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Simulation of substrate clamping on the properties of piezoelectric/piezomagnetic laminated composites

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Abstract

A theoretical model was established to calculate the magnetoelectric voltage coefficient of piezoelectric (PMN-PT)/piezomagnetic (CFO) laminated composites on Si substrate. Using this model, the transverse and longitudinal magnetoelectric voltage coefficient formula of laminated composites was derived in ideal interface coupling state simply. The magnetoelectric voltage coefficient (α_E) is calculated as function of the volume fraction of piezoelectrics and the thickness of Si substrate. It was found that the magnetoelectric voltage coefficient of PMN-PT/CFO laminated composites decreases with increase of Si substrate thickness, which shows strong clamping effect of the substrate. Meanwhile, with increase of substrate thickness, the compressive stress of substrate to PMN-PT enhances, which change the lattice constant of PMN-PT and lead to low optimum PMN-PT volume fraction *v*.

Keywords: B. Laminated composites; C. Magnetoelectric; C. Clamping; E. Substrate

1. Introduction

In recently years, multiferroic magnetoelectric (ME) materials, which simultaneously exhibit ferroelectricity and ferromagnetism, have drawn increasing interest due to their potential for applications as multifunctional devices, such as magnetic sensors, transformers, gyrators, microwave devices and so on [1–3].

Usually, the natural multiferroic single-phase compounds are rare, and their magnetoelectric responses are either relatively weak or occurs at temperatures too low for practical applications. In contrast, multiferroic composites, which incorporate both ferroelectric and ferromagnetic phases [4–6], typically yield giant magnetoelectric coupling response above room temperature, which makes them ready for technological applications [6–8]. The strongest ME coupling was expected in a layered structure due to the absence of leakage current and ease of poling to align the electric dipoles. Harshe [4] and Nan [9], respectively, proposed the simplified approximation and rigorous method to calculate the effective ME properties of piezoelectric/ferrite ceramic composites. Since then, many researchers have investigated the ME coupling behavior in the piezoelectric/ferrite ceramic composites both in the experimental [10–13] and theoretical [14–16] fields.

The laminated magnetoelectric composites were fabricated a lot for their low leakage current and strength of electric dipoles. $(1-x)Pb(Mg_{1/3}Nb_{2/3})O_3 - xPbTiO_3$ (PMN-PT), especially near its morphotropic phase boundary(MPB) with range of 32–35 mol% of PT, is one kind of the relaxor-type ferroelectrics which exhibits excellent piezoelectric properties than those of the most widely used $Pb(Zr,Ti)O_3$(PZT). And CFO is a well-known hard magnetic material, which exhibits high coercivity and moderate saturation magnetization.

In this paper, we focus on theoretical understanding the clamping effect of Si substrate on the magnetoelectric voltage coefficient (α_E) of PMN-PT/CFO laminated composites in transverse and longitudinal. Within the elastomechanics theory mode, we derived the transverse and longitudinal magnetoelectric voltage coefficient formula of laminated composites in ideal interface coupling state simply. And the α_E dependence of the volume fraction of piezoelectrics and the thickness of Si substrate were calculated. The relative mechanism was also discussed.

2. Theoretical consideration

Consider a simple bilayered composite of piezoelectric layer (PMN-PT) and magnetic layer(CFO) on Si substrate as shown

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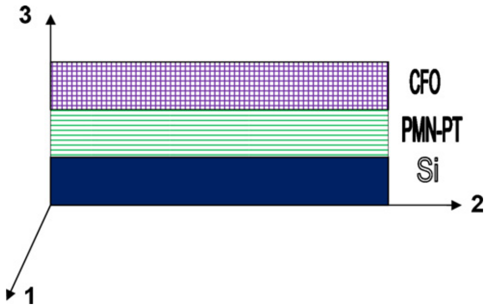

Fig. 1. Schematic illustration of the laminated composites of CFO/PMN-PT in the (1,2) plane on Si substrate.

