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RESEARCH ARTICLE

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Influence of the stress on magnetoelectric effect in magnetostrictive-PZT bilayers

Abstract In this letter, we investigate the influence of the stress on magnetoelectric (ME) effect in a magnetostrictive-PZT bilayer. ME voltage coefficient $\alpha_E^* = \frac{\delta E}{\delta H}$, where δE is the induced electric field for an applied alternating current (ac) magnetic field δH, is obtained by solving the stress-related piezoelectric constitutive equation and the conventional magnetostrictive equation with appropriate boundary condition. Based on the free-energy density function of the PZT film in stress state, we get the stress-related piezoelectric charge coefficient ${}^{\mathrm{p}}d_{31}^*$ and dielectric permittivity ${}^{\mathrm{p}}\varepsilon_{33}^*$. After taking the cobalt ferrite (CFO) as magnetostrictive phase, it is found that α_E^* increases with decreasing 2-d compressive stress for CFO-PZT, which not only is qualitatively consistent with previous experimental measurements, but also provides a possible route to improve the ME effect.

Keywords piezoelectric, magnetoelectric effect, magnetostrictive

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The magnetoelectric (ME) effect is defined as the dielectric polarization of a material in an applied magnetic field or an induced magnetization of an external electric field [1, 2], and it plays an important role in such devices as ME memory elements, transducers and smart sensors [3]. It is well known that there are two major sources for accomplishing this effect: (i) In single-phase multiferroics, the internal electric fields are enhanced by the presence of multiple long-range ordering, but there are few such single-phase materials and the effect is often weak; (ii) In product-property composites of magnetostrictive and piezoelectric materials, a weak alternating current (ac) magnetic field δH oscillating in the presence of a strong direct current (dc) bias field produces mechanical deformation resulting in a spin-electric dipole coupling and an ac electric field δE [4–6], where the ME voltage coefficient is defined as $\alpha_E = \frac{\delta E}{\delta H}$. Moreover, the lead zirconate titanate (PZT) was often taken as the piezoelectric phase because of its favorable piezoelectric capability. Therefore, most researches focus on these magnetostrictive-PZT composites. Compared with the single-phase multiferroic, the product-property composite could not only get strong ME effect, but also control the degree of ME effect via changing structural details of each phase such as the volume fraction, microstructure, composition, strain, etc.

Since Tellegen formulated the idea of synthesizing a product-property composite displaying ME effect in 1948 [7], the experimental studies of ME effects have received considerable attention in the succeeding years. Particulate composites were first used to research the ME effects, but they yielded α_E that were two to three orders-of-magnitude smaller than theoretical predictions. It has been found that such low values are primarily due to low resistivity for ferrites, which produces a leakage current that results in the loss of induced voltage [8]. These obstacles were overcome by using layered composite instead of particulate composite [9]. With the laminated Terfenol-D/PZT layered composite, Ryu *et al.* immediately acquired an ME voltage coefficient of up to 4.68 $\mathrm{V\cdot cm^{-1}\cdot Oe^{-1}}$, which exceeded the highest value gained from any particulate composites at that time by a fac-

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With great improvement having been achieved in the experimental field, many theoretical approaches were also used to investigate the ME coupling in product-property layered composite. The first prediction of large ME effects in layered composite was made by Harshe *et al.* [10], who calculated the longitudinal ME response of ferrite-PZT layered composites. Then, a more general way to the ME effects in layered composite was presented by Bichurin *et al.* [11]. They gained achievements for the consideration of the demagnetization factor and the introduction of a coupling parameter k, which was used to define the degree to which the deformation of the piezoelectric layer follows that of the magnetostrictive layer. By adjusting the suitable value of k, the theoretical model can explain some experimental results. However, it should be noticed that although the coupling parameter k was used to consider the actual interface condition, the physical meaning of coupling parameter k was still not very clear. To our best knowledge, detailed studies on the influence of the interface coupling on ME effect is lacking at present, and a substantial disagreement between theoretical and experimental data was observed.

