Strong flexural resonant magnetoelectric effect in Terfenol-D/epoxy-Pb(Zr,Ti)O_3 bilayer

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We report the bending resonant magnetoelectric effect observed in the bilayer of magnetostrictive $Tb_{0.30}Dy_{0.7}Fe_2$ (Terfenol-D)/epoxy and piezoelectric $Pb(Zr_{0.52}Ti_{0.48})O_3$ (PZT). It has been found that the bilayer has both giant magnetoelectric voltage coefficient and high coupling efficiency when it operates at the first bending resonance mode. The maximum bending resonant magnetoelectric voltage coefficient of 14.6 V/cm Oe at quite low frequency of 12.5 kHz was observed in the bilayer of small sizes of $17.4\times4.0\times1.44$ mm^3. This bending-resonance-type magnetoelectric effect shows promising application in transducers for magnetoelectric energy conversion. © *2005 American Institute of Physics*. [DOI: 10.1063/1.1935040]

Magnetoelectric (ME) effect has been receiving great and continuous interest due to its potential applications in sensors, memory device, and transducers for magnetoelectric energy conversion.[1–3] A large ME effect could be realized in a layered composite of the piezoelectric phase and the magnetostrictive phase by the product property.[4] That is, a magnetic field applied to the layered ME composite results in a mechanical strain in the magnetostrictive layer that is subsequently transferred to the piezoelectric layer by the mechanical stress-mediated interface coupling and lead to an electric polarization. In the past several years, various layered ME composites, such as $Tb_{1-x}Dy_xFe_{2-y}$(Terfenol-D)/Pb(Zr,Ti)O_3(PZT), ferrite / PZT, and Terfenol-D/PVDF laminates, were widely investigated.[5–11] The reported ME voltage coefficient is generally below ~5.0 V/cm Oe.

Recently, people found that the frequency affected significantly the ME coupling in the laminates. When the laminate operates in the resonance mode, its ME effect could be enhanced largely, generally yielding a ME voltage output of nearly two orders of magnitude over that of the nonresonant ME laminates.[12–16] To date, many experiments and calculations have been performed to optimize the resonant ME output for the laminates. The developed ME resonance modes include the longitudinal mode (for the rectangle laminates) (Ref. 13) and radial mode (for disk laminates).[12] It has been proved that the resonant ME laminates could be used potentially for the high-voltage miniature transformer applications.[14] However, a problem for the current ME resonance modes is that the operating frequencies are generally high, which could bring about significant eddy current loss for the magnetostrictive phase, especially for the large magnetostrictive earth rare alloys such as Terfenol-D, resulting in an inefficient magnetoelectric energy conversion. An alternative solution is to reduce the operating frequency. Unfortunately it will make the size of the laminate largely increase, which is disadvantageous to the actual application.

In this letter, we propose a bending-resonance-type ME bilayer. We note that the flexural deformation will take place if an external magnetic field is applied to the magnetostrictive/piezoelectric bilayer due to the nonsymmetrical stress distribution in the magnetostrictive and piezoelectric layers. When the bilayer operates in the bending vibration mode, its resonance frequency will be much lower than that of the other modes; meanwhile its size could be kept quite small and ME coupling is strong. In this Letter, we report our experiments on the flexural resonant ME bilayer of magnetostrictive Terfenol-D ($Tb_{0.30}Dy_{0.7}Fe_2$)/Epoxy and piezoelectric PZT [$Pb(Zr_{0.52}Ti_{0.48})O_3$], which yields a giant ME output with a quite low operating frequency and a high coupling efficiency.

Our ME bilayer was prepared by stacking and bonding the rectangle Terfenol-D / Epoxy composite (TEC) and PZT-5 pieces with epoxy binder. The PZT layer was polarized in the thickness direction and the TEC layer was magnetized along the longitudinal direction, as shown in Fig. 1. The volume content of Terfenol-D in the TEC is 0.8. The preparation process of the TEC can be found elsewhere.[17] The typical piezomagnetic coefficient $d_{31,t}$ of TEC and the piezoelectric coefficient $d_{31,p}$ of PZT were measured to be 5.9 nm/A at 1 kOe and 155 pm/V, respectively. Both the TEC and PZT layers have the same sizes of 17.4 mm(length, l)$\times$4.5 mm(width, w) $\times$0.72 mm(thickness, $d/2$).

