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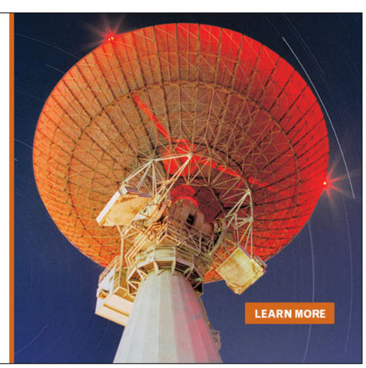

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Modeling of magnetoelectric effect in polycrystalline multiferroic laminates influenced by the orientations of applied electric/magnetic fields

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By using coarse graining model, the dependence of magnetoelectric (ME) coupling on the mutual orientations of magnetic and electric fields with respect to the orientation of layers in polycrystalline multiferroic laminates is investigated. It is shown that the ME coefficient, described by polarization change in response to the applied magnetic field, is proportional to the trace of effective piezomagnetic strain tensor projected onto laminate interfaces. The piezomagnetic strain significantly depends on the orientation of applied magnetic fields. The results obtained here demonstrate that the magnetoelectric effect in layered composites can be significantly improved by optimizing the configuration of applied electric/magnetic fields. © *2009 American Institute of Physics*. [DOI: 10.1063/1.3110062]

I. INTRODUCTION

The magnetoelectric (ME) effect in single phase or composite multiferroics is defined as the induced polarization response to an applied magnetic field or the induced magnetization response to an applied electric field. Obtaining large ME response is important for its potential applications in integrated multifunctional systems. The ME effect produced in multiferroic composites is found to be several orders higher than that in single phase multiferroic (see reviews[1,2] and references cited therein). Since polarization and magnetization in magnetic/piezoelectric composites interact by an overlap of the strain fields generated in magnetic and electric fields, the strength of the resultant ME coupling is mainly determined by a magnitude of strain mutually "transmitted" across the magnetic/piezoelectric interfaces. In this paper we consider a multilayer whose layers of magnetic and piezoelectric phases are nontextured polycrystals with grain sizes significantly less than thicknesses of the layers. Most of the sintered multiferroic ME composites fall in this category.

We note that the easy polarization/magnetization axes, as well as the effective piezoelectric/piezomagnetic tensors, in the polycrystalline multiferroic are determined not only by the intrinsic crystal symmetry as is the case for magnetic and piezoelectric single crystals, but also by the direction of poling electric/magnetic fields. This fact provides additional degree of freedom for the optimization of properties by varying configurations of the electric/magnetic fields. Indeed, under a fixed direction of the poling field, the measured ME coefficient varies with the change in angle between the poling field and the applied magnetic fields when the directions of applied ac and dc magnetic fields are parallel.[3,4] The ME coefficient also depends on the angle between the applied ac and dc magnetic fields, where significant enhancement in the magnitude of ME coefficient was obtained when the angle is about 45°.[5,6]

Among the reported composite microstructures in literature including laminate, particulate, and fibrous ones, most of the theoretical effort has been focused on understanding of ME effect in laminated composites, for two reasons: (i) simple geometry and fabrication and (ii) large ME effect.[3–14] The analytical foundation for describing ME coefficient of laminate composites was provided for the low-frequency response in Refs. 15 and 16, and at the dynamic resonance in Refs. 17 and 18. Both of these models assume homogenization of each layer and a perfect bonding between them. The influence of the imperfect interface bonding on the ME effect was investigated by introducing an interface coupling parameter,[19] by combing Green's function technique with perturbation theory,[20] and by using an extension of the shear lag model.[21] These studies demonstrated that the imperfect bonding significantly reduces the "transmission" of strain field across laminate interfaces and thus reduces the ME coefficient.

The objective of this manuscript is to conduct theoretical investigation of orientation-dependent ME properties in polycrystalline multiferroic laminates under various modes of applied electric/magnetic fields by using a simple coarse grain approximation.[15,16] We consider a trilayer system whose geometry is illustrated by Fig. 1. This system can be easily synthesized by sintering a polycrystalline "sandwich" consisting of $Ni_{0.6}Cu_{0.2}Zn_{0.2}Fe_2O_4$ (NCZF) and $0.9PbZr_{0.52}Ti_{0.48}O_3$-$0.1PbZr_{1/3}Nb_{2/3}O_3$ (0.9PZT-0.1PZN) (Ref. 7) layers. The top and bottom surfaces of the polycrystalline piezoelectric layer are assumed to be ideally bonded to the polycrystalline magnetic layers. The piezoelectric/ferromagnetic layers have very small electromechanical/magnetomechanical deformation in unpoled untextured polycrystals. Because of that we consider a poled state with the poling dc electric/magnetic fields designated as $\mathbf{E}^0$, $\mathbf{H}^0$, in

