JOURNAL OF APPLIED PHYSICS **105**, 061622 (2009)

Magnetoelectric study in Terfenol-D/$Tb_2(MoO_4)_3$ bilayer composite

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(Received 30 June 2008; accepted 30 October 2008; published online 16 March 2009)

The dependence of the magnetoelectric coefficients (α_E) in the Terfenol-D/$Tb_2(MoO_4)_3$ bilayer composite on the magnetic bias fields and the frequencies has been studied. The strain with butterfly loop of $Tb_2(MoO_4)_3$ under an external magnetic field was observed and the effect of ferroelasticity of the $Tb_2(MoO_4)_3$ on the magnetoelectric coupling was discussed. More than one peak of α_E as function of frequency of ac magnetic field were observed and it implies existence of bending resonance mode besides the longitudinal resonance mode in the bilayer composite. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3055418]

I. INTRODUCTION

Materials in which at least two ferroic orders (ferromagnetism, ferroelectricity, or ferroelasticity) occur simultaneously and allow coupling between different order parameters are known as multiferroics.[1,2] The magnetoelectric (ME) response is the appearance of an electric polarization $\boldsymbol{P}$ under a magnetic field $\boldsymbol{H}$ (i.e., the direct ME effect, designated as ME_H effect) and/or the appearance of a magnetization $\boldsymbol{M}$ upon applying an electric field $\boldsymbol{E}$ (i.e., the converse ME effect or ME_E).[3] Usually, the ME effect in the single phase materials is either too weak or occurs at too low temperature to be exploited practically. Alternatively and with greater design flexibility, multiferroic ME composites made by combining piezoelectric and magnetic substances together such as $CoFe_2O_4/PbZr_xTi_{1-x}O_3$ (PZT), Terfenol-D/PZT, and Terfenol-D-PVDF/PZT-PVDF have drawn significant interest in recent years due to their multifunctionality, in which the coupling interaction between piezoelectric and magnetic substances could produce a large ME response.[4] The ME voltage coefficient in a two-phase system is much larger than that in the single phase materials usually.[5–14] One of largest ME voltage coefficients of 500 $V\ cm^{-1}\ Oe^{-1}$ was reported recently for a high permeability magnetostrictive piezofiber laminate.[15] However, ferroelasticity in study of the ME effect is mentioned rarely. In this paper, the Terfenol-D($Tb_xDy_{1-x}Fe_2$)/$Tb_2(MoO_4)_3$ bilayer composites were fabricated. Then the ME effect and ferroelasticity effect were studied.

II. EXPERIMENTAL

The ME bilayer composite was prepared by stacking and bonding the rectangle Terfenol-D ($10\times4\times1\ mm^3$) and $Tb_2(MoO_4)_3$ single crystal ($12\times6\times1\ mm^3$ with the thickness along the [001] direction) pieces with epoxy binder as shown in Fig. 1. The grain orientation of Terfenol-D piece was [211], along the length direction, which can generate a highest magnetostrictive strain of ~1100 ppm along this direction. $Tb_2(MoO_4)_3$ (TMO) is a new type ferroelastic-ferroelectric (weak) single crystal growth by a Czochralski technology. The ME measurement instrument includes an electromagnet $H_{dc}\sim0-7$ kOe and a Helmholtz coil $H_{ac}=1$ Oe, which were applied along the length direction of the laminates. The induced ME voltages across the TMO layer were measured by an oscilloscope and a lock-in amplifier at the same frequency of the driving ac magnetic field.[16–18] The ME voltage coefficient α_E ($\partial V_{ME}/\partial H$) used designates the ME effect here.

III. RESULTS AND DISCUSSION

TMO is a ferroelastic single crystal with weak ferroelectricity. The x-ray diffraction patterns of TMO (not shown here) exhibit a tetragonal $C_{\bar{4}2_1m}$ structure at room temperature, and it has the similar structure as $Gd_2(MoO_4)_3$ (GMO). Figure 2(a) is the differential scanning calorimetry (DSC) result of TMO single crystal that reflects a phase change appearing at $T_c=432$ K. It is exact same with the T_c of GMO (a transition from the tetragonal $p_{\bar{4}2_1m}$ to the orthorhombic P_{ba2}). Figures 2(b) and 2(c) are relative dielectric constant ε and dielectric loss tan δ as functions of temperature in TMO single crystal respectively. Also a peak of relative dielectric constant ε and dielectric loss tan δ around T_c can be observed.

