H_{DC}$ . This indicates that one should choose the trilayered structure and the first resonant frequency for the ME device structures as the working frequency, although the ME peak of the bilayered sample is larger than that of the trilayered sample at the second resonant frequency.

## 4 Conclusions

In summary, the first and the second resonant frequencies in the 1–150 kHz range in cylindrical ME composites corresponding to the axial and the radial modes were analyzed. The magnetoelectric voltage coefficient of the trilayered structure is larger than that of the bilayered structure at the first resonant frequency, but this situation is reversed at the second resonant frequency. In order to obtain high performance ME devices, one should choose the trilayered structure and the first resonant frequency as the working frequency.

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