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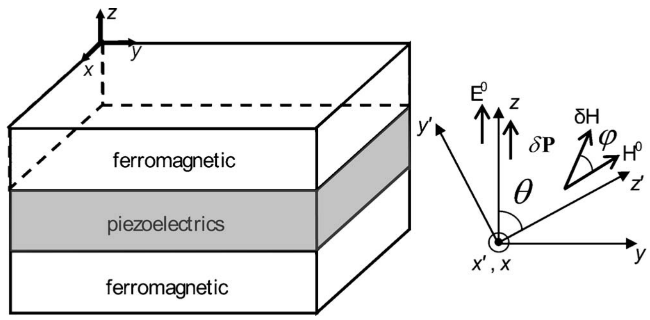

FIG. 1. Schematic illustration of configuration of applied electric/magnetic fields in trilayer polycrystalline multiferroic composites. The right figure shows a choice of coordinate systems (xyz) and $(x'y'z')$ located in the same "façade" plane of the left figure with the directions of z and z′ axes along the poling $\mathbf{E}^0$, $\mathbf{H}^0$ fields and parallel x and x' axes normal to the same plane.

Fig. 1. The applied ac magnetic field, $\delta\mathbf{H}$, is assumed to lie in the plane drawn through the poling direction of the vectors $\mathbf{E}^0$, $\mathbf{H}^0$. A "façade" of the trilayer shown in Fig. 1 coincides this plane. Since all polycrystalline layers are isotropic in the coarse graining approximation, and the vectors, $\mathbf{E}^0$, $\mathbf{H}^0$, $\delta\mathbf{H}$, are located in the same plane (yz, or $y'z'$) the system has the mirror symmetry with respect to this plane. As a result, the induced polarization, $\delta\mathbf{P}$, by $\delta\mathbf{H}$ is also located in the same plane.

II. THEORETICAL MODEL

The homogenized linear electromechanical/magnetomechanical responses of the constituent ferroelectric/ferromagnetic layers are described by constituent equations,

$$\varepsilon^p_{ij} = s^p_{ijkl}\sigma^p_{kl} + d^p_{kij}\delta E_k, \tag{1}$$

$$D_i = d^p_{ijk}\sigma^p_{jk} + k_{ij}\delta E_j, \tag{2}$$

where ε^p_{ij} and σ^p_{kl} are the strain and stress tensor components of the poled ferroelectric phase, D_i and δE_k are the vector components of the electric displacement and electric field, s^p_{ijkl} and k_{ij} are assumed to be isotropic effective compliance and permittivity tensors of the poled ferroelectric phase, respectively,

$$s^p_{ijkl} = \frac{1}{4\mu^p}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) - \frac{\upsilon^p}{2\mu^p(1+\upsilon^p)}\delta_{ij}\delta_{kl}, \tag{3}$$

$$k_{ij} = k_0\delta_{ij}, \tag{4}$$

where μ^p, υ^p, and k_o are the effective shear modulus, Poisson's ratio, and dielectric permittivity, respectively; d_{kij} is the component of the effective piezoelectric tensor. It has a cylindrical symmetry for considered poled untextured polycrystalline layers, the axis of symmetry being directed along the poling direction, $n^0_i = E^0_i/|\mathbf{E}^0|$. A general invariant form of the tensor d_{kij} is

$$d_{kij} = d_{31}n^o_k\delta_{ij} + \frac{d_{15}}{2}(\delta_{kj}n^o_i + \delta_{ki}n^o_j) + (d_{33} - d_{31} - d_{15})n^o_k n^o_i n^o_j, \tag{5}$$

where, d_{33}, d_{31}, and d_{15} are the effective longitudinal, transverse, and shear piezoelectric constants of the piezoelectric moduli in the matrix notation with respect to the poling direction, respectively. The constitutive equations of a magnetized ferromagnetic polycrystalline can be derived similarly

$$\varepsilon^m_{ij} = s^m_{ijkl}\sigma^m_{kl} + q_{kij}\delta H_k, \tag{6}$$

$$B_i = q_{ijk}\sigma^m_{jk} + \lambda_{ij}\delta H_j, \tag{7}$$

with

$$\lambda_{ij} = \lambda_0\delta_{ij}, \tag{8}$$

$$s^m_{ijkl} = \frac{1}{4\mu^m}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}) - \frac{\upsilon^m}{2\mu^m(1+\upsilon^m)}\delta_{ij}\delta_{kl}, \tag{9}$$

