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Bending response of terfenol-D/BaTiO₃ bilayer to external magnetic and electric fields

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Abstract

The interplay of ferroelectrics and ferromagnetics gives an additional freedom for device design. Based on thermodynamic theory, the bending response of a bilayer terfenol-D/BaTiO₃ system to external magnetic and electric fields was investigated. The vertical displacement induced by an asymmetry stress in the bilayer system can be controlled by the thicknesses of each layer, external electric field and magnetic field. A high relative static displacement of 55% can be obtained under a saturated magnetic field. The large bending response controlled by external electric and magnetic fields simultaneously may have potential applications for multifunctional devices.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

Advances in actuators, sensor networks and multifunctional devices are a challenge to materials with more flexibility and freedom for design [1, 2]. Multiferroic materials have gained much attention because of the interplay of ferroelectrics and ferromagnetics [3–5]. Although single phase multiferroic materials have been discovered with higher temperatures [6], the low magnetoelectric coupling effect still limits their application. An alternative route is to fabricate multiferroic materials. The laminate composites are very popular due to the easy fabrication and excellent properties, which may have potential applications in smart structures and multifunctional devices [7].

In this work, we theoretically investigated an intriguing bending actuator with a ferroelectric and ferromagnetic laminate structure. Actuators, as important integral components in a smart system, are extensively studied. Piezoelectric-based actuators such as THUNDER and

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RAINBOW exhibit enhanced displacement by using stress engineering [1]. Bending usually occurs due to the asymmetry stress, which in turn results in a vertical displacement. The asymmetric stress can be induced by the asymmetric configuration, asymmetric stress/strain, temperature gradient and content gradient [8]. To reach a large vertical displacement, the stress gradient should be large in the vertical direction. Recently, new structures were employed to enhance the displacement [9, 10] and a relative displacement of 200% related to the ferroelectric thickness was reached. However, traditional actuators usually use a single field (such as electric, magnetic, mechanic and thermal fields) to control the bending response. Using magnetostriction, controlled by the magnetic field, to give an asymmetric strain to the downside ferroelectric layer will give an additional freedom for device design. The new concepts will provide a more broad application.

2. Thermodynamic model

To induce large magnetostriction, we use the alloy terfenol-D as the source of the up-layer stress. Terfenol-D is widely



Figure 1. Schematic of the bending calculation model showing the asymmetric stresses and the direction of the external magnetic and electric fields.

used in transducers, actuators, sensors and multiferroic functional devices because of its low magneto-crystalline anisotropy, giant magnetostriction at a low magnetic field [11] and a little decrease in the magnetostriction under cyclic stress [12]. Recently, composites of terfenol-D were exploited to avoid high frequency limitation [13]. BaTiO₃, which is an environment-friendly material, is chosen as the bottom-layer ferroelectric material. By using efficient magnetostrictive stress mediated to the BaTiO₃ layer though the interface, the bending response of the system can be tuned conveniently.

We neglect the elastic effect of electrodes and epoxy in this model by assuming that the thicknesses of them are rather small compared with those of the BaTiO₃ and terfenol-D layers. As shown in figure 1, the BaTiO₃ thin film with the top and bottom SrRuO₃ electrodes [14] bonds with terfenol-D using hightemperature epoxy. The coordinate is located in the interface of the two layers. We considered a 3D model with in-plane symmetry. That is to say, the character is the same along the x and y directions, so we just consider the x direction. The lateral dimension is L. \bar{h} and h are the thicknesses of each layer. We assumed that the polarization P is along the z direction and the eigenstrain induced by P is $Q_{12}P^2$, in which Q_{12} is the electrostrictive coefficient. Terfenol-D, under the external magnetic field parallel to the film, will have a positive magnetostrictive strain, which is a nonlinear function of the external field if we neglect the external stress effect on the magnetostrictive properties [15]. Since the up-layer has a positive prestress while the down-layer has a negative eigenstrain, the system will bend to the direction as shown in figure 1.

