Enhanced magnetoelectric effect in longitudinal-transverse mode Terfenol-D/$Pb(Mg_{1/3}Nb_{2/3})O_3-PbTiO_3$ laminate composites with optimal crystal cut

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Magnetoelectric (ME) laminate composite consisting of optimal crystal cut thickness-polarized piezoelectric $0.7Pb(Mg_{1/3}Nb_{2/3})O_3-0.3PbTiO_3$ (PMN-PT) single crystal and the length-magnetized magnetostrictive $Tb_{0.3}Dy_{0.7}Fe_{1.92}$ (Terfenol-D) alloy has been fabricated. The cut optimization of PMN-PT crystal greatly enhances the longitudinally magnetized-transversely polarized (*L-T*) mode ME effect, which has a superior ME voltage coefficient α_E of ~3.02 V/cm Oe in low frequency band. Near the resonance frequency of 95 kHz, the coefficient dramatically increases and reaches the maximized value of 33.2 V/cm Oe, which is almost two times larger than the previously reported ⟨001⟩-oriented PMN-PT crystal based *L-T* mode laminate composite. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2943267]

I. INTRODUCTION

The magnetoelectric (ME) effect is the polarization *P* response to an applied magnetic field *H*, and the converse ME effect is a magnetization *M* response to an applied electric field *E*.[1] In the past decades, considerable research efforts have been put on the ME effect, first in single-phase materials, then in two-phase bulk composites, and lately in two-/three-phase laminate composites.[2–9] The magnetic-to-electric field conversion in ME composite is a magnetoelastoelectric coupling.[3] The ME coupling in laminate composite is realized by a stress mediated interaction between the magnetostrictive phase (or ferromagnet) and the piezoelectric (or ferroelectric) phase, which is often referred to as a multiferroic composite.[4] Some simple architecturally engineered nanostructured composites, such as $CoFe_2O_4$ nanopillars in a $BaTiO_3$ matrix, have also recently been reported.[5] It seems that these nanocomposites should have potential for high ME coupling because the two phase materials have a more intimate contact in nanodimension. But these nanostructured composites suffer from the drawback that the magnetostrictive and piezoelectric effects are dramatically decreased in nanodimension due to substrate's clamping effect.[6] To date, it is known that laminated composites of piezoelectric $0.7Pb(Mg_{1/3}Nb_{2/3})O_3-0.3PbTiO_3$ (PMN-PT) single crystal and magnetostrictive $Tb_{0.3}Dy_{0.7}Fe_{1.92}$ (Terfenol-D) alloy possess superior ME effect and ultrahigh magnetic field sensitivity due to their greater product effect of the piezoelectric effect and the magnetostrictive effect.[7] However, the orientation of piezoelectric PMN-PT single crystal was out of consideration in previous studies. Actually, based on the working principle of *L-T* mode laminate composite, the Terfenol-D mainly vibrates longitudinally when the applied magnetic field is along the length direction. Due to the mechanical coupling, the PMN-PT single crystal has to vibrate synchronously along the length direction and generates electric charge along the thickness direction. So the piezoelectric coefficient d_{31} and electromechanical coupling factor k_{31} should contribute greatly to the ME effect.[7] Consequently, designing new optimal cut-types in the single crystal piezoelectric phase to produce optimized transverse piezoelectric performance is a new method to enhance the applications of ME effect.

In this paper, we have developed an *L-T* mode ME laminate composite by sandwiching a special-oriented piezoelectric PMN-PT single crystal between two longitudinally magnetized magnetostrictive Terfenol-D plates.

II. EXPERIMENTS

A. Structure and fabrication

Figure 1 shows schematic diagram of the proposed sand-

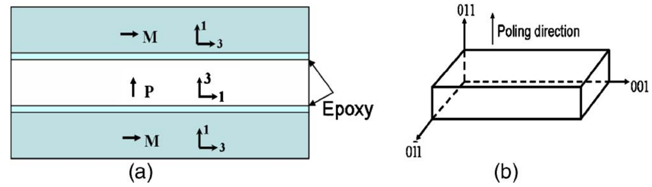

FIG. 1. (Color online) (a) Schematic diagram of the proposed magnetostrictive/piezoelectric laminated composites. The arrows designate the magnetization and polarization directions, respectively. (b) The newly designed special-oriented PMN-PT single crystal. The ⟨001⟩, ⟨0$\bar{1}$1⟩, and ⟨011⟩ crystallographic axes are oriented in the length, width, and thickness directions, respectively.