In addition to ambiguous details of interface coupling between PZT layer and magnetostrictive layer, the major discrepancy between theoretical and experimental results also comes from the fact that all the thermodynamic coefficients of PZT are measured on bulk crystal, not film. In particular, the piezoelectric and dielectric properties of PZT film may differ considerably from bulk. Furthermore, even if the film may be taken as "bulk" film, the stress state upon cooling through the Curie temperature can influence the piezoelectric and dielectric properties, in tetragonal PZT film. Therefore, another important problem is the influence of the stress built in film on ME effect. It is well known that there are several sources that contribute to the residual biaxial stresses built up in the epitaxial film. The most relevant ones are the intrinsic stress σ_{int} and the thermal stress σ_{th}. The magnitude of the intrinsic stress is largely determined by the film deposition conditions, and the thermal stress dominates during the process of cooling from the annealing temperature, which originates from the temperature-dependent lattice mismatch between the layers. At the same time, strain relaxation can occur through the misfit dislocation formation and twinning. Therefore, the previous works [12, 13] assume that the residual strain after the film growth process is close to zero. In fact, the strain can only be partially relaxed even if the films of each phase are thick enough. Aside from the intrinsic and thermal stress in PZT thick film, interfacial stresses may also be present in layered composite, which will contribute significantly to the total film stress. Thus, the influence of film stress, consisting of the interfacial stress and other stress resulting in residual biaxial stress in the films, on the ME effect is worth noticing in layered composite.

In this paper, we will investigate the influence of the stress on magnetoelectric (ME) effect in a magnetostrictive-PZT bilayer. A bilayer of stress state in the (1,2) plane consisting of PZT and magnetostrictive phases is shown in Fig.1. In this product-property structure, in order to get an effective ME coefficient in experiment, the magnetostrictive phase may be taken as the substrate, and as the bottom electrode in the device. This combination has many advantages as a model system. Without the constraint from the conventional substrate, such a geometry maximizes the efficiency of in-plane transfer of the magnetostrictive strain from magnetostrictive layer to piezoelectric layer. As we know, the stress not only affect the magnetic order temperature and the easy direction of magnetization in magnetostrictive phase, but also influence ferroelectric Curie temperature and thermodynamic properties in PZT phase. Because the mechanical strain-mediated ME coupling by magnetostrictive effect is a quasi-piezomagnetic effect, the influence of stress on the PZT phase is greater than that on the magnetostrictive phase. Therefore, for simplicity, here we only take the stress in the PZT layer into account. First, we briefly review a theoretical model valid for the above bilayer [10, 11]. The piezoelectric consititutive equations are

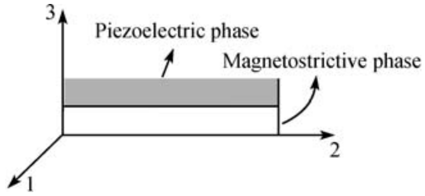

Fig. 1 Schematic diagram showing a bilayer of magnetostrictive and piezoelectric phases in the (1, 2) plane.

$$S_i^{\mathrm{p}} = {}^{\mathrm{p}}s_{ij} T_j^{\mathrm{p}} + {}^{\mathrm{p}}d_{ki}^{*} E_k^{\mathrm{p}} \qquad (1)$$

$$D_k^{\mathrm{p}} = {}^{\mathrm{p}}d_{ki}^{*} T_i^{\mathrm{p}} + {}^{\mathrm{p}}\varepsilon_{kn}^{*} E_n^{\mathrm{p}}$$

where T_j^{p} and S_i^{p} are stress and strain tensor components of the piezoelectric phase; D_k^{p} and E_k^{p} are the electric displacement and field; ${}^{\mathrm{p}}s_{ij}$ is compliance coefficients at constant electric field; ${}^{\mathrm{p}}d_{ki}^{*}$ and ${}^{\mathrm{p}}\varepsilon_{kn}^{*}$ are the piezoelectric charge coefficients and the dielectric permittivity matrix; the superscript "p" denotes the components for the piezoelectric layer. To consider the influence of stress, the superscript "*" is used to denote the coefficients for PZT thick film in biaxial stress state in order to differentiate it from those of stress-free bulk film. The magnetostrictive phase is assumed to have a cubic symmetry and is described by the following conventional equations:

$$S_i^{\mathrm{m}} ={}^{\mathrm{m}} s_{ij}T_j^{\mathrm{m}} +{}^{\mathrm{m}} q_{ki}H_k^{\mathrm{m}}$$

$$B_k^{\mathrm{m}} ={}^{\mathrm{m}} q_{ki}T_i^{\mathrm{m}} +{}^{\mathrm{m}} \mu_{kn}H_n^{\mathrm{m}} \tag{2}$$

where S_i^{m} and T_j^{m} are strain and stress tensor components of the magnetostrictive phase; H_k^{m} and B_k^{m} are the vector components of the magnetic field and magnetic induction, ${}^{\mathrm{m}}s_{ij}$ and ${}^{\mathrm{m}}q_{ki}$ are compliance and piezomagnetic coefficients; and ${}^{\mathrm{m}}\mu_{kn}$ is the permeability matrix, the superscript “m” denotes the components for the magnetostrictive layer.

We assume that the ME layered composite is unclamped, and the interface coupling is ideal. Thus, the ME voltage coefficients can be obtained by solving Eq.(1) and Eq.(2) with the following boundary conditions:

(i) The strains in both the phases along the transverse direction 1 and 2 are equal:

$$S_1^{\mathrm{p}} = S_1^{\mathrm{m}}$$

$$S_2^{\mathrm{p}} = S_2^{\mathrm{m}} \tag{3}$$

(ii) The total stresses are zero along the 1 and 2 direction, and no stress is present along the 3 direction

$$vT_1^{\mathrm{p}} + (1-v)T_1^{\mathrm{m}} = 0$$

$$vT_2^{\mathrm{p}} + (1-v)T_2^{\mathrm{m}} = 0$$

$$T_3^{\mathrm{p}} = T_3^{\mathrm{m}} = 0 \tag{4}$$

where ${}^{\mathrm{m}}v$ and ${}^{\mathrm{p}}v$ respectively denote the volume of magnetostrictive phase and piezoelectric phase, and $v = \dfrac{{}^{\mathrm{p}}v}{{}^{\mathrm{p}}v +{}^{\mathrm{m}} v}$ is defined as the volume fraction of ferroelectric composite.

Combining the continuity conditions for magnetic and electric fields and open-circuit conditions, detailed calculations will yield the following expressions for ME coefficients in the two different field orientations of importance: (i) $\alpha^*_{E,33}$ where the bias field H, δH, and δE are all parallel to each other and perpendicular to the sample plane (1,2); (ii) $\alpha^*_{E,31}$ where in-plane H and δH are parallel to each other and perpendicular to the out-of-plane δE:

Transverse ME effect:

$$\begin{aligned}\alpha^*_{E,31} =\, & v(1-v)({}^{\mathrm{m}}q_{11} +{}^{\mathrm{m}} q_{21})({}^{\mathrm{p}}d^*_{31})/\\ & ({}^{\mathrm{p}}\varepsilon^*_{33})({}^{\mathrm{m}}s_{11} +{}^{\mathrm{m}} s_{12})v + ({}^{\mathrm{p}}\varepsilon^*_{33})({}^{\mathrm{p}}s_{11}\\ & +{}^{\mathrm{p}}s_{12})(1-v) - 2({}^{\mathrm{p}}d^*_{31})^2(1-v)\end{aligned} \tag{5}$$

Longitudinal ME effect:

$$\begin{aligned}\alpha^*_{E,33} =\, & 2\mu_0 v(1-v)({}^{\mathrm{p}}d^*_{31})({}^{m}q_{31})/\\ & [\mu_0(v-1) -{}^{\mathrm{m}} \mu_{33}v][v({}^{\mathrm{m}}s_{11} +{}^{\mathrm{m}} s_{12})({}^{\mathrm{p}}\varepsilon^*_{33})\\ & +({}^{\mathrm{p}}s_{11} +{}^{\mathrm{p}} s_{12})({}^{\mathrm{p}}\varepsilon^*_{33})(1-v) - 2({}^{\mathrm{p}}d^*_{31})^2(1-v)]\end{aligned} \tag{6}$$

From the above expressions we find that ME effect is strongly related with not only the volume fraction of PZT, but also the intrinsic properties of each phase, especially the piezoelectric and dielectric properties of piezoelectric phase. As we mentioned previously, the piezoelectric and dielectric properties of the piezoelectric film in the stress state are stress-dependent. Now, we further consider the effect of stress on the piezoelectric and dielectric properties of the PZT thick film.