We firstly analyzed the vibrational modes of the bilayer. Before bonding, the first longitudinal magnetomechanical resonance (MMR) for the TEC piece and electromechanical resonance (EMR) for the PZT piece occur at 50.4 and 86.5 kHz, respectively. After bonding, both the MMR and EMR frequencies of the bilayer tend to be consistent, and the first

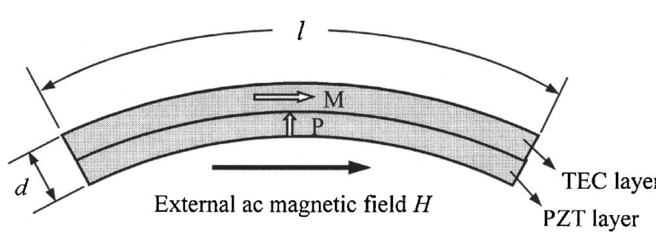

FIG. 1. Configuration and operation principle of the bending vibrational magnetoelectric TEC/PZT bilayer. The arrows *P* and *M* show the polarized direction of PZT and the magnetized direction of TEC, respectively.

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TABLE I. Bilayer parameters for the TEC and PZT.

	ρ_t or $\rho_p(\mathrm{kg/m^3})$	s^t_{11} or $s^p_{11}(\mathrm{m^2/N})$	v_t or v_p
TEC	7.64×10^3	42.5×10^{-12}	0.50
PZT-5	7.70×10^3	15.4×10^{-12}	0.50

longitudinal resonance of the bilayer shifts to a frequency between 50.4 and 86.5 kHz due to the acoustic velocity difference between the TEC and PZT. The average acoustic velocity $\overline{V}$ of the bilayer can be derived from the classical plate theory combined with a simple composite principle

$$\overline{V} = 1/\sqrt{\overline{\rho s_{11}}}, \tag{1}$$

where $\overline{\rho}$ is the average density of the bilayer that is determined by $\overline{\rho}=v_t\rho_t+v_p\rho_p$, and $\overline{s_{11}}$ is the equivalent elastic compliance of the bilayer that is followed by the parallel composite law and given by $\overline{s_{11}}=s^t_{11}s^p_{11}/(v_p s^t_{11}+v_t s^p_{11})$, where ρ_t or ρ_p are the density of the TEC and PZT, s^t_{11} or s^p_{11} are the elastic compliance of the TEC and PZT, v_t or v_p are the volume content of the TEC and PZT, respectively. The longitudinal resonance frequency is thus evaluated by

$$f^L_n = \frac{n}{2l}\overline{V}. \tag{2}$$

Meanwhile, the flexural vibrational modes appear in the bilayer due to the flexural deformation caused by the nonsymmetrical stress distribution in the TEC and PZT layers. Assuming that the two sides of the bilayer are free and applying the one-dimensional approximation, we describe the bending resonance frequencies of the bilayer as follows:

$$f^B_n = \frac{\pi d}{2\sqrt{12}l^2}\overline{V}\beta^2_n, \tag{3}$$

where $\beta_n \approx n+1/2$. Using the typical bilayer parameters listed in Table I, we predicted the first, second, and third bending resonance frequencies of the bilayer to be 11.7, 32.5, and 63.7 kHz, and 69.0 kHz for the first longitudinal resonance frequency. The further vibration modal analyses for the bilayer were subsequently performed by using a laser doppler vibrometer (Polytec, Germany). Figure 2 presents the measured frequency response of the surface displacement of one free side of the bilayer induced by an ac magnetic field of 5 Oe at magnetic bias of 0.7 kOe. One easily identifies that the resonance peak at 69.2 kHz is attributed to the first longitudinal resonance, while the resonance peaks at 12.1, 33.2, 60.1 kHz are ascribed to the first, second, and third bending resonance modes, respectively. The experimental and predicted results are well in agreement with each other. Modal analyses for the bilayer indicates that the first bending resonance frequency was much lower, only about one sixth of the first longitudinal resonance frequency. If such low frequency is required for the first longitudinal resonance, the length of the bilayer must be extended to be as long as ~100 mm.

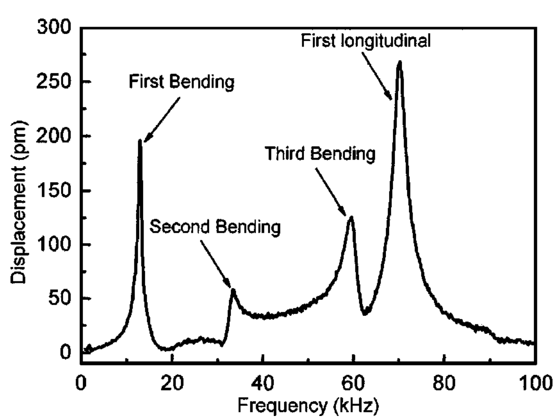

FIG. 2. Frequency response of the surface displacement of one free side of the bilayer induced by an ac magnetic field of 5 Oe at $H_{\mathrm{Bias}}=0.7$ kOe.