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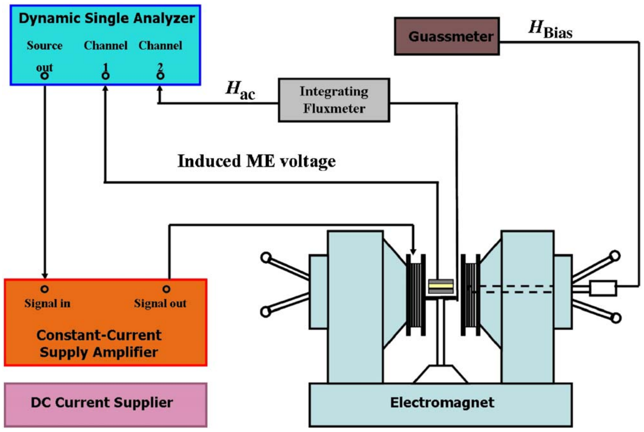

FIG. 2. (Color online) Schematic diagram of the ME measurement setup.

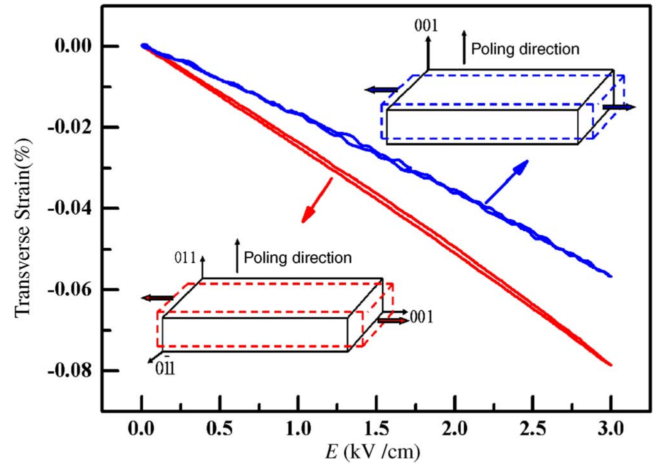

FIG. 3. (Color online) Strain vs E-field (unipolar) curves in the $\langle 001\rangle$-oriented and newly designed cut-type PMN-PT crystals.

wiched composite structure with the ME effect and the optimal cut-type of PMN-PT plate. The high quality PMN-PT single crystal was grown directly from the melt by modified Bridgman technique.[10] The as-grown single crystals were oriented along $\langle 001\rangle$, $\langle 011\rangle$, and $\langle 0\bar{1}1\rangle$ directions using an x-ray diffraction meter, and then diced to prepare samples with the crystal cut and dimensions of $12\langle 001\rangle^L \times 6\langle 0\bar{1}1\rangle^w \times 1\langle 011\rangle^T$ mm^3 (L: length, W: width, T: thickness). It has been shown in our previous work that PMN-PT plates, with this specially cut and poled along the $\langle 011\rangle$ thickness direction, possess ultrahigh transverse piezoelectric performance, i.e., ultrahigh thickness direction voltage response to length direction strain deformation.[11] The Terfenol-D plates were commercially supplied (Baotou Research Institute of Rare Earth, China) with the same dimensions as the PMN-PT plate and with the length direction along the [112] direction, which is the highly magnetostrictive crystallographic axis and the magnetization (M) was relatively easy to achieve.[12]

B. Measurement setup and procedure

The ME properties of the laminate composite were characterized at room temperature and zero stress bias using an in-house automated measurement system[13] shown in Fig. 2. The ME voltages (V) induced in the composites were measured as a function of ac magnetic field (H_{ac}), dc magnetic bias (H_{bias}), and ac magnetic field frequency (f) in the ranges of $10^{-7}-10^{-3}$ T, 0–1200 Oe, and 1–100 kHz, respectively. H_{ac} was provided by Helmholtz coils driven by a dynamic signal analyzer (Ono Sokki CF5220) via a constant-current supply amplifier (AE Techron 7572). H_{bias} was supplied by a water-cooled, U-shaped electromagnet (Myltem PEM-8005K) controlled by a dc current supply (Sorensen DHP200-15). H_{ac} and H_{bias} were monitored *in situ* by a pick-up coil connected to an integrating fluxmeter (Walker MF-10D) and a Gaussmeter (F. W. Bell 7030), respectively. All quantities were sampled and recorded by the dynamic signal analyzer and stored in a computer.