According to the Landau-Ginsburg-Devonshire formalism, the free-energy density function derived from the power-series expansion in terms of polarization components P_i and stress components T_j can be expressed as follows [14]:

$$\begin{aligned}G_1 =\, & \alpha_1(P_1^2 + P_2^2 + P_3^2) + \alpha_{11}(P_1^4 + P_2^4 + P_3^4)\\ & +\alpha_{12}(P_1^2P_2^2 + P_2^2P_3^2 + P_3^2P_1^2)\\ & +\alpha_{111}(P_1^6 + P_2^6 + P_3^6) + \alpha_{112}[P_1^4(P_2^2 + P_3^2)\\ & +P_2^4(P_1^2 + P_3^2) + P_3^4(P_1^2 + P_2^2)]\\ & +\alpha_{123}(P_1^2P_2^2P_3^2) - \frac{1}{2}s_{11}(T_1^2 + T_2^2 + T_3^2)\\ & -s_{12}(T_1T_2 + T_2T_3 + T_3T_1)\\ & -\frac{1}{2}s_{44}(T_4^2 + T_5^2 + T_6^2)\\ & -Q_{11}(T_1P_1^2 + T_2P_2^2 + T_3P_3^2) - Q_{12}[T_1(P_2^2 + P_3^2)\\ & -T_2(P_1^2 + P_3^2) + T_3(P_1^2 + P_2^2)]\\ & -Q_{44}(T_4P_2P_3 + T_5P_1P_3 + T_6P_1P_2)\end{aligned} \tag{7}$$

where P_i and T_i are the polarization and stress, α_i, α_{ij}, and α_{ijk} are the dielectric stiffness and higher-order stiffness coefficient at constant stress; s_{ij} is the elastic compliance coefficient at constant polarization; Q_{ij} is the cubic electrostrictive constant in polarization notation. Eq.(7) has two interesting solutions. One is for the prototypic cubic state($Pm3m$), another for the ferroelectric tetragonal state($P4mm$). It has already been identified that for PZT the piezoelectric charge coefficient d^*_{ki} is higher in its ferroelectric state than in its paraelectric state. Therefore, both theoretical and experimental works focus on the ferroelectric state of a film, especially the tetragonal state. The piezoelectric charge coefficient d^*_{ki}, piezoelectric voltage coefficient g^*_{ni} and dielectric permittivity ε^*_{kn} obey the following relationship:

$$d^*_{ki} = \varepsilon^*_{kn}g^*_{ni} \tag{8}$$

If the layered composite in 2-d stress state is unclamped, we have $T_1 = T_2 = h$, $T_3 = T_4 = T_5 = T_6 = 0$. The appropriate sign convention is adopted such that tensile stress is denoted by positive value of h, vice versa. Meanwhile the PZT film is assumed in tetragonal ($P4mm$) state, $P_1^2 = P_2^2 = 0$, $P_3^2 \neq 0$. The second partial derivative of Eq.(7) may give piezoelectric voltage coefficient g^*_{ni} and dielectric permittivity ε^*_{kn}[15]. Here we give the concrete expressions of g^*_{31} and ε^*_{33} since only these two coefficients will be useful in the following calculation:

$$g^*_{31} = 2Q_{12}P_3 \tag{9}$$

$$\varepsilon^*_{33} = \varepsilon_0 + [2(\alpha_1 - 2Q_{12}h + 6\alpha_{11}P_3^2 + 15\alpha_{111}P_3^4)]^{-1} \tag{10}$$

where ε_0 is the permittivity of the free space, P_3 is determined from the first partial derivative stability condition:

$$\partial G_1/\partial P_3 = 0$$

$$P_3^2 = -\alpha_{11} + [\alpha_{11}^2 - 3\alpha_{111}(\alpha_1 - 2Q_{12}h)]^{1/2}/3\alpha_{111} \tag{11}$$