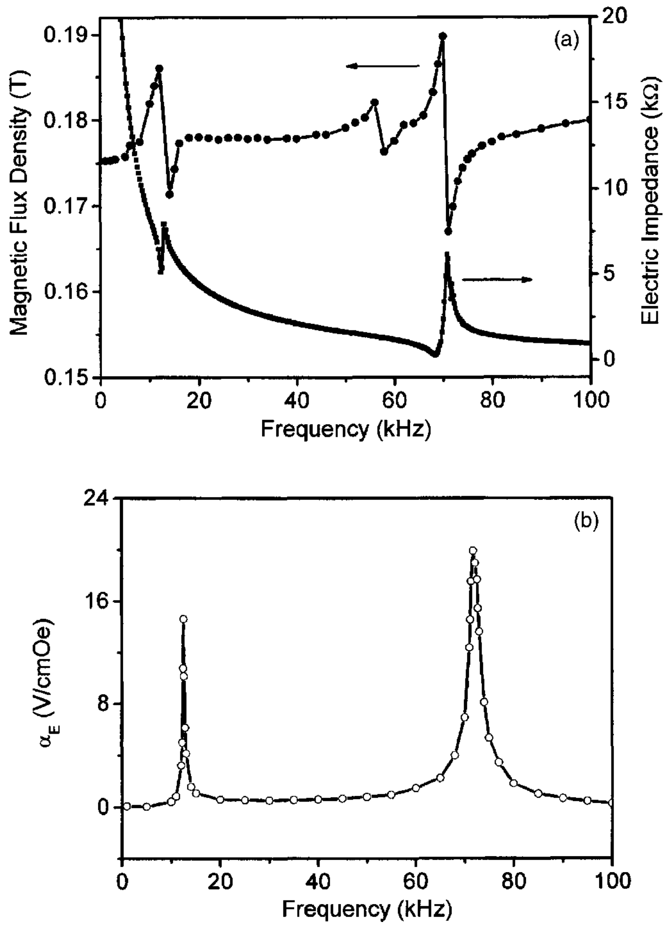

FIG. 3. (a) Frequency response of magnetic flux density for TEC piece and electric impedance for the PZT piece in the bilayer. (b) Frequency dependence of α_E for the bilayer. An optimal $H_{\mathrm{Bias}}=0.7$ kOe was set for maximum ME output.

We further investigate the magnetoelectric coupling in the bilayer. Figure 3(a) presents the frequency response of magnetic flux density for the TEC and electric impedance for the PZT in the bilayer. The effective electromechanical coupling coefficient k_p for the PZT and magnetomechanical coupling coefficient k_m for the TEC can be evaluated from the resonance frequency f_r and antiresonance frequency f_a, in the expression

$$k_{m \text{ or } p} = [1-(f_r/f_a)^2]^{1/2}. \tag{4}$$

By taking the characteristic frequencies from Fig. 3(a), the k_p for the first bending resonance is calculated to be ~0.33, slightly higher than that for the first longitudinal resonance, which is ~0.26. While the k_m for the first bending resonance is ~0.51, much higher than that for the first longitudinal resonance, which is only ~0.21. If the energy loss in the interface coupling is neglected, we evaluate the effective magnetoelectric coupling coefficient by $k_{\rm eff}=k_m k_p$. Accordingly, the evaluated $k_{\rm eff}$ for the first bending resonance is $\sim$0.17, being about three times of that for the first longitudinal resonance, which is only $\sim$0.05.

