



Enhanced magnetoelectric properties of Terfenol-D disk/Pb(Zr,Ti)O₃ ring multiferroic heterostructures with Pb(Zr,Ti)O₃ piezoelectric ring poled radially

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ABSTRACT

Terfenol-D disk/Pb(Zr,Ti)O₃ (PZT) ring multiferroic heterostructures were prepared and the magnetoelectric properties were characterized, for which PZT piezoelectric ring was poled radially. When the outer/inner diameter ratio of PZT ring was 1.6 and the bias and alternative magnetic fields were perpendicular to the thickness direction, the highest magnetoelectric voltage coefficient (dV/dH) at 1 kHz of 165 mV Oe⁻¹ was obtained, which was much higher than that for the previously reported disk-ring heterostructures with the piezoelectric phase poled along the thickness direction (75 mV Oe⁻¹). The enhanced magnetoelectric properties of the present heterostructures were due to the larger component of the piezoelectric coefficient and the increased effective length of the piezoelectric phase that contributed to the magnetoelectric coupling.

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1. Introduction

Magnetoelectric materials have drawn continuously increasing interest in the recent years due to their significant potential applications in actuator, sensors, and transducers [1,2]. Among the magnetoelectric materials, multiferroic heterostructures composed of magnetostrictive and piezoelectric phases have attracted special attentions because of their strong magnetoelectric effect at room temperature [2–13]. When a magnetic field is applied to a multiferroic heterostructure, stresses are generated in the magnetostrictive phase through reverse piezomagnetic effect, then they are passed to the piezoelectric phase and lead to the polarization change in the piezoelectric phase through piezoelectric effect [2]. So the magnetoelectric effect in multiferroic heterostructures is a product property derived from the mechanical coupling between the reverse piezomagnetic effect and piezoelectric effect, and the interfacial mechanical coupling plays a key role for the magnetoelectric effect.

Till now, most of the multiferroic heterostructures are of layered structure [2–5,7–13], and the magnetoelectric effect is derived from coupling the reverse piezomagnetic effect and piezoelectric effect through shear stresses. So it is an interesting and important issue to investigate the multiferroic heterostructures coupled through normal stresses. In the previous work, we have observed that the normal stresses-coupling CoFe₂O₄/Pb(Zr,Ti)O₃ (PZT) and Terfenol-D/PZT disk-ring heterostructures exhibit strong

magnetoelectric effect [14,15], and the theoretical prediction has been carried out on such heterostructures [14,16]. Similar multiferroic heterostructures with cylindrical and semiring configurations have also been reported [17–19]. For the disk-ring heterostructures in our previous work, the piezoelectric PZT phase is poled along thickness direction [14,15], which is perpendicular to the vibration plane since the thickness is much less than the outer and inner diameters of the ring. It has been reported that the magnetoelectric properties of the layered multiferroic heterostructures can be significantly enhanced when the piezoelectric phase is poled along the vibration direction [3,4]. Stronger magnetoelectric effect is also expected if the piezoelectric ring is poled radially for the magnetostrictive disk/piezoelectric ring multiferroic heterostructures.

2. Experimental

In the present work, magnetoelectric disk/piezoelectric ring multiferroic heterostructures with the piezoelectric ring poled radially are investigated, for which Terfenol-D and PZT-8 are adopted as the magnetostrictive and piezoelectric phases, respectively. The configuration of the present heterostructure is shown in Fig. 1. PZT ring with the inner diameter (D_{inner}) of 10 mm, outer diameter (D_{outer}) of 12, 14, 16, 18 or 20 mm and thickness of 1 mm is prepared by the standard solid state reaction method. The PZT ring is pasted with silver electrodes on the inner and outer side faces and poled radially under an electric field of 30 kV/cm in silicon oil at 120 °C. Then Terfenol-D disk with the diameter of a little smaller than 10 mm and thickness of 0.95 mm is inserted into PZT ring, and the small air gap between Terfenol-D disk and PZT ring is filled with silver epoxy. Thus, Terfenol-D disk/PZT ring heterostructures with $D_{\text{outer}}/D_{\text{inner}} = 1.2, 1.4, 1.6, 1.8$ and 2 are prepared. For magnetoelectric property measurement, the bias and alternative magnetic fields are applied along the same direction (x_3 or x_1 in Fig. 1), and the induced alternative voltage between the two electrodes on PZT inner and outer side faces is recorded. Magnetoelectric coefficient (dE/dH) is the most common parameter for describing the magnetoelectric properties, while magnetoelectric voltage coefficient (dV/dH) is more important for practical applications, so both the two parameters are