Up to now, we know only that the dielectric stiffness α_1 is renormalized to $\alpha_1 - 2Q_{12}h$ due to the stress and is a linear function of the stress h. The normalizing of the dielectric stiffness results in the shift in the Curie temperature $\Delta T = 4\varepsilon_0 CQ_{12}h$ and the stress-dependence of the piezoelectric charge coefficient d^*_{ki}, piezoelectric voltage coefficient g^*_{ni} and dielectric permittivity ε^*_{kn}. They will further cause the stress-dependence of ME effect.

In the following discussions, we apply material parameters given in Table 1 to discuss a bilayer of cobalt ferrite (CFO) and lead zirconate titanate (PZT). Since the strength of α^*_E depends sensitively on the concentration ratio of the two phases, we will first consider the transverse and longitudinal voltage coefficients as a function of the volume fraction v of PZT film in CFO-PZT. After substituting Eq.(8)–Eq.(11) into Eq.(5) and Eq.(6), the variations of transverse and longitudinal ME voltage coefficients $\alpha^*_{E,31}$, $\alpha^*_{E,33}$ with volume fraction v for a series of h are shown in Fig.2 and Fig.3. Of course, when the stress is set to be zero, the variations of transverse and longitudinal ME voltage coefficients with volume fraction v recover to the previous theoretical results [11]. At the same time, from Fig.2 and Fig.3 we also find that for a certain stress h, the ME coupling is absent in pure PZT ($v = 1$) or CFO ($v = 0$), and as v is increased from 0 to 1, $\alpha^*_{E,31}$ and $\alpha^*_{E,33}$ first increase to a maximum, then decrease gradually to 0. The stress-dependent behavior of ME effect is similar to the stress-free one.

Table 1 The material parameters of CFO and PZT used to calculate ME coupling (from Refs. [11, 16]).

	PZT	CFO
$a_1/(10^7\mathrm{m}\cdot\mathrm{F}^{-1})$	-2.669	
$a_{11}/(10^7\mathrm{m}^5\cdot\mathrm{C}^{-2}\cdot\mathrm{F}^{-1})$	-1.431	
$a_{111}/(10^9\mathrm{m}^9\cdot\mathrm{C}^{-4}\cdot\mathrm{F}^{-1})$	1.342	
$s_{11}/(10^{-12}\mathrm{m}^2\cdot\mathrm{N}^{-1})$	15.3	6.5
$s_{12}/(10^{-12}\mathrm{m}^2\cdot\mathrm{N}^{-1})$	-5	-2.4
$q_{33}/(10^{-12}\mathrm{m}\cdot\mathrm{A}^{-1})$		-1880
$q_{21}/(10^{-12}\mathrm{m}\cdot\mathrm{A}^{-1})$		556
$q_{11}/(10^{-12}\mathrm{m}\cdot\mathrm{A}^{-1})$		-1880
U_{33}/U_0		2
$e_0/(10^{-12}\mathrm{F}\cdot\mathrm{m}^{-1})$		8.85

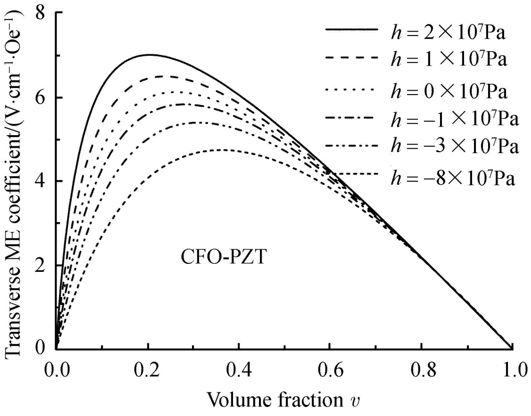

Fig. 2 Estimated dependence of transverse ME voltage coefficients on stress h and volume fraction v for the CFO-PZT bilayer.