in Fig. 1.The polarization direction coincides with the axis x_3. For a polarized piezoelectric phase with the symmetry ∞ m and magnetostrictive phase with cubic symmetry. We assume that the bilayer composites are homogeneous and the behavior is described by:

$$\begin{aligned} S_i &= s_{ij}T_j + d_{ki}E_k + q_{ki}H_k \\ D_k &= d_{ki}T_i + \varepsilon_{kn}E_n + \alpha_{kn}H_n \\ B_k &= q_{ki}T_i + \alpha_{kn}E_n + \mu_{kn}H_n \\ S_i^s &= s_{ij}^s T_j^s \end{aligned} \tag{1}$$

where S_i and T_j are strain and stress tensor components, E_k and D_k are the vector components of electric field and electric displacement, H_k and B_k are the vector components of magnetic field and magnetic induction, s_{ij}, d_{ki}, and q_{ki} are effective compliance, piezoelectric, and piezomagnetic coefficients, and ε_{kn}, μ_{kn}, α_{kn}, are effective permittivity, permeability, and ME coefficient, respectively.

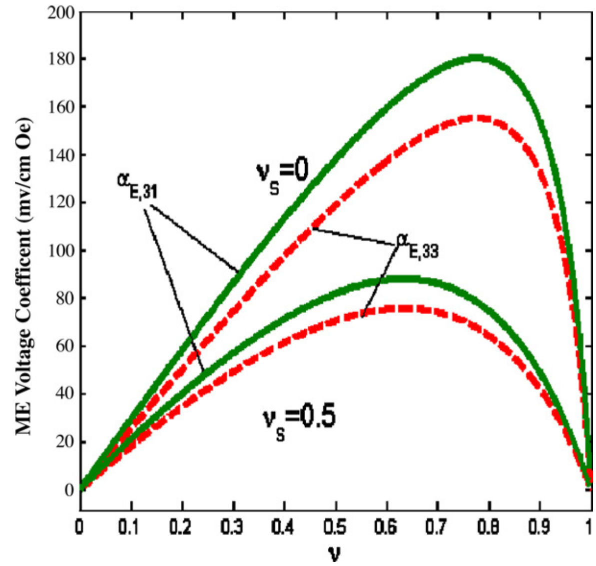

Fig. 2. Dependence of ME voltage coefficient on PMN-PT volume fraction v and substrate volume fraction v_s = 0 and 0.5 for the CFO/PMN-PT bilayer on Si substrate.

where $v = v^p/(v^p + v^m)$, $v_s = v^s/(v^p + v^m)$. Here v^p, v^m and v^s are the volume of piezoelectric, piezomagnetic phase and substrate, respectively. The superscripts p, m, s correspond to piezoelectric and piezomagnetic phase and substrate, respectively. The third term in Eq. (2) correspond to equilibrium condition (the total force projections on 1 and 2 axes are equal to zero). Using continuity conditions for magnetic and electric fields and open- and closed-circuit conditions, the expressions for transverse and longitudinal ME coefficient could be obtained as follows [16–19]:

$$a_{\mathrm{E},33} = -2q_{31}^m d_{31}^p v(1-v) \times \left\{ \left[(s_{11}^m + s_{12}^m)v + (s_{11}^p + s_{12}^p)(1-v) + (s_{11}^m + s_{12}^m) \times \frac{(s_{11}^p + s_{12}^p)}{s_{11}^s + s_{12}^s} v_s \right] \varepsilon_{33}^p - \frac{2(d_{31}^p)^2}{s_{11}^s + s_{12}^s}(s_{11}^m + s_{12}^m)v_s - 2(d_{31}^p)^2(1-v) \right\}^{-1} \tag{3}$$

$$a_{\mathrm{E},31} = (q_{11}^m + q_{21}^m) d_{31}^p v(1-v) \times \left\{ \left[(s_{11}^m + s_{12}^m)v + (s_{11}^p + s_{12}^p)(1-v) + (s_{11}^m + s_{12}^m) \times \frac{(s_{11}^p + s_{12}^p)}{s_{11}^s + s_{12}^s} v_s \right] \varepsilon_{33}^p - \frac{2(d_{31}^p)^2}{s_{11}^s + s_{12}^s}(s_{11}^m + s_{12}^m)v_s - 2(d_{31}^p)^2(1-v) \right\}^{-1} \tag{4}$$

