

# A new magnetoelectric resonance mode in bilayer structure composite of PZT layer and Terfenol-D/epoxy layer

Zhan Shi · Jing Ma · Ce-Wen Nan

Published online: 1 September 2007  
© Springer Science + Business Media, LLC 2007

**Abstract** Magnetoelectric (ME) composites with a bilayer structure were prepared by easily combining  $\text{Pb}(\text{Zr},\text{Ti})\text{O}_3$  layer and Terfenol-D/epoxy layer. A new second flexural ME resonance mode was investigated and confirmed by vibration modal analysis. A giant ME coefficient at the first flexural resonance mode (2.79 V/cmOe at 35 kHz) is much larger than that at the transverse resonance mode (1.31 V/cmOe at 122 kHz). A flat ME response of about 360 mV/cmOe occurs in the range from 50 to 80 kHz, which shows a large bandwidth. This bilayer structure is expected to achieve a giant ME effect at relatively low frequency.

**Keywords** Magnetoelectric · Bilayer · Resonance

## 1 Introduction

Magnetoelectric (ME) effect is defined as an induced polarization on applying a magnetic field or an induced magnetization on applying an electric field. Recently ME materials have drawn more and more interest due to its significant potential applications in actuators, sensors, and transducers [1–3]. In a multiferroic composite, ME effect is derived from the mechanical coupling between magnetic phase and piezoelectric phase, and thus ME property is also called as product property [4]. That is, when a magnetic field is applied on the composite, the magnetic phase changes its shape magnetostrictively, and then the strain transfers to the piezoelectric phase by mechanical coupling,

and finally induces a polarization. In ME coefficient measurement, the applied magnetic field is a large static magnetic field superposing a weak AC magnetic field [5].

Theoretical research predicted that a strong ME effect can be produced by combining piezoelectric materials of large piezoelectric effect and magnetic materials of large magnetostrictive effect [6]. Subsequently, the laminate composites of giant magnetostrictive alloy Terfenol-D and  $\text{Pb}(\text{Zr},\text{Ti})\text{O}_3$  (PZT) were investigated and their ME coefficient reached several V/cmOe at 1 kHz [7]. The giant ME (GME) materials are expected to be used in practical applications such as magnetic sensor or transducer [2, 3]. However, the Terfenol-D is so brittle that it is very hard to prepare, and is conducive to make large eddy current loss when the frequency of AC magnetic field is above 1 kHz. To overcome these two shortcomings of bulk Terfenol-D, the bulk Terfenol-D can be replaced with the composite of Terfenol-D particle and polymer [8]. The inert polymer is used as binder which would reduce the magnetostriction in a magnetic composite but increase the resistivity of the composite and thus the operating frequency to about 200 kHz. This makes it possible to study the ME response in a wide frequency range in the composites consisting of Terfenol-D.

Many researchers have found that the ME coefficients are significantly dependent on the frequency of the small AC magnetic field. It is a popular view that the ME coefficients at a resonance mode are significantly larger than that at low frequency. In several ME materials such as ferrite/PZT and Terfenol-D/PZT/PVDF, the GME effect is observed at high frequency resonance modes [8, 9]. There are several resonance modes in piezoelectric materials. Likely, in ME materials the longitude (L) resonance mode and transverse (T) resonance mode have been found in several kinds of structures [8, 10, 11]. However, for the L

Z. Shi · J. Ma · C.-W. Nan (✉)  
State Key Lab of New Ceramics and Fine Processing,  
Department of Materials Science and Engineering,  
Tsinghua University,  
Beijing 100084, China  
e-mail: cwnan@tsinghua.edu.cn

and T modes the resonance frequency is quite high, which will greatly increase the energy loss and make the converse efficient worse. Most recently, a strong flexural resonance mode at relatively low frequency is reported in a bilayer rectangle composite made of Terfenol-D/epoxy (TE) layer and PZT layer. The first flexural resonant ME mode and the ordinary longitudinal resonant ME mode are observed at 12.5 and 71.6 kHz [12]. However, the second and third flexural ME resonance modes have not been observed.

In this paper, we investigate the ME resonance modes in a bilayer disk made of TE layer and PZT layer and report new first and second order flexural ME resonance modes in this bilayer disk with GME coefficients.