The ME voltage coefficient α_E as a function of H_{dc} at frequencies of 35 and 40 kHz is shown in Fig. 3. With in-

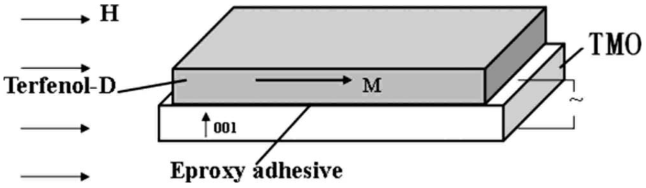

FIG. 1. Schematic illustration of the Terfenol-D/$Tb_2(MoO_4)_3$ bilayer composite.

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0021-8979/2009/105(6)/061622/3/$25.00

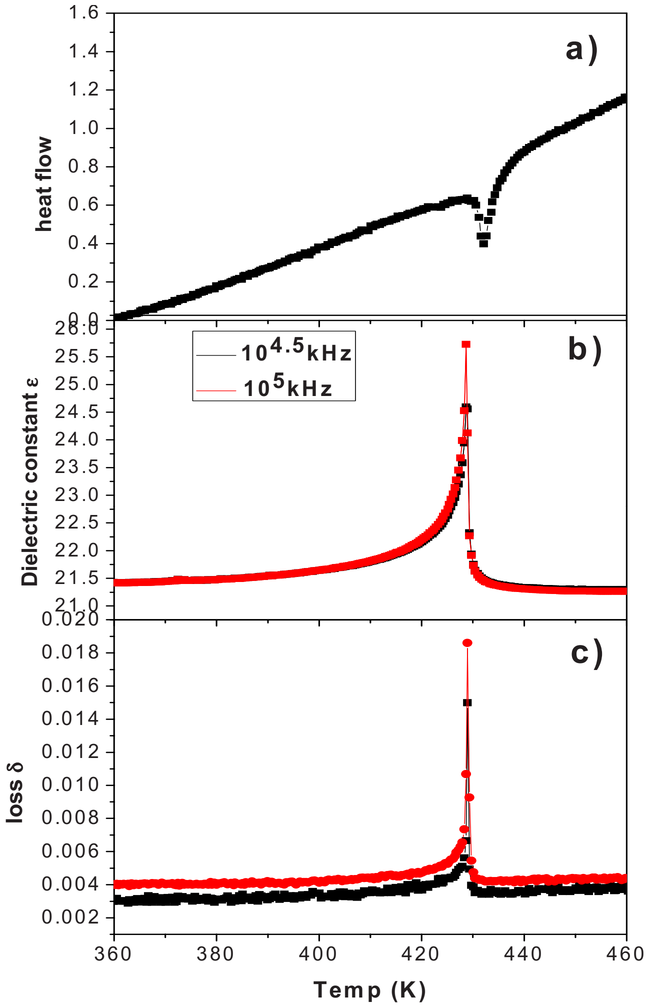

FIG. 2. (Color online) (a) DSC analysis of TMO; (b) dielectric constant ε of TMO; (c) dielectric loss tan δ of TMO.

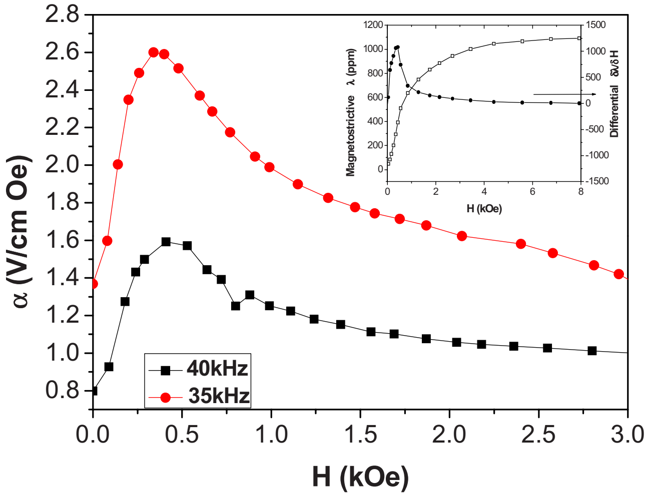

FIG. 3. (Color online) ME voltage coefficient as a function of applied H_{dc}.