For the solution of the set of Eq. (1), we assume (1,2) as the laminated composite plane (as shown in Fig. 1) and the direction 3 perpendicular to the sample plane. The laminated composite is poled with an electric field E along direction 3, the following boundary conditions are used:

$$\begin{aligned} & S_i^p = S_i^m = S_i^s, \quad (i = 1,2) \\ & T_i^p v + T_i^m(1-v) + T_i^s v^s = 0 \\ & T_3^p + T_3^m + T_3^s = 0 \end{aligned} \tag{2}$$

3. Results and discussion

In this model, we suppose that epilayer thickness is sufficiently large to neglect the influence of strain relaxation on average stresses in the structures which determine the ME voltage coefficient. The following material parameters [19] were used:

PMN-PT: $s_{11} = 11.73 \times 10^{-12}$ m^2/N, $s_{12} = -11.1 \times 10^{-12}$ m^2/N, $d_{31} = -360 \times 10^{-12}$ m/V, $\varepsilon_{33}/\varepsilon_0 = 3600$.
CFO: $s_{11} = 69 \times 10^{-12}$ m^2/N, $s_{12} = -2.4 \times 10^{-12}$ m^2/N, $q_{11} = -1880 \times 10^{-12}$ m/A, $q_{21} = q_{31} = 566 \times 10^{-12}$ m/A.
Si: $s_{11} = 7.67 \times 10^{-12}$ m^2/N, $s_{12} = -2.14 \times 10^{-12}$ m^2/N.

The PMN-PT volume fraction dependence of ME voltage coefficient of PMN-PT/CFO on Si substrate are shown in Fig. 2. From Fig. 2, it was found that for no Si substrate($v_s = 0$), the transverse and longitudinal α_E increases with the PMN-PT volume fraction v increasing, $\alpha_{E,31}$ and $\alpha_{E,33}$ attain the peak value of 180 mV/cm Oe and 155 mV/cm Oe for $v = 0.78$, respectively. And then $\alpha_{E,31}$ and $\alpha_{E,33}$ drop rapidly with increasing v. Pure PMN-PT ($v = 1$) and pure CFO ($v = 0$) do not reveal the ME effect. And the transverse ME coefficient $\alpha_{E,31}$ is larger than the longitudinal ME coefficient $\alpha_{E,33}$, which indicates a stronger transverse coupling than that of the longitudinal case in such laminate composites. But for Si substrate volume fraction of $v_s = 0.5$, the peak values of $\alpha_{E,31}$ and $\alpha_{E,33}$ of the PMN-PT/CFO laminate composites are 84 mV/cm Oe and 72 mV/cm Oe for $v = 0.63$, respectively, which also shows the optimum PMN-PT volume fraction v shifts to lead-lack compositions.

Fig. 3 shows the $\alpha_{E,33}$ of the system as the function of PMN-PT volume fraction v and Si substrate volume fraction v_s. It was found that the $\alpha_{E,33}$ of the systems decreased rapidly with the increase of v_s and the $\alpha_{E,33}$ of the systems vanishes when v_s arrived at 10 or more. That means there is strong clamping effects of Si substrate on the $\alpha_{E,33}$ of the system. Otherwise, the optimum PMN-PT volume fraction v shifts to lead-lack compositions with increase of Si substrate volume fraction v_s, which may be the fact that the substrate have the compressive stress to the PMN-PT in plane, the compressive stress enhances with the increase of substrate thickness, which change the lattice constant of PMN-PT and then lead to the low optimum PMN-PT volume fraction v.

