Magnetoelectric study in Terfenol-D/Tb$_2$(MoO$_4$)$_3$ bilayer composite

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The dependence of the magnetoelectric coefficients ($\alpha_E$) 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 $\alpha_E$ 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 $P$ under a magnetic field $H$ (i.e., the direct ME effect, designated as $ME_H$ effect) and/or the appearance of a magnetization $M$ upon applying an electric field $E$ (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$_2$O$_4$/PbZrTi$_{1−x}$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 magnetostriective piezofiber laminate.$^{15}$ However, ferroelasticity in study of the ME effect is mentioned rarely. In this paper, the Terfenol-D/Tb$_2$(Dy$_{1−x}$Fe$_x$)/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 $\sim$1100 ppm along this direction. Tb$_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}$$\sim$0–7 kOe and a Helmholz 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 $\alpha_E$ ($\partial V_{ME}/\partial H$) 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 $C_{4z/m}$ 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 $P_{4z/m}$ to the orthorhombic $P_{ba2}$). Figures 2(b) and 2(c) are relative dielectric constant $\varepsilon$ and dielectric loss tan $\delta$ as functions of temperature in TMO single crystal respectively. Also a peak of relative dielectric constant $\varepsilon$ and dielectric loss tan $\delta$ around $T_c$ can be observed.

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

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increasing $H_{dc}$, the ME voltage coefficient $\alpha_E$ increased and reached its maximum value at $H_{dc} \approx 0.5$ kOe, then the $\alpha_E$ decreased to a stable value with increasing $H_{dc}$. The inset of Fig. 3 is the magnetostriction $\lambda$ and piezomagnetic coefficient $\delta\lambda/\delta H$ of Terfenol-D. A similar dependence of the $\alpha_E$ 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 $\alpha_E$ has a correlation with the piezomagnetic coefficient $\delta\lambda/\delta H$ of Terfenol-D as observed in (Tb$_x$ Dy$_{1-x}$)$_2$Fe$_2$O$_2$/[Pb(Fe$_{1/2}$Nb$_{1/2}$)$_2$O$_3$–0.48 PbTiO$_3$] laminated composites in our laboratory.

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 under various magnetic fields. The inset is the magnetostriction of Terfenol-D dependence 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 (stress-strain lag) and nonzero strain (spontaneous strain) in contrast, which dem-
demonstrates 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 magnetostriiction of Terfenol-D. Figure 4(b) is the ME coupling coefficient $\alpha_E$ 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 $\alpha_E$ 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 $\alpha_E = 3.04$ 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 magnetostriictive and piezoelectric layers, 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 $\alpha_E$ 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 magnetostriictive 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 $\alpha_E = 12$ V/cm Oe at 150 kHz for the EMR and $\alpha_E = 3.04$ 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.

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