

# Giant magnetoelectric effect of a hybrid of magnetostrictive and piezoelectric composites

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A magnetoelectric (ME) hybrid structure is constructed by an efficient coupling between magnetostrictive Terfenol-D ( $\text{Tb}_{0.27-0.30}\text{Dy}_{0.73-0.70}\text{Fe}_{1.90-1.95}$ )/epoxy and piezoelectric lead–zirconate–titanate [ $\text{Pb}(\text{Zr}_{0.52}\text{Ti}_{0.48})\text{O}_3$ ]/epoxy composites. Significant ME effect produced by the piezoelectric  $k_{31}$  coupling with the longitudinal vibration of the magnetostrictive component over a wide range of frequency is observed. It is revealed that the ME effect can be enhanced by applying an optimized magnetic bias. A magnetoelectric voltage coefficient as high as 8700 mV/cm Oe is recorded at the resonance frequency of 59.2 kHz for the structure with an optimized magnetic bias of 0.7 kOe. Our measurement confirms that the eddy current loss remains negligibly small at an operating frequency as high as  $\sim 200$  kHz, predicting very promising applications of the present ME structure. © 2003 American Institute of Physics. [DOI: 10.1063/1.1577404]

## I. INTRODUCTION

Magnetoelectric (ME) effect is defined as a variation of dielectric polarization in a system as a response to an applied magnetic field, or an induced magnetization by an external electric field.<sup>1</sup> With this effect, an effective conversion between electric energy and magnetic energy becomes possible. The ME materials thus represent one kind of smart structures which have received continuous attentions as potential sensors for magnetic field measurements and transducers for magnetoelectric conversion.<sup>2,3</sup> Since the ME effect was observed in antiferromagnetic  $\text{Cr}_2\text{O}_3$  compound,<sup>4</sup> quite a lot of ME materials and structures have been discovered and developed. Unfortunately, for almost all ME compounds ever discovered, the ME effect, characterized by the magnetoelectric voltage coefficient, e.g.,  $\alpha_E = (\partial E / \partial H)_T$  with  $E$  the probed electric field and  $H$  the applied magnetic field, and  $T$  the temperature, is too small ( $\alpha_E \sim 20$  mV/cm Oe) to be utilized for practical purposes.<sup>4–6</sup>

However, a much higher ME effect has been identified in specially designed composites in which the magnetostrictive phase is combined with the piezoelectric one so that an efficient magnetomechanical–piezoelectric coupling between the two phases is achieved.<sup>7</sup> Since then, much effort has been made along this line. For example, Van den Boomgaard

*et al.*<sup>8,9</sup> synthesized a bulk composite of CFO (cobalt ferrite) with  $\text{BaTiO}_3$ , but the ME yield was  $\sim 50$  times lower than that predicted theoretically. The CFO–lead–zirconate–titanate (PZT) multilayered structures prepared by Harshe *et al.*<sup>10</sup> showed an  $\alpha_E \sim 75$  mV/cm Oe, which was still much lower than the predicted value. Recently, strong ME effect was demonstrated in a nickel ferrite (NFO)/PZT multilayered structure prepared by Srinivasan *et al.* using the tape casting technique.<sup>11,12</sup> The reported  $\alpha_E$  value was  $\sim 1500$  mV/cm Oe, which was in excellent agreement with the theoretical prediction.

These earlier studies allow us to emphasize the essential role of two issues in fabricating magnetoelectric composites with enhanced ME effect, on one hand, the two phases chosen must have, respectively, large magnetostrictive and piezoelectric effects. On the other hand, an effective coupling between the magnetomechanical transfer and piezoelectric transfer is the key factor for an enhanced ME effect. In 2001, Ryu *et al.* developed a sandwich structure stacked by giant magnetostrictive material Terfenol-D ( $\text{Tb}_{0.27-0.30}\text{Dy}_{0.73-0.70}\text{Fe}_{1.90-1.95}$ ) and PZT disks. The measured largest value of  $\alpha_E$  reached 5900 mV/cm Oe.<sup>13,14</sup> However, the wider application of this structure is seriously limited by the presence of high eddy current loss in Terfenol-D above a few keloherz.<sup>15</sup> To overcome this difficulty, Terfenol-D/polymer composite is an excellent alternative because of its lower eddy current loss in the region of high frequency.<sup>16</sup>