Usually, ferromagnetic materials are saturated in room temperature. The stress effect on the spontaneous magnetization and the magnetostriction is small compared with the effect of domain rotation under an external magnetic field, especially for the polycrystalline terfenol-D. For simplicity, we neglect the small electrostriction induced stress effect on the magnetostriction of terfenol-D. The total free energy of the bilayer can be written as the summation of the free energy of the two layers [16]:

$$F = F_{\rm P}^{0} + F_{\rm M}^{0}(H) + \int_{-\bar{h}}^{h} \left[\frac{1}{2} A(T - T_{\rm c0}) P^{2} + \frac{1}{4} B P^{4} + \frac{1}{6} C P^{6} + \frac{1}{2} D \left(\frac{\mathrm{d}P}{\mathrm{d}z} \right)^{2} - E P(z) + f_{\rm ela} \right] \mathrm{d}z, \tag{1}$$

where $F_{\rm P}^0$ and $F_{\rm M}^0(H)$ are the initial reference states of the ferroelectric and ferromagnetic layers, respectively. $F_{\rm M}^0(H)$ is just the function of the external magnetic field. *A*, *B*, *C* and *D* are the expansion coefficients of the Landau free energy. *T* is the ambient temperature and $T_{\rm c0}$ is the Curie temperature of the bulk BaTiO₃. Neglecting the depolarization field, *E* is only the external electric field [17]. The last term $f_{\rm ela}$ is the elastic coupling energy density of the system and related to the external fields, thicknesses of each layer and the polarization of the ferroelectric layer.

The temporal evolution of the polarization field can be described by the time-dependent Ginzburg–Landau equation [16]:

$$\frac{-1}{M}\frac{\partial P}{\partial t} = \frac{\delta F}{\delta P} = AP + BP^3 + CP^5 - D\frac{\mathrm{d}^2 P}{\mathrm{d}z^2} - E + \frac{\delta f_{\mathrm{ela}}}{\delta P},$$
(2)

where M is the kinetic coefficient related to the domain wall mobility.

In the following, we use Hsueh's method [18] to calculate the coupling elastic energy density f_{ela} and its variation to the polarization. The terfenol-D and BaTiO₃ layers are considered as cubic elastic media with effective elastic constants $G = C_{11} + C_{12} - 2C_{12}^2/C_{11}$ and $\bar{G} = \bar{C}_{11} + \bar{C}_{12} - 2\bar{C}_{12}^2/\bar{C}_{11}$, respectively. C_{11}, C_{12} and $\bar{C}_{11}, \bar{C}_{22}$ are corresponding elastic moduli [9].

First, let the BaTiO₃ layer, which has the same length as the initial state of terfenol-D, extend to the terfenol-D layers under a magnetic field. The unconstrained and incompatible in-plane elastic strains in the ferroelectric layer will be $\varepsilon_{11} = \varepsilon_{22} = \varepsilon_H - Q_{12}P^2$. Second, relax both layers. If there is no slippage or fracture in the bonded bilayer, a uniform stress, corresponding to a uniform strain, will be imposed on each layer. The residual uniform stress c, related to the electrostriction, magnetostriction, elastic properties and thicknesses of each layer, can be calculated according to the zero force equilibrium function $\int_{-h_m}^0 \bar{G}c \, dz + \int_0^h G(c + \varepsilon_H - Q_{12}P^2) \, dz = 0$; thus, the uniform strain component is obtained as

$$\varepsilon = -Gh\left(\varepsilon_H - \frac{Q_{12}}{h}\int_0^h P^2 \,\mathrm{d}z\right)/(\bar{G}\bar{h} + Gh). \tag{3}$$