III. RESULTS AND DISCUSSION

It is well known that PMN-PT single crystal has superior piezoelectric effect and electromechanical coupling performance. However, the piezoelectric effect of PMN-PT is anisotropic, depending significantly on the crystal cut type and the poling direction. Consequently, optimization of the crystal cut and poling processing of PMN-PT single crystals should be considered to produce optimal piezoelectric performance for different piezoelectric resonator modes in versatile applications. Figure 3 shows the electric-field induced strain patterns for the $\langle 001\rangle$-oriented and newly designed cut-type 0.70PMN-0.30PT crystal, respectively, using a unipolar field with amplitude of $E<3$ kV/cm and frequency of 0.2 Hz. From the slope of the plot, the piezoelectric coefficients of d_{31} were determined. The k_{31} was determined using the resonance-antiresonance technique by a HP 4194A impedance analyzer following the IEEE standards, and d_{33} is directly measured by a quasistatic Berlincourt d_{33} meter (50 Hz) (see Table I). From the comparison in Table I, it is obviously observed that the newly designed cut-type PMN-PT crystal is much superior for transverse mode application than the $\langle 001\rangle$-oriented one.

The ME voltage coefficient α_E, defined as $|dE/dH_{ac}|$, of the as-prepared crystal cut optimized laminate composite was then measured for various H_{bias} at H_{ac} of 1 Oe peak and f of 1 kHz, as shown in Fig. 4. α_E initially increases rapidly with H_{bias} and reaches a maximum value of $\sim$3.02 V/cm Oe at an optimal H_{bias} of 400 Oe, then decreases with increasing H_{bias}. In addition, the laminated composite has almost a linear response to H_{bias} in the range of $0<H_{bias}<200$ Oe. Correspondingly, the relationship of the laminated composite can be used to detect small dc magnetic field.[8]

Figure 5 illustrates the induced ME voltage V across the PMN-PT plate as a function of applied H_{ac} over the range of $10^{-7}<H_{ac}<10^{-3}$ T at $f=1$ kHz. It is clear that V has an

TABLE I. Piezoelectric parameters for $\langle 001\rangle$-oriented and newly designed cut-type PMN-PT crystals.

	d_{31} ($\times 10^{-12}$ C/N)	d_{33} ($\times 10^{-12}$ C/N)	k_{31}	$\varepsilon_{33}^T/\varepsilon_0$
$\langle 001\rangle$-oriented	−1126	2420	0.62	4276
$[001]^L \times [\bar{1}10]^W \times [110]^T$-oriented	−2645	2060	0.95	6200

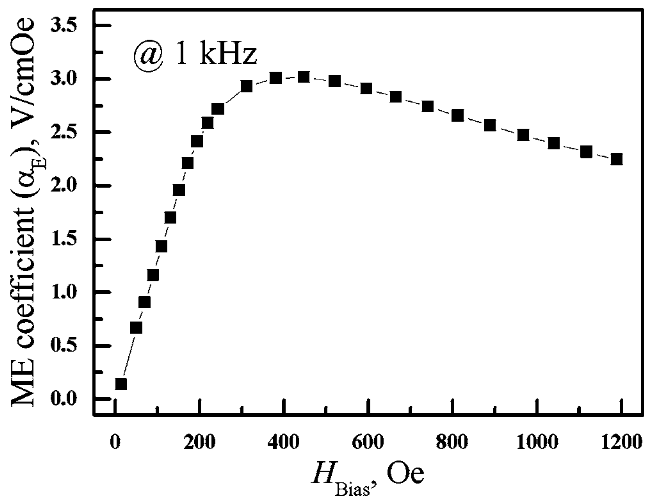

FIG. 4. ME voltage coefficient α_E of the new laminated composite as a function of H_{bias}. These data were taken using $H_{ac}=1$ Oe peak at $f=1$ kHz.

excellent linear response to H_{ac} in the whole measured range for various H_{bias}. A higher detection sensitivity of $10^{-10}-10^{-11}$ T could be obtained if shielding magnetic noise could be adopted and composite fabrication could be improved.[7] From the slope the plot, the ME voltage coefficient α_E for various H_{bias} can also be determined, which coincides reasonably well with α_E versus H_{bias}, as shown in Fig. 4.

The dependence of α_E on f at various H_{bias} is shown in Fig. 6(a). It is noted that no remarkable dispersion of α_E is observed for all cases apart from the variations associated with the ME resonances. The largest α_E is observed at $H_{bias}=400$ Oe for the whole frequency range. In particular, the maximal resonance α_E located at the ME resonances frequency (f^{MER}) of 95.25 kHz under this optimal H_{bias} is as large as 33.2 V/cm Oe. This resonance α_E at f^{MER} is over 13 times larger than its nonresonance α_E of ~3.02 V/cm Oe. Figure 6(b) shows H_{bias} dependence of f^{MER} and α_E at f^{MER}. The results clearly demonstrate that the variation of α_E at f^{MER} is similar to the α_E at nonresonant frequency, as shown in Fig. 4. However, f^{MER} decreases with increasing H_{bias},