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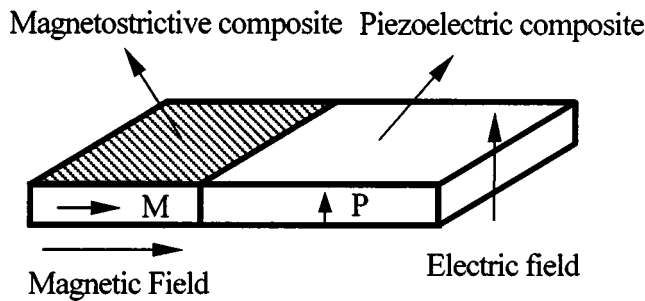


FIG. 1. Schematic of the magneto-electric composite structure. The arrows  $P$  and  $M$  show the poling direction of piezoelectric composite and magnetic domain direction of magnetostrictive composite, respectively.

Moreover, its tailorable properties enable it to meet the special ME coupling.<sup>16–18</sup>

In this article, we report a ME composite structure based on 0-3 Terfenol-D/Epoxy composite [magnetostrictive component (MSCP)] combined with 0-3 Pb ( $Zr_{0.52}Ti_{0.48}$ )O<sub>3</sub>/epoxy composite [piezoelectric component (PECP)]. The structure operates on the longitudinal vibrational mode and a very strong ME yield at the resonance frequency is observed. The PZT/epoxy composite is chosen as the PECP for two reasons. One is that an effective interface matching between MSCP and PECP can be easily achieved by introducing the PZT/epoxy composite. The other is that an efficient ME coupling between the two components is allowed, noting that the acoustic impedance of the two components can be adjusted to optimization by changing the contents of the polymer. Figure 1 is a schematic illustration of the ME composite structure. The arrows  $P$  and  $M$  show the poling direction of the PECP and magnetic domain orientation of the MSCP, respectively. Therefore, the MSCP is activated in the longitudinal vibrational mode. When an external ac magnetic field is applied, the mechanical deformation in the MSCP is acoustically transferred to the PECP, thereby resulting in an induced electric field due to the transverse piezoelectric coupling  $k_{31}$ .

## II. EXPERIMENT

The ME composite structure was prepared by using a compression technique. Terfenol-D powder (from Baotou Rare-earth Institute, China) with an average size of 100  $\mu\text{m}$  and PZT-502 ceramic powder (from PKI, USA) with an average size of 60  $\mu\text{m}$  were separately mixed with epoxy resin (Spurr low-viscosity embedding media, Polyscience Inc.) and precompressed in the rectangular Plexiglass molds to form the Terfenol-D/Epoxy and PZT/epoxy composites with dimensions of 20 mm $\times$ 20 mm $\times$ 10 mm (thickness). The volume fractions of Terfenol-D and PZT in the composites are 0.7 and 0.6, respectively. Subsequently, the two precompressed composites were placed into a compaction die with cavity dimensions of 20 mm $\times$ 20 mm $\times$ 25 mm (depth) in turn and laminated. The laminated composite was then compacted under a pressure of 50 MPa. In order to align the Terfenol-D powders, compaction was carried out under a static magnetic field of 1.5 kOe, the direction of which is parallel to that of the pressure. After that, the laminated com-

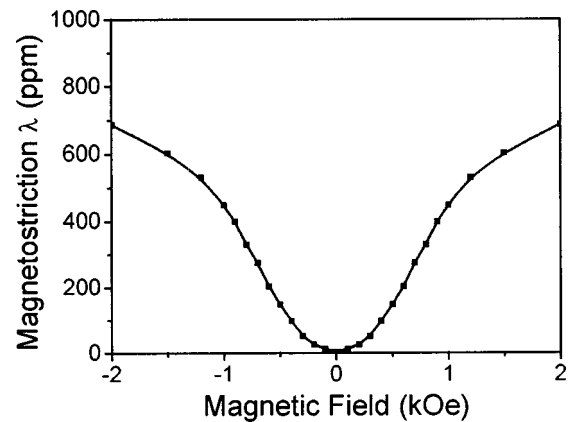


FIG. 2. Magnetostriction response of the Terfenol-D composite as function of applied magnetic field.

posite was cured at 70 °C for 8 h and then sliced into the samples, as shown in Fig. 1. The dimensions of the Terfenol-D composite and PZT composite in the sample were 6.8 mm (length) $\times$ 6.2 mm $\times$ 0.7 mm and 9.1 mm (length) $\times$ 6.2 mm $\times$ 0.7 mm, respectively. Finally, the PZT/epoxy component was covered by air-dried silver electrodes and poled in silicone oil at 90 °C and 35 kV/cm for 25 min.