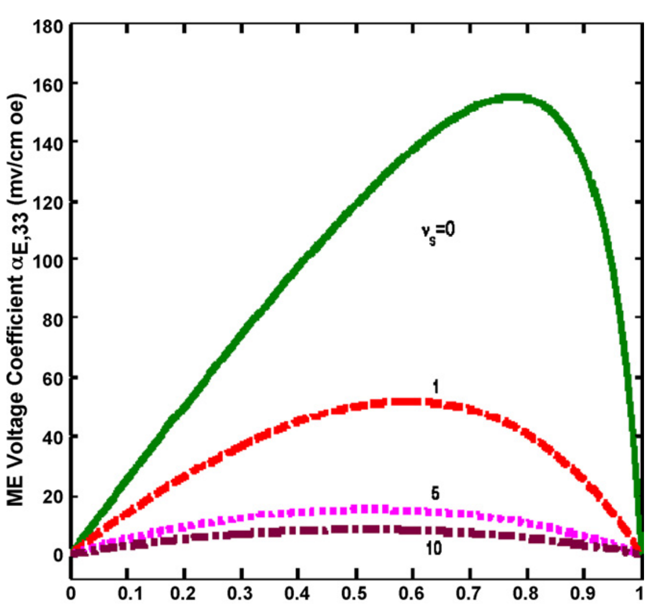

Fig. 3. $\alpha_{E,33}$ of the systems as the function of PMN-PT volume fraction v and Si substrate volume fraction v_s.

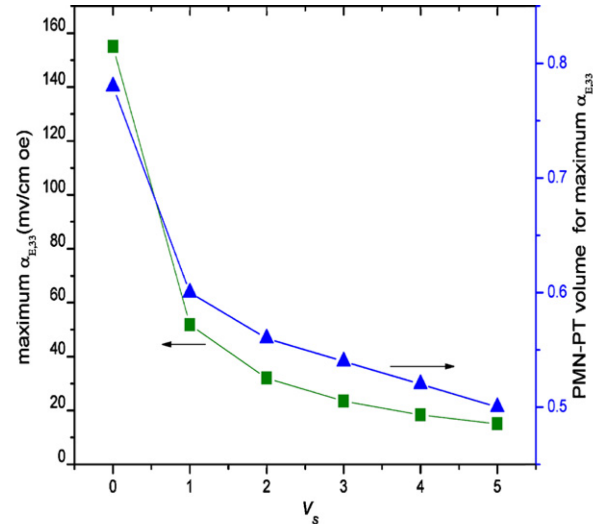

Fig. 4. Variation with substrate volume fraction v_s of the peak value of $\alpha_{E,33}$ of the system and the corresponding PMN-PT volume fraction.

Fig. 4 shows the variation in the peak value of $\alpha_{E,33}$ with the substrate volume fraction v_s along with the PMN-PT volume fraction v corresponding to peak $\alpha_{E,33}$. It was found that with increase of v_s, the $\alpha_{E,33}$ peak value decreased which indicate the increase in substrate thickness leads to decrease in properties of the composite. And the corresponding the PMN-PT volume fraction v also decreased, which also may ascribe to the change of PMN-PT lattice constant due to enhanced compressive stress with the increase of substrate thickness.

4. Conclusions

A theoretical model of PMN-PT/CFO laminated composite on Si substrate was established. Although the estimates here are based on bulk material parameters, it can easily be refined to take into account parameters for cobalt ferrite and PMN-PT bilayer on Si substrates. It was found that the strength of ME interactions is weaker with the increasing of thickness of Si substrate due to the strong clamping effects of Si substrate. With the increase of substrate thickness, the compressive stress of substrate to PMN-PT enhances, which change the lattice constant of PMN-PT and then lead to the low optimum PMN-PT volume fraction v.

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