### 2 Experimental

The bilayer disk was prepared by bonding the PZT disk with TE disk. First, PZT disk with thickness 0.3 mm was electroded and polarized in silicon oil under a poling field of 2 kV/mm and at poling temperature of 70°C. The piezoelectric constant of PZT is 440 pC/N measured by a standard  $d_{33}$  meter, and its dielectric constant is 953 at 1 kHz measured by HP4194 impedance analyzer. Terfenol-D powder with average particle size of about 50  $\mu\text{m}$  (from Beijing Institute of Non-Ferrous Metals, China) was mixed with low viscosity epoxy to get TE mixture. Then the PZT disk was put into a mould and TE mixture was poured into the mould. The epoxy hardened at room temperature, and then we cut off the redundant epoxy and got the sample (see Fig. 1). The diameter and total thickness of the sample are 13.2 and 2 mm, respectively, and the thickness ratio  $t_{\text{PZT}}/L=0.85$ . The volume fraction of Terfenol-D particles in the TE layer was about 0.23.

To measure the electrical properties, we connected two thin wires to the electrodes of PZT as the output terminates (Fig. 1). ME response was measured under a mechanical free boundary condition in the frequency range from 100 Hz to 158 kHz using a self-construct ME test equipment, which had been introduced in our previous work [8]. The bias magnetic field was applied along the polarized direction of PZT and the ME response was got by measuring the charge output on the PZT layer. Vibration modal analysis was performed by using a laser Doppler vibrometer (Polytec, German).

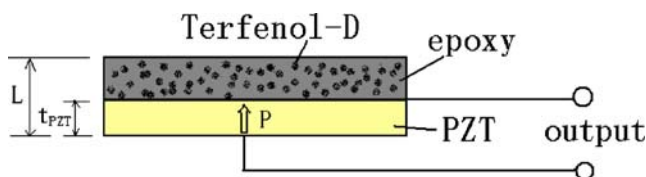


Fig. 1 Schematic illustration of TE/PZT bilayer disk

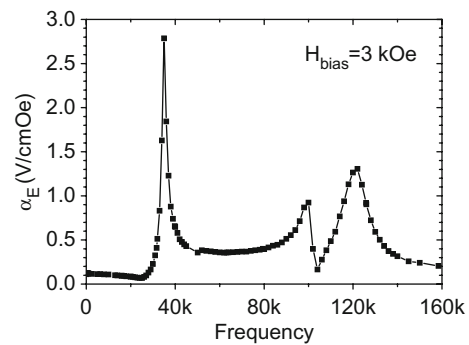


Fig. 2 ME coefficient of the TE/PZT bilayer disk as the function of frequency

### 3 Results and discussion

Figure 2 illustrates the frequency dependence of the ME coefficient  $\alpha_E$  ( $\alpha_E=dE/dH$ ) of this bilayer disk at 3 kOe. Three resonance peaks are obviously observed and the peak values are 2.79 V/cmOe at 35 kHz, 0.924 V/cmOe at 100 kHz, and 1.31 V/cmOe at 122 kHz respectively. The resonance peak at 122 kHz is attributed to the T resonance, which was discussed in many sandwich laminated composites [8, 9]. However, the observation of three ME resonance peaks in laminated composites has not been reported before.

Figure 3 shows the electric impedance spectrum of the sample by using HP4194 impedance analyzer. As shown in Fig. 3, there are also three resonance peaks at about 35, 100, and 122 kHz, respectively. Comparing Fig. 2 with Fig. 3, we can find that the frequencies of the ME resonance peaks are in good agreement with those of the piezoelectric resonance peaks. In the electric impedance test, the sample is driven by an AC electric field, while in the ME coefficient test it is driven by a weak AC magnetic field. These two different driving fields can both provide T strain in the composite. So vibration behavior observed in these two kinds of tests is the same. When the frequency of the applied AC magnetic field matches the piezoelectric resonant frequency of the composite, the magnetostrictive strain induces the piezoelectric resonance of the composite by converse piezoelectric effect and results in the ME