The piezoelectric coefficient of the PZT composite as measured by a piezo  $d_{33}$  meter (Model ZJ-3B) is  $d_{33}=50$  pm/V. The magnetostriction coefficient  $\lambda$  for the Terfenol-D composite was measured by using a MTI-2000 optical fiber displacement sensor. The ME coefficient  $\alpha_E=(\partial E/\partial H)_T$  is determined by the induced electric field generated in the PECP under a small ac magnetic field less than 5 Oe (generated by a solenoid) superposed onto a dc magnetic bias  $H_{\text{Bias}}$  of up to 2.1 kOe (generated by a pair of permanent NdFeB magnets). Both the ac magnetic field and magnetic bias are parallel to the longitudinal direction of the MSCP. The frequency  $f$  of the ac magnetic field varies in the range of 0–200 kHz. The solenoid was driven by a signal generator and power amplifier. The induced electric field was measured with a high input impedance circuit and oscilloscope. The ac magnetic field and flux density were, respectively, measured by using a pick-up coil and a search coil connecting with a fluxmeter.

## III. RESULTS AND DISCUSSION

Figure 2 shows magnetostriction response of the Terfenol-D composite as function of applied magnetic field. One can observe the changing slope and saturation strain in the butterfly-shaped curve of the double-sided strain versus applied magnetic field. The typical magnetostriction coefficient  $\lambda$  for the Terfenol-D composite at 1.0 kOe is  $\sim 450$  ppm. Figure 3 presents the flux density of the MSCP in the sample versus magnetic field frequency  $f$  at various  $H_{\text{Bias}}$ . The measured response is quite similar to the relative permeability  $\mu_r$  response reported earlier.<sup>16</sup> Given a magnetic bias  $H_{\text{Bias}}$ , the fundamental longitudinal resonance appears at  $f \sim 60$  kHz and the peak at  $\sim 120$  kHz represents the second harmonic resonance.

Figure 4 shows the dependence of  $\alpha_E$  on  $f$ , i.e., disper-

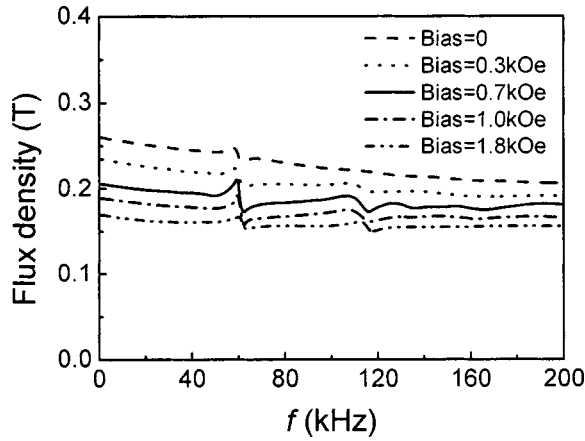


FIG. 3. Flux density of MSCP in the sample as a function of applied magnetic field frequency  $f$ .