$$q_{kij} = q_{31}e^o_k\delta_{ij} + \frac{q_{15}}{2}(\delta_{kj}e^o_i + \delta_{ki}e^o_j) + (q_{33} - q_{31} - q_{15})e^o_k e^o_i e^o_j, \tag{10}$$

where ε^m_{ij} and σ^m_{kl} are the strain and stress tensor components, μ^p and υ^m are the effective shear modulus and Poisson's ratio of the magnetized ferromagnetic phase, δH_k and B_i are the vector components of the ac magnetic field and magnetic induction, q_{kij} and λ_{ij} are the components of the effective piezomagnetic tensor and permeability tensor, respectively, and $e^0_i = H^0_i/|\mathbf{H}^0|$, q_{33}, q_{31}, and q_{15} are the effective longitudinal, transverse, and shear piezomagnetic constants of the piezomagnetic moduli with respect to the direction of $\mathbf{H}^0$.

After applying an ac magnetic field, $\delta\mathbf{H}$, we calculated the induced polarization, $\delta\mathbf{P}$. The effective ME coefficient tensor is then characterized by $\alpha_{ij} = \delta P_i/\delta H_j$. If we assume that the value of k_0 in the laminate is much larger than that in the vacuum, then $\delta P_i = k_0\delta E_i$. In the following calculations all tensors and vectors are presented in the (xyz) coordinate system (Fig. 1) unless the use of other system is specially noted. In this case, by definition, $\mathbf{E}^0 \| \boldsymbol{n}_o = (0,0,1)$. According to Eq. (2) the electrostatic boundary condition can be written as

$$D_3 = d_{31}(\sigma^p_{11} + \sigma^p_{22}) + k_0\delta E_3 = 0. \tag{11}$$

From Eq. (11) we have

$$\delta P_3 = -d_{31}(\sigma^p_{11} + \sigma^p_{22}). \tag{12}$$

Equation (12) shows that the magnitude of induced polarization is related to the trace of in-plane biaxial stretch in the ferroelectric layer transmitted from the ferromagnetic layer. Based on the boundary conditions for the force balance and displacement continuity in the trilayer, we have

$$\sigma^p_{31} = \sigma^p_{32} = \sigma^p_{33} = 0, \quad \sigma^m_{31} = \sigma^m_{32} = \sigma^m_{33} = 0, \tag{13}$$

$$\sigma^m_{11}t_m + \sigma^p_{11}t_p = 0, \quad \sigma^m_{22}t_m + \sigma^p_{22}t_p = 0, \tag{14}$$

$$\varepsilon^p_{11} = \varepsilon^m_{11}, \quad \varepsilon^p_{22} = \varepsilon^m_{22}, \tag{15}$$

where t_p and t_m are the thicknesses of the ferroelectric and ferromagnetic layers, respectively. Since the poling direction is $n_o = (0,0,1)$ in the (xyz) coordinate system, using Eqs. (1), (6), and (13) into Eq. (15) gives

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$$(s_{11}^p + s_{12}^p)\sigma_{11}^p + d_{31}\delta E_3 + (s_{22}^p + s_{12}^p)\sigma_{22}^p + d_{31}\delta E_3 = (s_{11}^m + s_{12}^m)\sigma_{11}^m + \varepsilon_{11}^{mo} + (s_{22}^m + s_{12}^m)\sigma_{22}^m + \varepsilon_{22}^{mo}, \quad (16)$$

where ε_{11}^{m0} and ε_{22}^{m0} are the components of the piezomagnetic strain, ε_{ij}^{m0}, s_{11}^p, s_{22}^p, s_{12}^p, s_{11}^m, s_{22}^m, and s_{12}^m are the related components of compliance of piezoelectric and magnetic layers, respectively, with relationship $s_{22}^p = s_{11}^p$ and $s_{22}^m = s_{11}^m$. Using Eq. (14) in Eq. (16) gives

$$\sigma_{11}^p + \sigma_{22}^p = \frac{\varepsilon_{11}^{m0} + \varepsilon_{22}^{m0}}{(s_{11}^p + s_{12}^p) + (s_{11}^m + s_{12}^m)\frac{t_p}{t_m} - \frac{2(d_{31})^2}{k_0}}. \quad (17)$$

Combining Eqs. (12) and (17) gives the value of the ME coefficient, $\alpha_{3H} = \delta P_3 / |\delta \mathbf{H}|$ as

$$\alpha_{3H} = \frac{-d_{31}}{|\delta \mathbf{H}|} \frac{\varepsilon_{11}^{m0} + \varepsilon_{22}^{m0}}{(s_{11}^p + s_{12}^p) + (s_{11}^m + s_{12}^m)\frac{t_p}{t_m} - \frac{2(d_{31})^2}{k_0}}. \quad (18)$$

