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Magnetic control of polarization and ferroelastic strain switching in Terfenol-D/Bi₁₂GeO₂₀ laminate

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A profound interplay between ferroelastic strain, polarization, and external magnetic field was observed in both bilayer and trilayer laminate Tb_{0.27–0.30}Dy_{0.73–0.70}Fe_{1.90–1.95} (Terfenol-D)/Bi₁₂GeO₂₀. The bilayer shows a much higher shear strain induced magnetoelectric effect than the trilayer. A series of experimental results related to the ferroelasticity in Bi₁₂GeO₂₀ such as a butterfly loop of the surface strain of Bi₁₂GeO₂₀ to magnetic field and a reproducible magnetic-field-induced strain switching were observed for the bilayer laminate. © 2009 American Institute of Physics. [DOI: 10.1063/1.3089571]

In the past few years there has been a revival of interest in multiferroic materials in which two or three of the electric, elastic, and magnetic orderings coexist.¹ Multiferroics, mainly focusing on the interaction between the ferromagnetic phase and ferroelectric phase, are considered now as promising materials for magnetic field probes, storage media, spintronic devices, sensors, and actuators,^{2–4} especially the magnetostrictive/piezoelectric laminate composites which show much larger magnetoelectric (ME) effect compared to most single phase and other multiphase ME materials due to the stress mediated product property between magnetostrictive effect and piezoelectric effect.^{5–12} However, even though the mediated elastic stress/strain plays a key role in ME effect, there seems to be no concrete research on the elastic strain although a stress mediated interface coupling parameter was introduced in some papers.^{13,14} In our previous study of a ferromagnetic-ferroelastic/ferroelectric Ni₂MnGa/Pb(Zr_xTi_{1-x})O₃ composite, a peak of ME coefficients (α) was observed at the temperature of martensitic transition of Ni₂MnGa alloy. It is due to an attached ferroelastic deformation induced by reorientation of the variants (or martensitic twins) of the martensitic phase in the Ni₂MnGa alloy under a magnetic field.¹⁵ In this work, we report that laminate composites of Tb_{0.27–0.30}Dy_{0.73–0.70}Fe_{1.90–1.95} (Terfenol-D)/Bi₁₂GeO₂₀ (BGO) exhibit profound interplay between electric polarization, ferroelastic deformation, and applied magnetic field at room temperature.

In the experiment, bilayer and trilayer laminates were synthesized by bonding slices of magnetostrictive Terfenol-D and (110) plane piezoelectric BGO single crystal through epoxy adhesive. Two sides of BGO crystals were coated with silver electrodes. The size of both Terfenol-D and BGO slices is about 15 × 4 × 1 mm³. Terfenol-D alloy was chosen as the magnetostrictive material for its giant magnetostrictive strain (~1200 ppm) with an expeditious response (~1 μs) at room temperature.¹⁶ It is (211) oriented along the length

direction (the easy magnetized direction). As to the piezoelectric phase, we tried to choose the lead-free material BGO single crystal. The BGO single crystal is a widely used multifunctional material due to its piezoelectric effect, electrooptic effect, and good ultrasonic properties. For ME measurement,^{11,15} the samples were subjected to a bias magnetic field H and an alternating current (ac) magnetic field δH . An electromagnet was used to provide $H \sim 0–8$ kOe and a Helmholtz coil was used to generate $\delta H = 1$ Oe. The magnetic fields were set along the length direction of the sample. ME voltage δV across BGO crystal was measured with an oscilloscope and a lock-in amplifier. Thus the ME voltage coefficient was obtained as $\alpha = \delta V / d \delta H$, where d is the thickness of BGO. In order to better understand the elastic coupling, a length deformation on the surface of BGO driven by the magnetostriction of Terfenol-D in the bilayer laminate was measured by a resistance strain gauge stucked to the bottom surface of BGO.

Figure 1 shows the magnetostriction of Terfenol-D as a function of applied magnetic field H . Like the other traditional magnetic materials, overlapped magnetostriction curves without loops were observed. The maximum magnetostrictive strain of Terfenol-D is 1100 ppm. An important observation in Fig. 1 is the always positive values of the magnetostriction, which means that the Terfenol-D can be only elongated under external magnetic field even under the

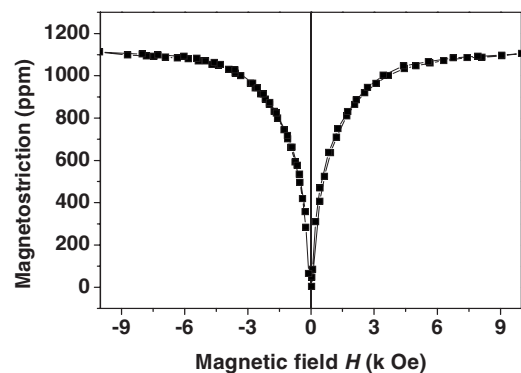


FIG. 1. Magnetostriction of Terfenol-D as a function of magnetic field.