Third, with the development of the asymmetric stresses, bending occurs in the final state. The elastic strain depending on *z* can be written as $\varepsilon(z) = c - (z - t_b)/r$, for $-\bar{h} \le z \le +h$, in which $z = t_b$ is the location of the bending axis. *r* is the radius of curvature of the system. From the force equilibrium

$$\int_{-\bar{h}}^{0} \bar{G} \frac{z - t_{\rm b}}{r} \, \mathrm{d}z + \int_{0}^{h} G \frac{z - t_{\rm b}}{r} \, \mathrm{d}z = 0$$

and the torque equilibrium in the final state

$$\int_{-\bar{h}}^{0} \sigma_{s} z \, \mathrm{d}z + \int_{0}^{h} \sigma_{f} z \, \mathrm{d}z = \int_{-\bar{h}}^{0} \bar{G} \varepsilon(z) z \, \mathrm{d}z + \int_{0}^{h} G \bigg[\varepsilon(z) + \varepsilon_{H} - \frac{Q_{12}}{h} \int_{0}^{h} P^{2} \, \mathrm{d}z \bigg] z \, \mathrm{d}z = 0,$$

the location of the bending axis and the radius of curvature can be determined as

$$t_{\rm b} = \frac{Gh^2 - \bar{G}\bar{h}^2}{2(\bar{G}\bar{h} + Gh)},\tag{4}$$

$$r = \frac{\bar{G}\bar{h}^{2}(2\bar{h}+3t_{\rm b}) + Gh^{2}(2h-3t_{\rm b})}{3\left[Gh^{2}\left(c+\varepsilon_{H}-\frac{Q_{12}}{h}\int_{0}^{h}P^{2}\,{\rm d}z\right) - \bar{G}c\bar{h}^{2}\right]}.$$
(5)

Finally, we can find the total elastic energy density f_{ela} . The variation of f_{ela} with respect to spontaneous polarization is

$$\delta f_{\rm ela} / \delta P = -4Q_{12}G[c - (z - t_{\rm b})/r + \varepsilon_H]P + 4GQ_{12}P^3.$$
(6)

Substituting $\delta f_{\rm ela}/\delta P$ in equation (2), we can rewrite the equation as

$$\frac{-1}{M}\frac{\partial P}{\partial t} = \frac{\delta F}{\delta P} = A^*P + B^*P^3 + CP^5 - D\frac{\mathrm{d}^2 P}{\mathrm{d}z^2} - E,\qquad(7)$$

where the renormalized coefficients are

$$A^* = A(T - T_{c0}) - 4GQ_{12}[c - (z - t_b)/r + \varepsilon_H],$$

$$B^* = B + 4GQ_{12}^2,$$
(8)

The static and steady-state polarization can be solved by letting the right side of equation (7) to be zero. The average polarization $\langle P \rangle = \int_0^h P \, dz/h$, the uniform strain component *c* and the position of the bending axis t_b can also be obtained.

According to figure 1, the static bending displacement is given by

$$d(H) = r(H) - \sqrt{r(H)^2 - (L/2)^2}.$$
 (9)

As shown in equation (9), the vertical displacement d is increased with the increase in the lateral length L. For application, we are more concerned with the relative displacement. We define two static relative displacements ΔD_t and ΔD_H related to the thickness and the external field, respectively:

$$\Delta D_t = \frac{d(H)}{h + \bar{h}},$$

$$\Delta D_H = \frac{d(H) - d(H = 0)}{d(H = 0)}.$$
(10)

The dynamic d with respect to E can be obtained by applying a cyclic sinusoidal external field $E = E_0 \sin(2\pi t/T') = E_0 \sin(2\pi t f')$ along the z direction, where E_0 , T' and f' are the amplitude, period and frequency, respectively.

3. Results and discussions

Using the material constants taken from [4, 9], we can solve the polarization and the vertical displacement at a given lateral dimension and thicknesses of each layer. As an example, we took the thicknesses of both the BaTiO₃ and terfenol-D layers to be 0.1 mm and the lateral dimension *L* to be 50*h*.



