Enhanced magnetoelectric effect in longitudinal-transverse mode Terfenol-D/Pb(Mg\textsubscript{1/3}Nb\textsubscript{2/3})O\textsubscript{3}–PbTiO\textsubscript{3} laminate composites with optimal crystal cut

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Magnetoelastic (ME) laminate composite consisting of optimal crystal cut thickness-polarized piezoelectric 0.7Pb(Mg\textsubscript{1/3}Nb\textsubscript{2/3})O\textsubscript{3}–0.3PbTiO\textsubscript{3} (PMN-PT) single crystal and the length-magnetized magnetostrictive Tb\textsubscript{0.3}Dy\textsubscript{0.7}Fe\textsubscript{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 $\alpha_E$ of $\sim$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 $E$. 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 nanostructured magnetoelectric/piezoelectric laminated composites. The magnetic-to-electric field conversion in ME composite is a magnetoelectric coupling. 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. Some simple architecturally engineered nanostructured composites, such as CoFe\textsubscript{2}O\textsubscript{4} nanopillars in a BaTiO\textsubscript{3} matrix, have also recently been reported. 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. To date, it is known that laminated composites of piezoelectric 0.7Pb(Mg\textsubscript{1/3}Nb\textsubscript{2/3})O\textsubscript{3}–0.3PbTiO\textsubscript{3} (PMN-PT) single crystal and magnetostRICTive Tb\textsubscript{0.3}Dy\textsubscript{0.7}Fe\textsubscript{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. 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. 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 a 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-
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. The as-grown single crystals were oriented along (001), (110), and (011) directions using an x-ray diffraction meter, and then diced to prepare samples with the crystal cut and dimensions of $12(001)^2 \times 6(011)^v \times 1(011)^{v}$ 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 (011) thickness direction, possess ultrahigh transverse piezoelectric performance, i.e., ultrahigh thickness direction voltage response to length direction strain deformation. 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.

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 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 (Mytement 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 (001)-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 $d_{33}$ were determined. The $k_{33}$ 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 ⟨001⟩-oriented one.

The ME voltage coefficient $\alpha_E$, defined as $\frac{|dE|}{dH_{ac}}$, 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. $\alpha_E$ 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.

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

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<th>TABLE I. Piezoelectric parameters for ⟨001⟩-oriented and newly designed cut-type PMN-PT crystals.</th>
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<td>$d_{31}$ ($\times 10^{-12}$ C/N)</td>
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<tr>
<td>(001)-oriented</td>
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<td>[001]$^2$ × [110]$^\parallel$</td>
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<td>[110]$^\parallel$-oriented</td>
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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 ⟨001⟩-oriented and newly designed cut-type PMN-PT crystals.
excellent linear response to $H_\text{ac}$ in the whole measured range for various $H_{\text{bias}}$. A higher detection sensitivity of $10^{-10}$ to $10^{-11}$ T could be obtained if shielding magnetic noise could be adopted and composite fabrication could be improved.\textsuperscript{7} From the slope the plot, the ME voltage coefficient $\alpha_E$ for various $H_{\text{bias}}$ can also be determined, which coincides reasonably well with $\alpha_E$ versus $H_{\text{bias}}$, as shown in Fig. 4.

The dependence of $\alpha_E$ on $f$ at various $H_{\text{bias}}$ is shown in Fig. 6(a). It is noted that no remarkable dispersion of $\alpha_E$ is observed for all cases apart from the variations associated with the ME resonances. The largest $\alpha_E$ is observed at $H_{\text{bias}}=400$ Oe for the whole frequency range. In particular, the maximal resonance $\alpha_E$ located at the ME resonances frequency ($f_{\text{MER}}$) of 95.25 kHz under this optimal $H_{\text{bias}}$ is as large as 33.2 V/cm Oe. This resonance $\alpha_E$ at $f_{\text{MER}}$ is over 13 times larger than its nonresonance $\alpha_E$ of 3.02 V/cm Oe. Figure 6(b) shows $H_{\text{bias}}$ dependence of $f_{\text{MER}}$ and $\alpha_E$ at $f_{\text{MER}}$. The results clearly demonstrate that the variation of $\alpha_E$ at $f_{\text{MER}}$ is similar to the $\alpha_E$ at nonresonant frequency, as shown in Fig. 4. However, $f_{\text{MER}}$ decreases with increasing $H_{\text{bias}}$ reaching the smallest value of 95.25 kHz at $H_{\text{bias}}=400$ Oe, and then increasing with increasing $H_{\text{bias}}$. Physically, the initial change in both $\alpha_E$ (at $f_{\text{MER}}$) and $f_{\text{MER}}$ with increasing $H_{\text{bias}}$ can be explained by the $H_{\text{bias}}$-induced motion of the available non-180\(^\circ\) domain walls in the Terfenol-D plates.\textsuperscript{13} That is, as $H_{\text{bias}}$ increases to the 400 Oe critical value, the compliance associated with increased deformation contribution from this non-180\(^\circ\) domain-wall motion is maximized, resulting in a maximum in strain (and hence $\alpha_E$ at nonresonant frequency and $\alpha_E$ at $f_{\text{MER}}$) and a minimum in stiffness (and hence $f_{\text{MER}}$). Beyond this optimal and also critical value of $H_{\text{bias}}$, constraining of non-180\(^\circ\) domain-wall motion due to interaction with $H_{\text{bias}}$ gives rise to a decrease in strain and an increase in stiffness. It is noted that the deformation contribution from the motion of 180\(^\circ\) domain walls is insignificant as it produces changes in magnetization without accompanying strain.\textsuperscript{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 $(001)^T \times (01\bar{1})^n \times (011)^T$ between two Terfenol-D plates enhances the elastolectric coupling in the magnet-to-electric field conversion, which results in a significantly increased ME voltage coefficient, approximately two times larger than the previous $L$-$T$ mode laminate com-
posite 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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