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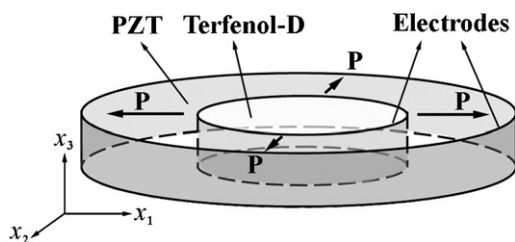


Fig. 1. Configuration of Terfenol-D disk/PZT ring multiferroic heterostructure.

investigated in the present work. The magnetoelectric voltage coefficient can be calculated by the induced alternative voltage divided by the applied alternative magnetic field. When the magnetic fields are along x_3 and x_1 directions, the corresponding magnetoelectric voltage coefficients are denoted by dV_r/dH_3 and dV_r/dH_1 , respectively. The magnetoelectric coefficient is also attained as the magnetoelectric voltage coefficient divided by the effective length of the piezoelectric phase $((D_{\text{outer}} - D_{\text{inner}})/2)$. The measurement is conducted in the frequency range between 1 and 200 kHz. For simplification, the present Terfenol-D disk/PZT ring heterostructure with the PZT piezoelectric ring poled radially is denoted by T/P(r), and the Terfenol-D disk/PZT ring and PZT disk/Terfenol-D ring heterostructures with the PZT phase poled along thickness direction in the previous work [15] are denoted by T/P(t) and P(t)/T, respectively.

3. Results and discussion

Fig. 2 shows the magnetoelectric voltage coefficients (dV_r/dH_3 and dV_r/dH_1) for the present T/P(r) heterostructure with $D_{\text{outer}}/D_{\text{inner}} = 1.6$ at 1 kHz as functions of bias magnetic field (H_{dc}). With increasing the bias magnetic field, the magnetoelectric voltage coefficient increases to a peak value, then decreases for both dV_r/dH_3 and dV_r/dH_1 . This can be explained by the bias magnetic field-dependent piezomagnetic coefficient of Terfenol-D, which first increases and then decreases with increasing the bias magnetic field [5]. The bias magnetic field where dV_r/dH_3 reaches the peak value is much higher than that for dV_r/dH_1 , while the peak value for the former is significantly lower than that for the latter. The different dependences for dV_r/dH_3 and dV_r/dH_1 on bias magnetic field can be explained by the different components of piezomagnetic coefficient of Terfenol-D that contribute to the magnetoelectric coupling for the two cases. Since the outer and inner diameters are much larger than the thickness for the present T/P(r) heterostructure, the vibration occurs in $x_1 - x_2$ plane for both dV_r/dH_3 and dV_r/dH_1 . That is to say, the applied magnetic field and the vibration are perpendicular for dV_r/dH_3 , while they are both in $x_1 - x_2$ plane for dV_r/dH_1 . So only q_{31} component of the piezomagnetic coefficient of Terfenol-D contributes to the magnetoelectric coupling for dV_r/dH_3 , while q_{33} component dominates for dV_r/dH_1 . It is well known that q_{33} reaches

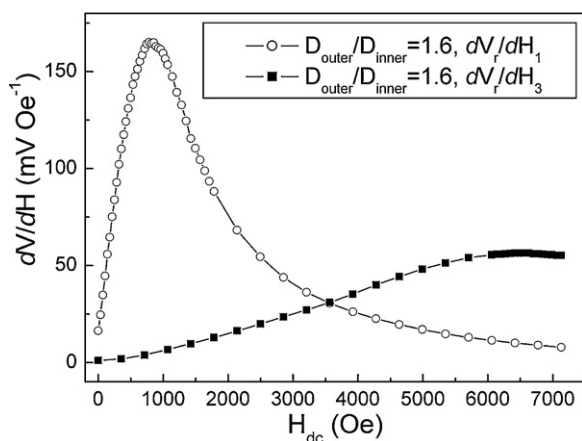


Fig. 2. Magnetoelectric voltage coefficients of T/P(r) heterostructure with $D_{\text{outer}}/D_{\text{inner}} = 1.6$ at 1 kHz as functions of bias magnetic field.