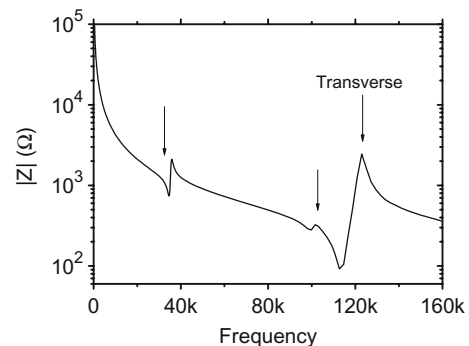
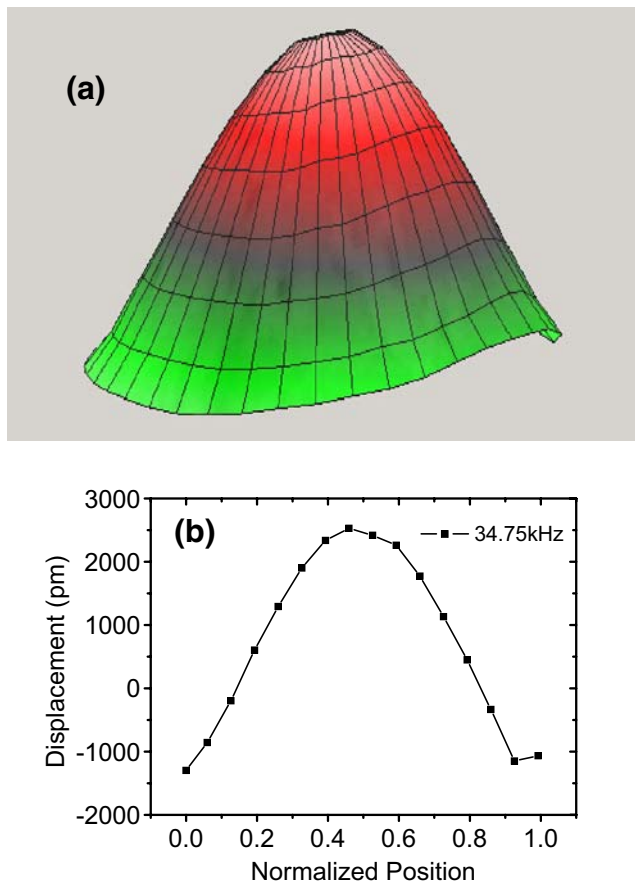


Fig. 3 Electric impedance spectrum of the PZT disk in the bilayer



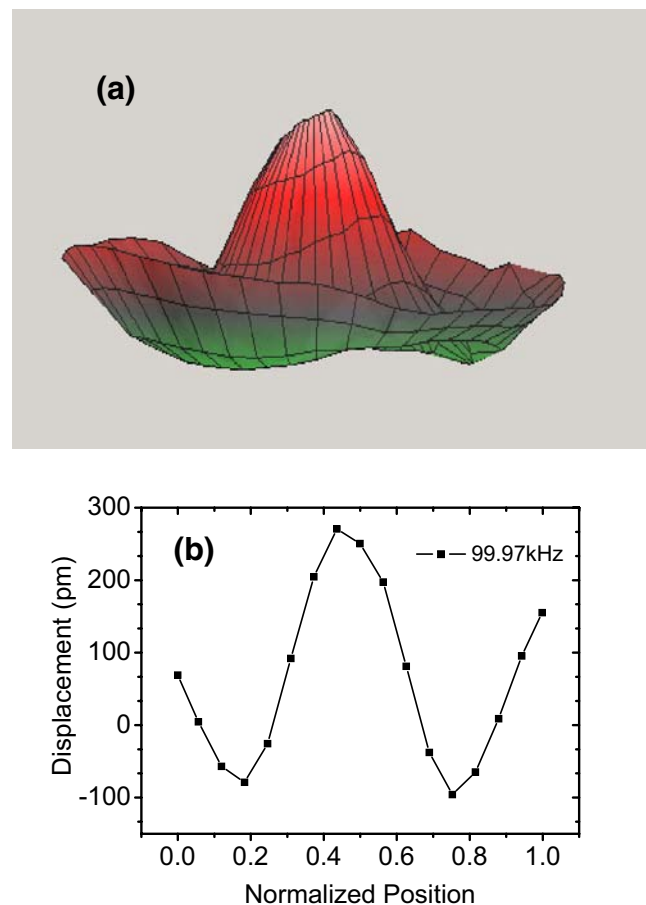
**Fig. 4** (a) Vibration mode shape and (b) the displacement distribution along the radial axle of bilayer disk at 34.75 kHz

resonance. Thus, the ME resonance is attributed to the piezoelectric resonance.

To investigate the origin of the first and second resonant peaks, we further perform the vibration modal analysis by using a laser Doppler vibrometer. The sample is driven by 5 V<sub>pp</sub> sine voltage in a frequency range from 100 Hz to 200 kHz. By measuring the out-of-plane vibration displacement of the top surface of bilayer disk, the first and second order flexural resonant peaks are found at 34.75 and 99.97 kHz. Then a single frequency test is performed at 34.75 kHz as shown in Fig. 4. Figure 4(a) illustrates the mode shape at 34.75 kHz, which is a typical first flexural resonant mode [13]. Figure 4(b) gives the radial distribution of out-of-plane displacement, with 0 and 1 representing the edges and 0.5 representing the center point by normalizing the surface position. Similarly, Fig. 5 shows the results of vibration modal analysis at 99.97 kHz, which confirms a typical second flexural resonant mode [13]. The displacement of the second flexural resonance is about 1/9 of that of the first one. For the bilayer structure piezoelectric transducer, the first and second order flexural resonant modes have been well studied by numerical calculation, finite element analysis approach and experiments [13]. Although the resonant frequencies in the vibration modal

analysis in the bilayer piezoelectric transducer have a little difference from these in ME coefficient spectrum, they are in well agreement with each other.