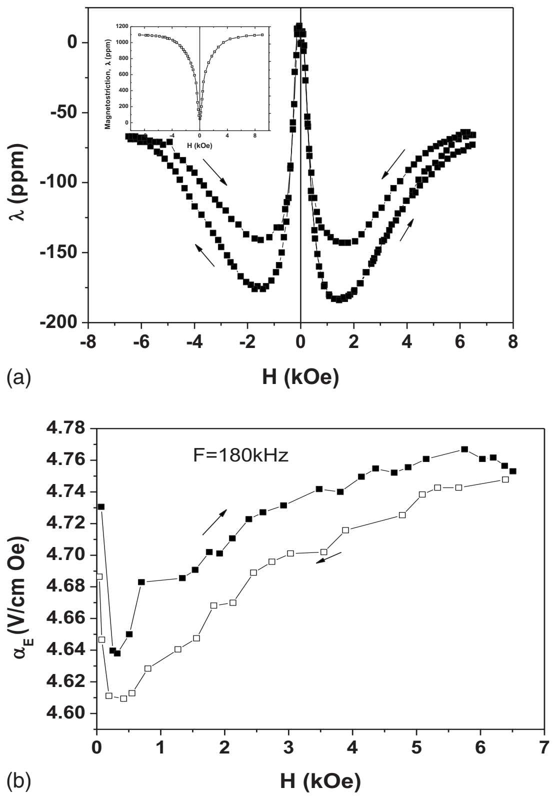

FIG. 4. (a) The surface longitudinally strain of TMO crystal under various magnetic fields. The inset is the magnetostriction of Terfenol-D dependence on H_{dc}. (b) The ME coupling coefficient loop with an increasing and decreasing of H_{dc}.

creasing H_{dc}, the ME voltage coefficient α_E increased and reached its maximum value at $H_{dc} \sim 0.5$ kOe, then the α_E decreased to a stable value with increasing H_{dc}. The inset of Fig. 3 is the magnetostriction λ and piezomagnetic coefficient $\delta\lambda / \delta H$ of Terfenol-D. A similar dependence of the α_E and the piezomagnetic coefficient on the field was observed in Fig. 3 and both the largest values appear at H_{dc} (0.5 kOe). It indicates that the α_E has a correlation with the piezomagnetic coefficient $\delta\lambda / \delta H$ of Terfenol-D as observed in $(Tb_xDy_{1-x}Fe_2)/[0.52\ Pb(Fe_{1/2}Nb_{1/2})O_3-0.48\ PbTiO_3]$ laminated composites in our laboratory.[19]

Figure 4(a) describes the longitudinally strain of TMO single crystal in the Terfenol-D/$Tb_2(MoO_4)_3$ bilayer composite under various magnetic fields. The strain was measured by some strain gages as the sensor and an instrument with Wheatstone bridge to record the value of strain. During the measurement, one strain gage is placed on the surface of TMO single crystal and another is placed on a piece of silicon as comparison for the Si has no strain response to the magnetic field. When a magnetic field is applied to the composite, the Terfenol-D piece will produce a deformation for its magnetostriction. The deformation will pass to the TMO piece (as an external stress) and then result in strain of TMO. The strain of the TMO in the Terfenol-D/$Tb_2(MoO_4)_3$ bilayer composite is a compressive one ($\lambda < 0$) under various magnetic fields, as shown in Fig. 4(a). When the Terfenol-D piece elongates, the TMO piece shortens at first due to the bending strain caused by the modulus discrepancy of Terfenol-D and TMO, then elongates with Terfenol-D. The inset of Fig. 4(a) is a dependence of the magnetostriction of Terfenol-D on H_{dc}. It exhibits as a symmetric curve and overlaps with increasing and decreasing H_{dc} and no strain was observed at null H_{dc}. However the strain of TMO as a function of H_{dc} has a butterfly loop (strain-stress lag) and nonzero strain (spontaneous strain) in contrast, which dem-