Figure 2. Static relative displacements of the bilayer versus the thickness of the terfenol-D layer with different thicknesses of the BaTiO₃ layer. The inset shows the static relative displacements of the bilayer versus the thickness of the terfenol-D layer.

As we know, the lateral dimension, the thicknesses of each layer and their relative ratio are key factors to the bending response [9]. Since the moduli of the two materials are similar $(G = 1.79 \times 10^{11}, \bar{G} = 1.46 \times 10^{11})$, we can approximate and simplify equations (3)–(5) by letting $G \doteq \overline{G}$. Substituting the simplified equations (3) and (4) to equation (5), we can obtain $\bar{r} = (h + \bar{h})^3 / [6\bar{h}h(\varepsilon_H - Q_{12}\langle P^2 \rangle)]$. It is clear that r is larger than $4\sqrt{h\bar{h}}/[3(\varepsilon_H - Q_{12}\langle P^2 \rangle)]$ and only equal at $\bar{h} = h$. If the total thickness of the two layers is a constant, there exists the minimum value of r, and we can obtain the smallest radius curvature corresponding to the largest vertical displacement. We note that this inequation is just under the assumption $G \doteq G$. If the effective elastic constants are not equal to each other, a careful calculation is needed to find the smallest r and the corresponding thickness ratio. The relationship of the displacement and the thicknesses of the terfenol-D layer under the magnetic field of 200 KOe can be found in figure 2. In this example, the vertical displacement d increases with increasing thickness of the BaTiO₃ layer at a given thickness of the terfenol-D layer. The maximum d can also be simulated. According to figure 2, the best thickness ratio is about, which is consistent with the above simplified calculation. The relative displacement ΔD_t changes little with the thickness of the BaTiO₃ layer and also reaches a maximum at $\bar{h} = h$.

The two relative displacements with respect to the total thickness and the one without magnetic field are calculated by using equation (10). The thickness of each layer is 0.1 mm and the lateral dimension L is 5 mm. The results are given in figure 3. As defined in equation (10), ΔD_t is the relative displacement related to the thickness of the film. Since the films bond at a high temperature above the Curie temperature of BaTiO₃, the phase transition will take place when the temperature was cooled down to the room temperature. The corresponding spontaneous strain will make the system bend even in the absence of the magnetic field because of the asymmetric stress state. The initial relative



Figure 3. Static relative displacements of the bilayer versus the magnetic field. The inset shows the static vertical displacement versus the magnetic field.



Figure 4. Dynamics of the displacement of the bilayer subjected to a sinusoidal applied electric field under an external bias magnetic field.

displacement ΔD_t , which can also be called a prestress induced bending displacement, is about 35%. As described above, the value will be different with the given different thickness parameters. The relative displacement related to the magnetic field ΔD_H is also shown in figure 3. The magnetic field induced relative displacement can be as high as 55%, which is higher than the graded layers and the FE/PE layer ones with the same configuration. Since the slope of each curve of ΔD_t is similar to each other, so the ΔD_H is similar even at a different ratio of the thickness of the two layers.

The dynamic response under combined external electric and magnetic fields is given in figure 4. As we applied a sinusoidal electric field, the spontaneous electrostriction is a butterfly loop under this electric field. The vertical displacement, which is almost linear with the electrostriction, is also a butterfly loop. As shown in figure 4, the displacement can reach to 160% of the ferroelectric thickness and reduce to as low as 0.3h. The high drop height is about 0.13 mm, 65% of the total layer thickness. We should note that, if the magnetic field is very small, and the magnetostriction will be not be big enough to make the bend always towards to one direction, the loop will be bizarre under the sinusoidal electric field.

4. Conclusions

The bending response of a terfenol-D/BaTiO₃ bilayer system on the external magnetic and electric fields is studied using an elastic and thermodynamic model. The bending responses due to the inhomogeneous stress induced by the magnetic field in the bilayer can produce large vertical displacements. The elastic and thermodynamic model developed in this paper is applicable to many other multilayer systems.

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