The ME voltage coefficient $\alpha_E = dE/dH$, which is determined by the induced electric field E under a small ac magnetic field $H=5$ Oe, was then tested for the bilayer. The ac magnetic field H was superimposed onto a dc magnetic bias $H_{\rm Bias}$ and both were parallel to the longitudinal direction of the bilayer. The induced electric field E was measured using a lock-in amplifier (SRS Inc, SR830). Figure 3(b) plots the profile of α_E vs frequency for the bilayer at an optimized $H_{\rm Bias}=0.7$ kOe at which the ME effect is maximal. The maximum values of $\alpha_E=14.6$ V/cm Oe at 12.5 kHz for the first bending resonance and $\alpha_E=19.9$ V/cm Oe at 71.6 kHz for the first longitudinal resonance were observed. The ME output in the second and third bending resonance modes were not observed due to their weak vibration. According to the resonant ME frequency f_r and 3-dB frequency bandwidth Δf, we obtained the effective mechanical quality factor $Q_m = f_r/\Delta f$ to be $\sim$48 for the first bending resonance and $\sim$51 for the first longitudinal resonance.

It is well known that the energy conversion efficiency η for a transducer is relative to the effective mechanical quality factor Q_m and the effective coupling coefficient $k_{\rm eff}$.[18] High $k_{\rm eff}$ and Q_m values will lead to a large η value. With respect to the ME coupling of the bilayer operated in the first bending resonance mode, the effective magnetoelectric coupling coefficient $k_{\rm eff}$ is much higher, while its Q_m value is comparable to that of the first longitudinal resonance, though the α_E value induced by the first bending resonance is somewhat lower than that of the first longitudinal resonance. Therefore, we infer that the magnetoelastoelectric energy conversion operated in the first bending resonance mode is more efficient in the bilayer. In addition, the much smaller geometric size for the bending resonance mode is also advantageous to miniaturize the devices. These advantages show that this magnetostrictive/piezoelectric bilayer operated at the bending resonance mode is more suitable to the transducer application for magnetoelectric energy conversion.

In summary, a giant ME voltage output operated at quite low frequency has been found in the first bending resonant TEC/PZT bilayer with very small size. It was also shown that the first bending resonant ME coupling efficiency was higher than the others. The current experimental results indicate that this bending-resonance-type ME effect has promising applications in the transducer devices for magnetoelectric energy conversion.

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[1] T. H. O'Dell, Electron. Power **11**, 266 (1965).
[2] L. P. M. Bracke and R. G. van Vliet, Int. J. Electron. **51**, 255 (1981).
[3] S. X. Dong, J. F. Li, and D. Viehland, Appl. Phys. Lett. **83**, 2265 (2003).
[4] J. van den Boomgaard and R. A. J. Born, J. Mater. Sci. **13**, 1538 (1978).
[5] G. Srinivasan, E. T. Rasmussen, J. Gallegos, R. Srinivasan, Yu. I. Bokhan, and V. M. Laletin, Phys. Rev. B **64**, 214408 (2001).
[6] G. Srinivasan, E. T. Rasmussen, B. J. Levin, and R. Hayer, Phys. Rev. B **65**, 134402 (2002).
[7] J. Ryu, A. V. Carazo, K. Uchino, and H. E. Kim, Jpn. J. Appl. Phys., Part 1 **40**, 4948 (2001).
[8] J. Ryu, S. Priya, A. V. Carazo, and K. Uchino, J. Am. Ceram. Soc. **84**, 2905 (2001).
[9] K. Mori and M. Wuttig, Appl. Phys. Lett. **81**, 100 (2002).
[10] C. W. Nan, L. Liu, N. Cai, J. Zhai, Y. Ye, and Y. H. Lin, Appl. Phys. Lett. **81**, 3831 (2002).
[11] C. W. Nan, G. Liu, and Y. H. Lin, Appl. Phys. Lett. **83**, 4366 (2003).
[12] M. I. Bichurin, D. A. Filippov, V. M. Petrov, V. M. Laletsin, N. Paddubnaya, and G. Srinivasan, Phys. Rev. B **68**, 132408 (2003).
[13] S. X. Dong, J. R. Cheng, J. F. Li, and D. Viehland, Appl. Phys. Lett. **83**, 4812 (2003).
[14] S. X. Dong, J. F. Li, D. Viehland, J. Cheng, and L. E. Cross, Appl. Phys. Lett. **85**, 3534 (2004).
[15] U. Laletsin, N. Padubnaya, G. Srinivasan, and C. P. DeVreugd, Appl. Phys. A **78**, 33 (2004).
[16] S. X. Dong, J. F. Li, and D. Viehland, Appl. Phys. Lett. **85**, 2307 (2004).
[17] J. G. Wan, J.-M. Liu, H. L. W. Chan, C. L. Choy, G. H. Wang, and C. W. Nan, J. Appl. Phys. **93**, 9916 (2003).
[18] F. X. Zhang and K. Shun, *Piezoelectricity* (National Defence Industry Press, China, 1984).