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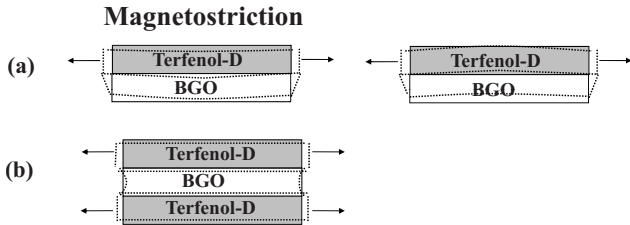


FIG. 2. Schematic drafts for the possible deformation of the laminate composites Terfenol-D/BGO. (a) Flexural deformation of the bilayer due to the discrepancy of Young's modulus between Terfenol-D and BGO; (b) trilayer deformation.

inversing direction magnetic field. Due to the magnetostriction, the Terfenol-D vibrates along the length direction under a bias magnetic field H and an ac magnetic field δH . It drives the BGO crystals to vibrate similarly. Consequently, the BGO generates an alternating electric voltage on both sides of the electrodes. Since the ME coupling occurs via the interlayer mechanical strain, paying much attention to the mechanical strain is important. The different mechanical deformations of the bilayer and trilayer laminates were assumed as shown in Fig. 2, where Fig. 2(a) shows the flexural deformation of the bilayer due to the different Young's modulus between Terfenol-D and BGO and Fig. 2(b) shows the deformation of BGO in the trilayer under the Terfenol-D magnetostriction drive. It is obvious from Fig. 2 that the BGO crystal in the bilayer laminate has a much stronger shear strain deformation than that in the trilayer laminate. Actually, BGO single crystal with [110] orientation is often used to give birth to thickness shear mode vibration in multifunctional devices and transducers.

BGO crystal is of cubic structure and point group 23. Its piezoelectric coefficient tensor is as follows:

$$\begin{pmatrix} 0 & 0 & 0 & d_{14} & 0 & 0 \\ 0 & 0 & 0 & 0 & d_{14} & 0 \\ 0 & 0 & 0 & 0 & 0 & d_{14} \end{pmatrix}.$$

The d_{14} term is 0.4×10^{-10} C/N. According to the piezoelectric equation, we can get the following results: $P_1 = d_{14}\sigma_4$, $P_2 = d_{14}\sigma_5$, and $P_3 = d_{14}\sigma_6$, in which P and σ represent the polarization and stress, respectively, i.e., the ME effect in Terfenol-D/BGO should be entirely caused by the shear stress (strain). It is very different from the relevant experiments and theoretical estimate in that the ME coefficient of this kind of bilayer and sandwich laminate structure is proportional to the piezoelectric term d_{31} or d_{33} usually.¹⁷⁻¹⁹ Experimental results (P_3) in this work exhibit that shear stress (strain) can also contribute to the ME coupling and d_{14} piezoelectric coefficients can also be considered to be used.^{20,21} Usually, the reported results of the ME coefficient in a bilayer are much less than that in a trilayer for the weak interface elastic coupling due to the glue bond; however, the ME coefficient α measured here in Terfenol-D/BGO laminates is different.

Figure 3 shows the dependence of the ME voltage coefficient α on the measuring frequency in bilayer Terfenol-D/BGO. The electromechanical resonance leads to a significant enhancement by a factor of more than 10 compared to the low frequency value in ME coupling in Fig. 3. The resonance frequency depends on the mechanical compliance, which will be affected by the symmetry of elastic strain, so the low

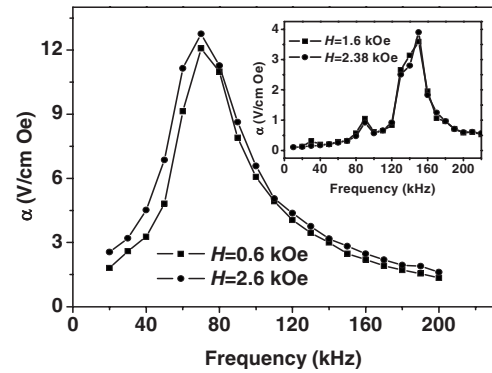


FIG. 3. Frequency dependence of ME coefficient for the bilayer Terfenol-D/BGO; the inset is for that of the trilayer laminate for comparison.

resonance frequency (around 70 kHz) in the bilayer compared with the one in the trilayer (about 150 kHz shown in the inset figure in Fig. 3) should be partly due to the flexural elastic coupling, which is in coincidence with the report by Zhang *et al.*²² As to the bilayer laminate, the largest ME coefficient α of 13 V/cm Oe was obtained, which is much larger than the value of 4 V/cm Oe obtained in the trilayer laminate (shown in the inset figure in Fig. 3). From Fig. 2, we can observe that the thickness vibration of BGO is depressed in the trilayer and the bilayer laminate has a much stronger shear strain deformation; it may be the origin for the weaker ME coefficient α measured in the trilayer laminate.