sion of the ME effect, under different  $H_{\text{Bias}}$ . In all cases, no remarkable frequency dispersion is observed except for the resonance ranges, indicating an excellent frequency stability of the ME effect. At  $H_{\text{Bias}}=0$ , two peaks located at  $f=59.8$  and  $29.9$  kHz, respectively, are observed. The corresponding peak values for  $\alpha_E$  are 400 and 180 mV/cm Oe, which are comparable to the values reported.<sup>8–12</sup> The peak at  $f=59.8$  kHz is obviously attributed to the fundamental longitudinal vibration of the sample, as confirmed by the measured response of the flux density in Fig. 3, whereas the weaker peak at  $f=29.9$  kHz is caused by the double-sided strain response of the Terfenol-D composite versus applied magnetic field. In more detail, the magnetostriction of the Terfenol-D composite is symmetric with magnetization and the double-sided strain versus applied magnetic field or “butterfly” results, as shown in Fig. 2. Hence the induced strain of the MSCP shows a doubling of the applied magnetic field frequency  $f$ , which leads to the piezoelectric vibration of the PECP at  $2f$  by the magnetomechanical–electric coupling. When the applied magnetic field frequency  $f$  is 29.9 kHz, the sample vibrates at a frequency of  $2f=59.8$  kHz, which agrees exactly with the fundamental longitudinal resonance of the sample, thus a resonance peak of  $\alpha_E$  appears. Once a nonzero magnetic bias  $H_{\text{Bias}}$  is applied, this so-called frequency doubling of the Terfenol-D composite will be gradu-

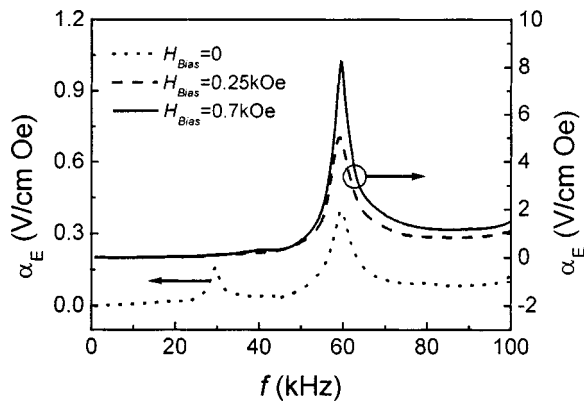


FIG. 4. Frequency dependence of magnetolectric voltage coefficient  $\alpha_E$  with different magnetic bias  $H_{\text{Bias}}$ .

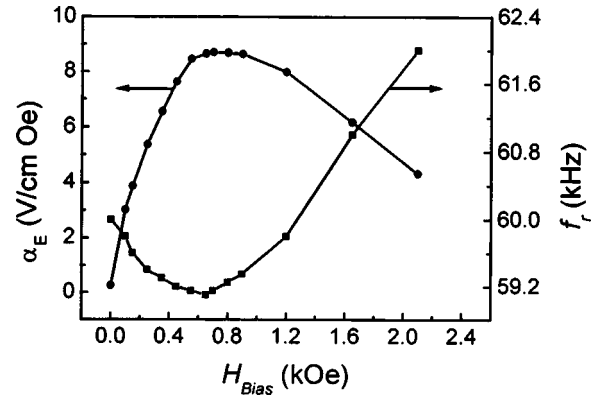


FIG. 5. Magnetolectric voltage coefficient  $\alpha_E$  measured at the resonance and corresponding resonance frequency  $f_r$  as functions of magnetic bias  $H_{\text{Bias}}$ .

ally suppressed,<sup>19</sup> and the consequent ME effect will become weaker. In fact, the peak of  $\alpha_E$  at  $f=29.9$  kHz almost disappears when the applied magnetic bias is above 0.2 kOe in this work.