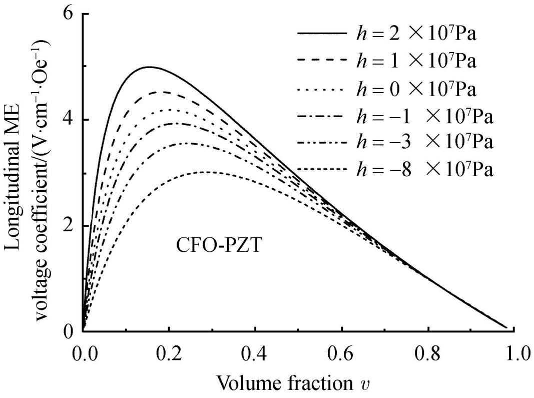

Fig. 3 Estimated dependence of longitudinal ME voltage coefficients on stress h and volume fraction v for the CFO-PZT bilayer.

On the other hand, it is more important to examine the influence of stress on ME effect. Figure 2 and Fig.3 show that $\alpha^*_{E,31}$ decreases as h changes from positive (tensile stress) to negative (compressive stress), and the positions of the peak values shift to PZT-rich compositions. Similar characteristics are observed in the curves of $\alpha^*_{E,33} \sim h$. In the region of $h < 0$, compared with the experimental research conducted by Srinivasan in the layer of cobalt zinc ferrite-lead zirconate titanate(CZFO-PZT) [17], our numerical results are qualitatively consistent with theirs. As has been found in their experimental measurements, the stress existing in PZT thick film can influence the composite's ME effect significantly. The XRD data show that ferrite is free of strain, but the PZT layer is strained in heterostructures of CZFO-PZT (compressive strain). In addition, the strain decreases with increasing

Zn substitution, accompanied by an improvement of the ME effect. They attribute the enhancement of ME effect to the increase of interface bonding of the substitution of Zn. In their theory, α_E values are for the bulk values of the material parameters rather than utilizing those of films in stress states, although they reinforce the need for the determination of material parameters for the constituent phases in the layered composites. A parameter k is introduced in order to describe the interface bonding influenced by a variety of factors. Unfortunately, they could only speculate on possible reasons, including built-in stress in the PZT layer, for the unique and favorable interface bonding in the system. Here we point clearly that decreasing compressive stress may cause the effective improvement of the ME effect, which corresponds to increasing Zn substitution in their experiments. Naturally, we extend our theory to the region of $h > 0$ (tensile stress). Figure 2 and Fig.3 show that the tensile stress may increase ME effect if the PZT film is still in the out-of-plane polar in the presence of suitable tensile stress, which provides us a possible route to improve the ME effect. We hope further investigations with this situation will be helpful to support this suggestion.

The stress dependence of ME effect, with respect to the stress-free bulk materials, can easily be understood by considering the change in nature of PZT layers in the stress state. The stress dependence of ME effect originates from the stress dependence of PZT parameters in stress state. The magnetostrictive phase is considered as the substrate and the influence of 2-d stress on the PZT phase is greater than that on the magnetostrictive phase, that is, ${}^{\mathrm{m}}q_{11}$, ${}^{\mathrm{m}}q_{21}$ and ${}^{\mathrm{m}}\mu_{33}$ may be considered approximately as constants. Then, from Eq.(5) and Eq.(6), we find that all parameters in expressions are independent of stress h except for d_{31}^* and ε_{33}^*. Consequently, the main influence of h on ME effect comes from the influence of h on d_{31}^* and ε_{33}^*. Therefore, we need to make clear the concrete influence of h on the two coefficients. According to Eq.(8)–Eq.(11), the dielectric permittivity ε_{33}^* and the piezoelectric charge coefficient d_{31}^* as a function of h are calculated and displayed in Fig.4 for PZT. It is shown that both their amplitudes will increase with increasing tensile stress or decreasing compressive stress. That is to say, through ε_{33}^* and d_{31}^*, the 2-d stress h has a great effect during the mechanical deformation resulting in induced electric polarization. Although the increase of the dielectric coefficient hampers ME effect, the increase of the piezoelectric coefficient enhances ME effect. Finally, with increasing tensile stress or decreasing compressive stress, the competition between piezoelectric charge coefficient and dielectric permittivity causes the increase of ME effect.