Like our previous work in Terfenol-D/0.52 Pb(Fe_{1/2}Nb_{1/2})O₃-0.48PbTiO₃ laminate composites,²¹ non-negligible ME coefficient under null dc magnetic field was observed (not shown here). The nonzero α under zero dc magnetic field probably comes from the spontaneous and remnant ferroelastic strain, and the ferroelasticity of BGO may be responsible for it. In order to confirm the existence of ferroelastic effect, the surface strain of BGO crystal was measured as a function of the applied bias magnetic field in bilayer Terfenol-D/BGO. As reported, under an applied magnetic field, overlapped positive magnetostriction curves without loops were observed for Terfenol-D in Fig. 1. While on the contrary, a different response of the strain of BGO to magnetic field (under driving of the Terfenol-D magnetostriction) describes the H -induced longitudinal strain on the surface of BGO crystal. The strain of BGO as a function of H was a butterfly loop, as shown in Fig. 4, which is quite different to the magnetostriction of Terfenol-D. Regarding the ferroelastic domain pair $\{S_1, S_2\}$, applied stress could

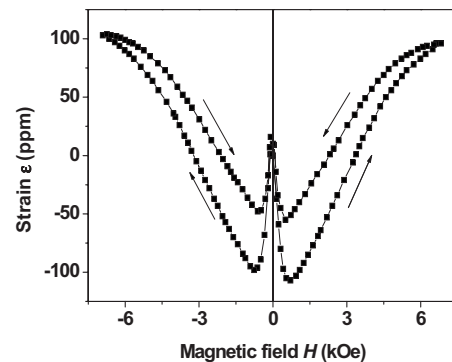


FIG. 4. The dependence of the surface strain of BGO crystal on external applied magnetic field.

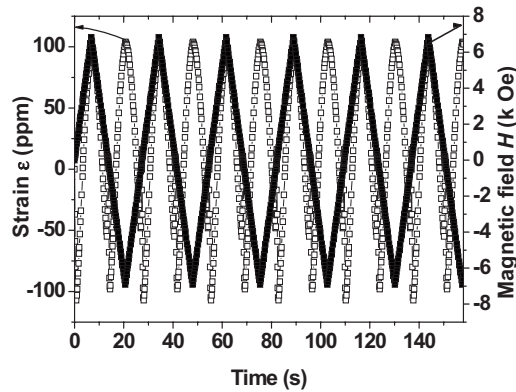


FIG. 5. Reproducible strain reversal controlled by magnetic field. It shows the strain switching by linearly varying the magnetic field. The solid dot curve corresponds to the magnitude of $H(t)$, while the hollow plane curve corresponds to the strain $\varepsilon(t)$ generated.

switch one ferroelastic domain state to another. So the variation of strain has a lag compared with applied stress and a ferroelastic hysteresis loop between the strain and stress will be observed. The strain of BGO crystal in Fig. 4 is in well agreement with its ferroelasticity (as predicted by Aizu²³) and the hysteresis and remnant strain will strongly influence the ME effect in Terfenol-D/BGO.

Curiously, the sign of the strain in BGO can be switched completely by increasing the applied magnetic field as shown in Fig. 5, which means the extension and contraction of BGO can be freely controlled by the external magnetic field. It is very different from the traditional magnetostriction and electrostriction materials. The strain sign reversal by applied magnetic field results in a startling reproducible “strain switching” shown in Fig. 5. The magnetic field was varied as $H(t) = H_0(t - nT)$, where t is the time, n is the cycle number, and T is the cycle period. The solid dot curve of Fig. 5 corresponds to the magnitude of $H(t)$, while the hollow plane curve corresponds to the strain $\varepsilon(t)$ generated. In this figure, the strain $\varepsilon(t)$ changes the sign periodically and leaves a small remnant strain ~ 10 ppm when $H(t) = 0$. It is in accordance with Fig. 4 and shows a highly reproducible magnetic-field-induced strain switching although the data were obtained from only a few of cycles. It points a new avenue to freely control the extension and contraction strain by a magnetic field and may give a possible application of the strain memory devices. Because the cubic structure of BGO can delete the influence of thermal expansion, the thickness shear vibration has small impedance and loss, which may be of great potential application.

In conclusion, the coupling between magnetic field, electrical polarization, and ferroelastic strain in bilayer and

trilayer Terfenol-D/BGO composites was investigated. Stronger magnetic-field-induced ME effect was observed in bilayer laminate than that in the trilayer due to the larger thickness shear deformation. Furthermore, a profound interplay between ferroelastic strain of BGO and the applied magnetic field was also observed, which was demonstrated by a butterfly loop of the strain to magnetic field and a highly reproducible magnetic-field-induced strain switching in the bilayer laminate.

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