Since the magnetostriction slope of the Terfenol-D composite around the zero magnetic bias point is so shallow, vibration of the MSCP under an ac magnetic field is very inefficient, which causes the ME effect to be very weak in this region. By applying a magnetic bias  $H_{\text{Bias}}$ , however, ac operation of MSCP can be moved to steeper regions of magnetostriction curve, eventually getting into the middle of the burst region, where much larger strains are realized for a given ac magnetic field, hence much stronger ME effects are achieved. As shown in Fig. 4, with  $H_{\text{Bias}}$  increasing to 0.25 and 0.7 kOe, the peak intensities of  $\alpha_E$  at fundamental longitudinal resonance are enhanced by 1 order of magnitude, reaching up to  $\sim 5100$  and  $\sim 8700$  mV/cm Oe, respectively. The detailed  $H_{\text{Bias}}$  dependence of  $\alpha_E$  is given in Fig. 5. With increasing  $H_{\text{Bias}}$  from zero,  $\alpha_E$  increase rapidly before reaching a maximum at  $H_{\text{Bias}} \sim 0.7$  kOe, and then drops slowly. At the same time, the resonance frequency shifts as a function of  $H_{\text{Bias}}$ , decreases with increasing  $H_{\text{Bias}}$  before reaching a minimum at  $H_{\text{Bias}} \sim 0.7$  kOe, and then rises slowly. To understand this magnetic bias dependence further, one notes the negative  $\Delta E$  effect for the MSCP,<sup>18,20</sup> where  $\Delta E = (E_{H_{\text{Bias}}} - E_0)/E_0$  with  $E_{H_{\text{Bias}}}$  and  $E_0$  the Young's modulus at a given magnetic bias  $H_{\text{Bias}}$  and zero bias, respectively. At the small ac magnetic field, with increasing  $H_{\text{Bias}}$  from zero, the mobility of the non- $180^\circ$  domain wall in the MSCP is enhanced and magnetically favored domains grow, which leads to the negative  $\Delta E$  effect (the elastic moduli at constant magnetic field strength  $E_H$  decreases). At the so-called optimized magnetic bias (here it is  $H_{\text{Bias}} = 0.7$  kOe), the motion of the non- $180^\circ$  domain is maximum and  $E_H$  reaches minimum. The MSCP is softened enough that the magnetostrictive coupling factor  $k_{33}$  is the highest, leading to the largest strain response. Assuming that the coupling between the MSCP and PECP has nothing to do with the magnetic bias, which seems reasonable, the observed  $\alpha_E$  has a maximum at  $H_{\text{Bias}} = 0.7$  kOe. In fact,  $k_{33}$  can be calculated by<sup>17</sup>

$$k_{33} = [1 - (f_r^E/f_d^E)^2]^{1/2}, \quad (1)$$

where  $f_r^E$  and  $f_a^E$  are the resonance frequency and antiresonance frequency, respectively. By taking these characteristic frequency from Fig. 3, the calculated maximum  $k_{33}$  at  $H_{\text{Bias}} = 0.7$  kOe is 0.311, corresponding to a maximum  $\alpha_E = 8700$  mV/cm Oe at 59.2 kHz, which is much higher than the values of previously reported magnetoelectric materials and structures.<sup>4,10,11,13</sup>

Moreover, the problem of high eddy current loss for Terfenol-D over the high frequency range has been overcome in the Terfenol-D/epoxy composite. The measurement demonstrates that the upper frequency limit for this ME composite structure can be extended up to 100 kHz and higher, as shown in Fig. 4. According to the classical eddy current theory,<sup>21</sup> the critical operating frequency  $f_c$  at which serious eddy current loss is expected can be calculated by

$$f_c = \frac{2\rho}{\pi\mu_r\mu_0 t^2}, \quad (2)$$

where  $\rho$  is the electric resistivity and  $t$  is the effective diameter of the MSCP. Taking  $\rho = 390 \mu\Omega\text{m}$ ,  $t = 7$  mm, and  $\mu_r = 1.85$  measured at  $H_{\text{Bias}} = 0.7$  kOe, one has  $f_c \sim 2.2$  MHz, which is  $\sim 1000$  times higher than the critical frequency for bulk Terfenol-D. In fact, apart from the resonance near 60 and 120 kHz, the measured relative permeability  $\mu_r$  as a function of  $f$  is flat enough until  $f \sim 200$  kHz. This verifies that the eddy current loss is negligible over a wide range of frequency and the present ME composite structure can be used for high frequency applications at least up to 200 kHz.

#### IV. CONCLUSION

In conclusion, we have developed a ME composite structure which combined the Terfenol-D/epoxy and PZT/epoxy composites as the MSCP and PECP, respectively. It operates by the longitudinal vibration of the MSCP driving the piezoelectric output of the PECP. Giant magnetoelectric voltage coefficient at the fundamental longitudinal resonance mode of the MSCP with low eddy current loss has been observed, demonstrating a promising application potential

for this ME composite structure. The frequency dependence and magnetic bias dependence of the magnetoelectric coefficient suggest that even higher conversion efficiency can be accomplished if the coupling parameters, such as resonance frequency, coupling coefficient, and acoustic impedance etc. are further optimized.

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