According to the vibration modal analysis shown in Fig. 4 and Fig. 5, we confirm that the ME resonance peaks at 35 and 100 kHz are attributed to the first and second flexural resonant modes, respectively. According to the displacement values in Fig. 4(b) and Fig. 5(b), the first flexural resonance mode is much stronger than the second one. The larger displacement at first flexural resonance mode results in a stronger ME response as shown in Fig. 2. Furthermore, the first flexural resonance ME coefficient at a much lower frequency is about 2 times larger than that at T mode. This is of benefit to minimization of the ME devices with high ME response. The second flexural ME resonance mode is observed for the first time in laminated disk, at which the ME coefficient is remarkable and the operating frequency is also lower than that at T mode. Between the first and second order resonant peak, especially from 50 to 80 kHz, the ME coefficient is almost constant and with a high value of about 360 mV/cmOe, which has a large bandwidth of about 30 kHz. Although a larger ME coefficient around resonant frequency achieves several V/cmOe, it only takes



**Fig. 5** (a) Vibration mode shape and (b) the displacement distribution along the radial axle of bilayer disk at 99.97 kHz

place in a very narrow frequency range. Moreover the amplitude of the ME coefficient always changes violently around the resonant frequency, resulting in a very small bandwidth. Thus the flat high ME response at a wide frequency range in this bilayer structure is suitable to be used in a magnetic devices.

#### 4 Conclusions

In summary, not only the first flexural ME resonance mode and T ME resonance mode, but also the second flexural ME resonance mode has been observed for the first time. In addition, the ME coefficient at the first ME resonance mode is larger than that at the ME T mode for the laminate disk, and the much lower frequency for the first ME flexural resonance mode is an advantage for the miniaturization of the ME devices. A large ME coefficient of about 30 kHz bandwidth in this bilayer disk has been observed, which is expected to be used in a magnetic devices.

**Acknowledgement** The authors thank Ms. En Zhu Li and Dr. Xiang Cheng Chu's discussions and help in the vibration modal analysis. This work was supported by the NSF of China (Grant No. 50232030)

and the Ministry of Sciences and Technology of China through the 973-Project (Grant No. 2002CB613303).

#### References

1. T. Ueno, J. Qiu, J. Tani, *IEEE Trans. Magn.* **39**(6), 3534 (2003)
2. S.X. Dong, J.F. Li, D. Viehland, *Appl. Phys. Lett.* **83**(11), 2265 (2003)
3. S.X. Dong, J.F. Li, D. Viehland, J. Cheng, L.E. Cross, *Appl. Phys. Lett.* **85**(16), 3534 (2004)
4. C.-W. Nan, *Phys. Rev. B* **50**, 6082 (1994)
5. J.Y. Zhai, N. Cai, Z. Shi, Y.H. Lin, C.-W. Nan, *J. Appl. Phys.* **95**(10), 5685 (2004)
6. C. W. Nan, M. Li, J. H. Huang, *Phys. Rev. B* **63**, 144415 (2001)
7. J. Ryu, S. Priya, A. V. Carazo, K. Uchino, H.E. Kim, *J. Am. Ceram. Soc.* **84**(12), 2905 (2001)
8. N. Cai, J. Zhai, C.-W. Nan, Y. Lin, Z. Shi, *Phys. Rev. B* **68**, 224103 (2003)
9. M.I. Bichurin, D.A. Filippov, V.M. Petrov, V.M. Laetsin, N. Paddubnaya, G. Srinivasan, *Phys. Rev. B* **68**, 132408 (2003)
10. M. Zeng, J.G. Wan, Y. Wang, H. Yu, J.-M. Liu, X.P. Jiang, C.-W. Nan, *J. Appl. Phys.* **95**(12), 8069 (2004)
11. S.X. Dong, J.R. Cheng, J.F. Li, D. Viehland, *Appl. Phys. Lett.* **83**(23), 4812 (2003)
12. J.G. Wan, Z.Y. Li, Y. Wang, M. Zeng, G.H. Wang, J.-M. Liu, *Appl. Phys. Lett.* **86**, 202504 (2005)
13. X.B. Hu, L.T. Li, X.C. Chu, Z.L. Gui, *Mater. Sci. Eng. B Solid-state Mater. Adv. Technol.* **99**, 316 (2003)