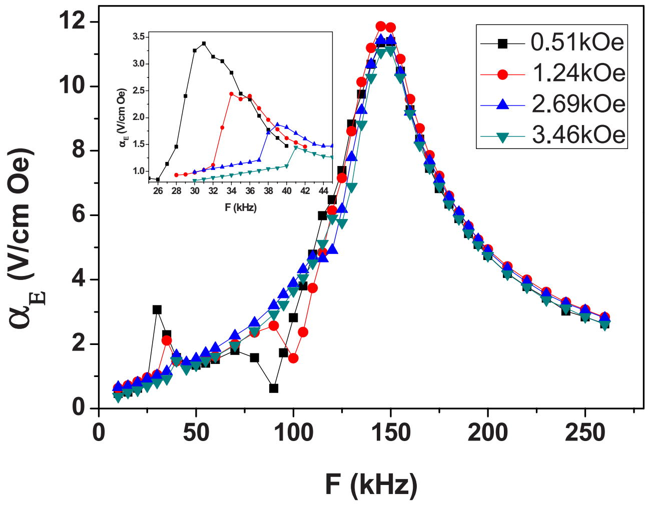

FIG. 5. (Color online) Frequency responses of ME voltage coefficient under H_{dc}=0.51, 1.24, 2.69, and 3.46 kOe, respectively. The inset indicates the bending resonance frequencies under different H_{dc}.

onstrates an existence of the ferroelasticity of TMO. This ferroelasticity effect has a strong influence on the strain of TMO besides the pure deformation induced by the magnetostriction of Terfenol-D. Figure 4(b) is the ME coupling coefficient α_E versus H_{dc} during increasing and decreasing of the field, also a loop and a nonzero value at null field are observed obviously, which demonstrated the influence of the ferroelastic effect on ME effect. It is valuable to study further the effect of ferroelasticity on the ME effect.

Figure 5 plots the dependence of α_E on frequency of ac magnetic field over a wide frequency range of $0 < f < 260$ kHz for the bilayer at H_{dc}=0.51, 1.24, 2.69, and 3.46 kOe, respectively. The maximum value of $\alpha_E = 12$ V/cm Oe was got at 150 kHz, which could be attributed to the electromechanical resonance (EMR). Furthermore, a peak of α_E=3.04 V/cm Oe is also observed at lower frequency around 30 kHz. Usually, a flexural vibrational mode will appear in the bilayer due to the flexural deformation caused by the nonsymmetrical stress distribution in the magnetostrictive and piezoelectric layers,[20] so the peak at 30 kHz comes from this vibrational mode caused by the nonsymmetrical stress distribution of the TMO and Terfenol-D. The inset of Fig. 5 indicates the bending resonance frequencies under different H_{dc} and the peak position shifts toward high H_{dc}. It is shown that the α_E at the bending vibration resonance is also much higher than one in single phase ME materials. The bending vibrational mode is more complex and sensitive to the different conformation. The bending conformation will alter largely under various H_{dc} due to the nonsymmetrical stress distribution in the magnetostrictive and piezoelectric layers which causes the peak position shifts toward high H_{dc} of the resonance frequencies.

IV. CONCLUSIONS

In summary, the coupling between magnetic field, polarization and the ferroelastic strain in bilayer Terfenol-D/$Tb_2(MoO_4)_3$ composite were investigated. The maximum values of α_E=12 V/cm Oe at 150 kHz for the EMR and α_E=3.04 V/cm Oe at 30 kHz for the bending resonance were observed. Ferroelasticity of TMO and its influence on the ME effect were demonstrated by the strain-stress lag and the loop of ME coupling coefficient versus H_{dc}. The effect of ferroelasticity on ME coupling needs further study.

ACKNOWLEDGMENTS

This research was supported by the NSFC (Grant Nos. 50672034 and 50832002), the National Key Basic Research Program of China (Grant Nos. 2009CB623300, 2009CB929501, and 2006CB921804), and the Research Grant No.G-YF79 of the Hong Kong Polytechnic University, NJU test foundation.

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