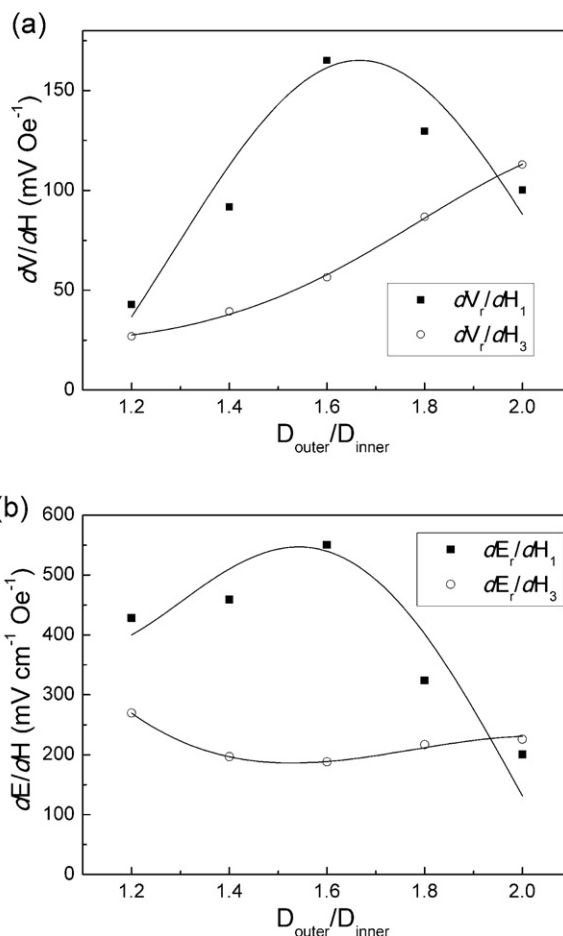


Fig. 3. Peak (a) magnetoelectric voltage coefficients and (b) magnetoelectric coefficients of T/P(r) heterostructures at 1 kHz as functions of $D_{\text{outer}}/D_{\text{inner}}$.

a higher peak value at a much lower bias magnetic field comparing with q_{31} [3,5], and this leads to the different dependences for dV_r/dH_3 and dV_r/dH_1 on the bias magnetic field.

The peak magnetoelectric voltage coefficients of the T/P(r) heterostructures show strong dependence on the ratio of the outer/inner diameters ($D_{\text{outer}}/D_{\text{inner}}$) of the PZT ring, as shown in Fig. 3(a). dV_r/dH_3 increases with increasing $D_{\text{outer}}/D_{\text{inner}}$, while dV_r/dH_1 first increases and reaches the maximum value when $D_{\text{outer}}/D_{\text{inner}} = 1.6$, and then decreases. For the magnetoelectric coefficient, the dependence of dE_r/dH_1 on $D_{\text{outer}}/D_{\text{inner}}$ is similar to dV_r/dH_1 , while dE_r/dH_3 first decreases and then increases with increasing $D_{\text{outer}}/D_{\text{inner}}$, as shown in Fig. 3(b). The different dependences of magnetoelectric voltage coefficient and magnetoelectric coefficient on $D_{\text{outer}}/D_{\text{inner}}$ are due to the $D_{\text{outer}}/D_{\text{inner}}$ -dependent effective length of the piezoelectric phase $((D_{\text{outer}} - D_{\text{inner}})/2)$.

The magnetoelectric properties of the present T/P(r) heterostructures are significantly enhanced comparing with the previous T/P(t) and P/T(t) heterostructures, for which the piezoelectric phase is poled along thickness direction. The highest magnetoelectric coefficient of the present T/P(r) heterostructures is $550 \text{ mV cm}^{-1} \text{ Oe}^{-1}$ for dE_r/dH_1 and $D_{\text{outer}}/D_{\text{inner}} = 1.6$, and it is significantly higher than that for T/P(t) ($400 \text{ mV cm}^{-1} \text{ Oe}^{-1}$) [15]. T/P(r) and T/P(t) heterostructures are of similar configurations, and the main difference between them is the poling direction of the PZT piezoelectric ring. PZT ring is poled along the radial direction for T/P(r), which is in the $x_1 - x_2$ vibration plane, so d_{33} component of the piezoelectric coefficient of PZT dominates for the magnetoelectric coupling. While, the thickness direction along which PZT ring is poled for T/P(t) heterostructures is perpendicular to the