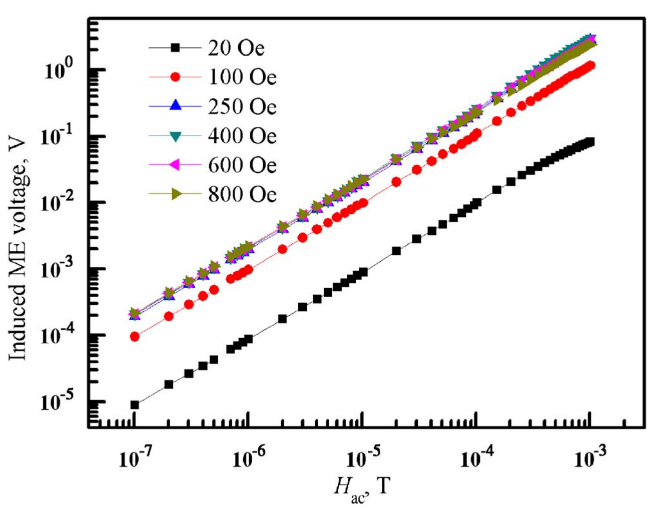

FIG. 5. (Color online) Induced ME voltage V as a function of applied H_{ac} over the range of $10^{-7}<H_{ac}<10^{-3}$ T at $f=1$ kHz for various H_{bias}.

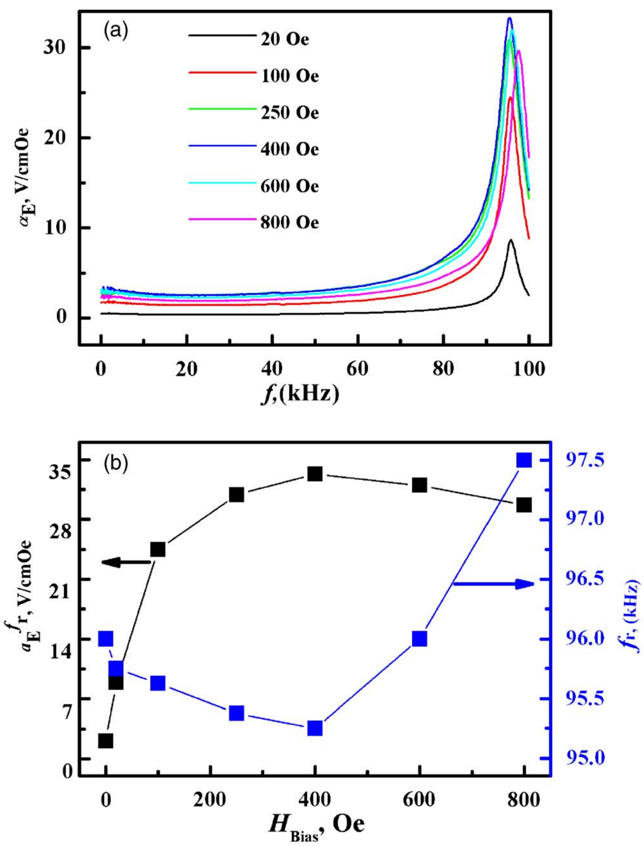

FIG. 6. (Color online) (a) Frequency response of ME voltage coefficient α_E for various H_{bias}, and (b) H_{bias} dependence of ME voltage coefficient at ME resonances $\alpha_E^{f_r}$ and ME resonances frequency f_r.

reaching the smallest value of 95.25 kHz at $H_{bias}=400$ Oe, and then increasing with increasing H_{bias}. Physically, the initial change in both α_E (at f^{MER}) and f^{MER} with increasing H_{bias} can be explained by the H_{bias}-induced motion of the available non-180° domain walls in the Terfenol-D plates.[13] That is, as H_{bias} increases to the 400 Oe critical value, the compliance associated with increased deformation contribution from this non-180° domain-wall motion is maximized, resulting in a maximum in strain (and hence α_E at nonresonant frequency and α_E at f^{MER}) and a minimum in stiffness (and hence f^{MER}). Beyond this optimal and also critical value of H_{bias}, constraining of non-180° domain-wall motion due to interaction with H_{bias} gives rise to a decrease in strain and an increase in stiffness. It is noted that the deformation contribution from the motion of 180° domain walls is insignificant as it produces changes in magnetization without accompanying strain.[9]

IV. SUMMARY

In summary, our specially designed *L-T* mode ME laminate composite by sandwiching PMN-PT plate with the optimal cut type of $\langle 001\rangle^L \times \langle 0\bar{1}1\rangle^w \times \langle 011\rangle^T$ between two Terfenol-D plates enhances the elastoelectric coupling in the magnetic-to-electric field conversion, which results in a significantly increased ME voltage coefficient, approximately two times larger than the previous *L-T* mode laminate composite of ⟨001⟩-oriented PMN-PT single crystal. This reveals a new method to enhance the ME effect of the ME composite.

ACKNOWLEDGMENTS

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