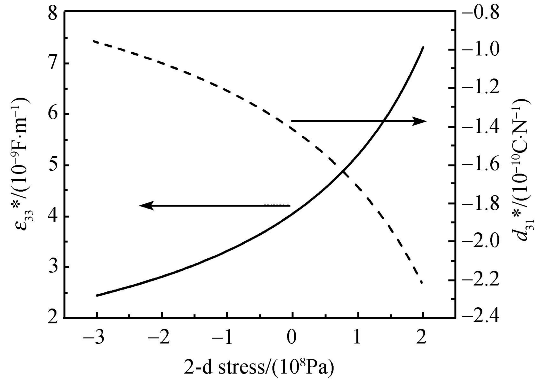

Fig. 4 The dielectric permittivity ε_{33}^* and the piezoelectric charge coefficient d_{31}^* as a function of stress h for PZT.

In addition, in order to have a better understand for the 2-d stress dependence of ME effect, in Fig.5 and Fig.6 we plot the maximum $\alpha_{E,31}^*$, $\alpha_{E,33}^*$ and corresponding PZT volume fraction $v_{\max}$ as functions of stress h. As shown in Fig.2 and Fig.3, with the increase of tensile stress or decrease of compressive stress, $\alpha_{E,31}^*$, $\alpha_{E,33}^*$ and $v_{\max}$ decrease gradually. All these show that the stress in the film play a major role in determining the ME coupling strength. In designing a magnetostrictive-PZT bilayer, it is crucial to find an optimum stress/strain in the film besides choosing the reasonable concentration of PZT film in order to obtain the greater ME coupling strength.

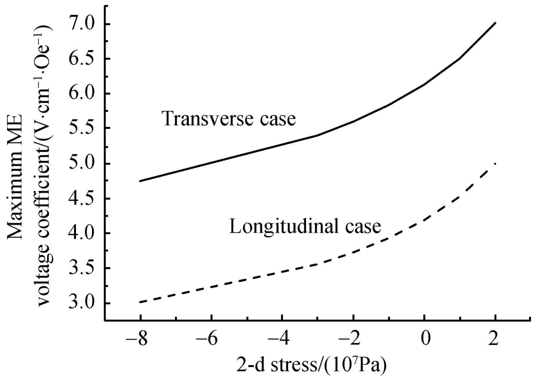

Fig. 5 Maximum ME voltage coefficients as functions of stress h.

In summary, the effect of biaxial stress in PZT film on the ME voltage coefficients for ME layered composite has been studied, in which the magnetostrictive phase is stress-free and PZT layer is stressed. The ME voltage coefficients expression are achieved by solving the stress-related piezoelectric constitutive function and conventional magnetostrictive equations with the help of appropriate boundary conditions. As far as CFO-PZT is concerned, it has been demonstrated that the ME voltage coefficients will increase with decreasing compressive stress, which is qualitatively consistent with previous experimental measurements.

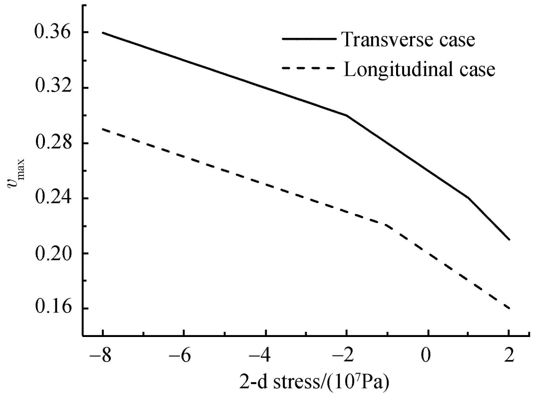

Fig. 6 The variation of $v_{\max}$ which is the PZT volume fraction corresponding to the maximum ME voltage coefficient with stress h.

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