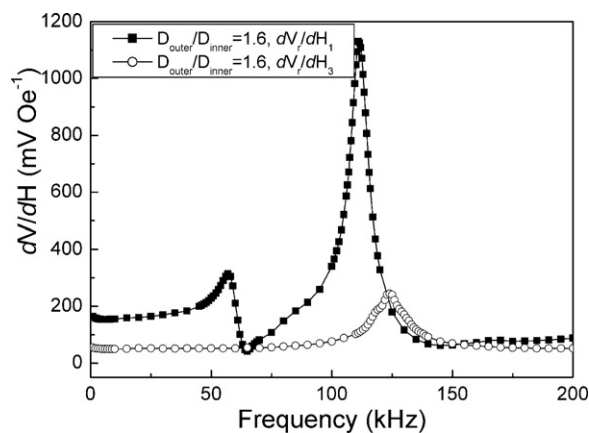


Fig. 4. Magnetolectric voltage coefficients of T/P(r) heterostructure with $D_{\text{outer}}/D_{\text{inner}} = 1.6$ as functions of frequency.

$x_1 - x_2$ vibration plane, and d_{31} component instead of d_{33} contributes to the magnetolectric coupling. The larger value of d_{33} than d_{31} [3] is responsible for the enhanced magnetolectric coefficients of the T/P(r) heterostructures comparing with the T/P(t) heterostructures. The enhancement of the magnetolectric voltage coefficient for T/P(t) heterostructures is more significant comparing with the magnetolectric coefficient. The corresponding highest magnetolectric voltage coefficient for T/P(r) heterostructures is 165 mV Oe^{-1} , while the highest value for T/P(t) heterostructures is only 40 mV Oe^{-1} [15]. Furthermore, the highest magnetolectric voltage coefficient for T/P(r) heterostructures is 2.2 times of that for the P/T(t) heterostructures (75 mV Oe^{-1}), although the highest magnetolectric coefficient for the former ($550 \text{ mV cm}^{-1} \text{ Oe}^{-1}$) is lower than that for the latter ($750 \text{ mV cm}^{-1} \text{ Oe}^{-1}$) [15]. The more significantly enhanced magnetolectric voltage coefficient for T/P(r) heterostructures is not only due to the larger value of d_{33} than d_{31} , but also because of the increased effective length of PZT phase that contributes to the magnetolectric coupling. The effective length is just the thickness of PZT phase (1 mm) for T/P(t) and P(t)/T heterostructures, while it is $(D_{\text{outer}}/D_{\text{inner}})/2$ and varies from 1 to 5 mm for T/P(r) heterostructures since PZT ring is poled along radial direction.

The magnetolectric effect for T/P(r) heterostructures is frequency-dependent, and Fig. 4 shows dV_r/dH_3 and dV_r/dH_1 as functions of frequency for T/P(r) heterostructure with $D_{\text{outer}}/D_{\text{inner}} = 1.6$ under the bias magnetic fields where the magnetolectric voltage coefficients at 1 kHz reach the peak values. Resonant peaks that correspond to electromechanical resonance can be observed for both dV_r/dH_3 and dV_r/dH_1 . When the bias and alternative magnetic fields are along the x_3 direction, the magnetostrictive and piezoelectric phases vibrate radially, so one symmetrical resonant peak that corresponds to radial vibration mode is observed for dV_r/dH_3 . The corresponding resonant peak can also be observed for dV_r/dH_1 , for which the resonant frequency is a little lower than that for dV_r/dH_3 , and the peak value is much

higher. While, another weaker resonant peak with asymmetrical shape can also be observed at a lower frequency for dV_r/dH_1 . The T/P(r) heterostructures are non-centrosymmetrical along x_1 axis, so when the magnetic fields are along x_1 axis, the vibration should be regarded as the combination of the radial vibration and the vibration along x_1 direction [7,15], and the latter should be responsible for the additional asymmetrical resonant peak.

4. Conclusions

Terfenol-D disk/PZT ring multiferroic heterostructures with PZT piezoelectric ring poled radially have been prepared and characterized. The present T/P(r) heterostructures exhibit significantly enhanced magnetolectric properties, and the highest magnetolectric voltage coefficient at 1 kHz is as high as 165 mV Oe^{-1} , which is 2.2 times of that for the Terfenol-D/PZT disk-ring heterostructures with the piezoelectric phase poled along thickness direction. The larger component of piezoelectric coefficient and increased effective length of the PZT piezoelectric phase that contribute to the magnetolectric coupling are responsible for the enhanced magnetolectric properties for the present heterostructures.

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