Under the given system configuration, the piezomagnetic strain in the $(x'y'z')$ coordinate system is

$$\varepsilon_{ij}'^{m0}(\delta \mathbf{H}) = \begin{pmatrix} q_{31}\delta H_3 & 0 & q_{15}\delta H_1 \\ 0 & q_{31}\delta H_3 & q_{15}\delta H_2 \\ q_{15}\delta H_1 & q_{15}\delta H_2 & q_{33}\delta H_3 \end{pmatrix}, \quad (19)$$

where $\delta \mathbf{H} = (\delta H_1, \delta H_2, \delta H_3)$ in the $(x'y'z')$ coordinate system. The components of the piezomagnetic strain in the (xyz) coordinate can be obtained by the $(x'y'z') \to (xyz)$ coordinate transformation, $\hat{\alpha}$,

$$\varepsilon_{ij}^{m0} = \alpha_{ik}\alpha_{jl}\varepsilon_{kl}'^{m0}, \quad (20)$$

where

$$\alpha_{ij} = \begin{bmatrix} 1 & 0 & 0 \\ 0 & \cos\theta & \sin\theta \\ 0 & -\sin\theta & \cos\theta \end{bmatrix}. \quad (21)$$

Substituting Eqs. (19) and (21) into Eq. (20) with $\delta H_1 = 0$, $\delta H_2 = \delta H \sin\varphi$, and $\delta H_3 = \delta H \cos\varphi$ gives

$$(\varepsilon_{11}^{m0} + \varepsilon_{22}^{m0}) = \{[q_{31}(1 + \cos^2\theta) + q_{33}\sin^2\theta]\cos\varphi + 2q_{15}\sin\theta\cos\theta\sin\varphi\}\delta H. \quad (22)$$

If we introduce a projection tensor $\mathbf{A} = \mathbf{I} - \mathbf{n} \otimes \mathbf{n}$, which maps a three-dimensional (3D) tensor on the two-dimensional (2D) plane normal to the unit vector $\boldsymbol{n}$, then the 2D projection of the tensor ε_{ij}^{mo} on the layer planes with its normal direction, $\mathbf{n} = (0,0,1)$, is

$$\mathbf{A} \times \boldsymbol{\varepsilon}^{m0} \times \mathbf{A} = \begin{pmatrix} \varepsilon_{11}^{m0} & \varepsilon_{12}^{m0} \\ \varepsilon_{21}^{m0} & \varepsilon_{22}^{m0} \end{pmatrix}. \quad (23)$$

As follows from Eq. (23), $\varepsilon_{11}^{m0} + \varepsilon_{22}^{m0}$ entering Eq. (18) is a trace of the piezomagnetic in-plane strain, which is a projection of a homogenized 3D piezomagnetic strain tensor, $\boldsymbol{\varepsilon}^{m0}$, onto the laminate interface. Thus Eq. (18) indicates that the value of the ME coefficient is proportional to the trace of in-plane piezomagnetic strain, which in turn depends on the orientation of both ac and dc magnetic fields, as shown in Eq. (22). If we measured the value of α_{3H} in unit of

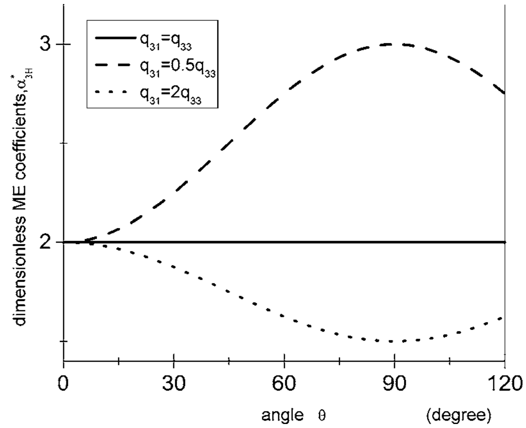

FIG. 2. The dimensionless $\mathrm{ME_H}$ coefficient, α_{3H}^*, as a function of the angle θ between the poling (dc) electric and magnetic fields under condition of parallel directions $\boldsymbol{H}^{\circ}$ and $\boldsymbol{\delta H}$ at different values of q_{31}, q_{33}.

$-d_{31}q_{33}/\{(s_{11}^p + s_{12}^p) + (s_{11}^m + s_{12}^m)(t_p/t_m) - [2(d_{31})^2/k_0]\}$, then substituting Eq. (22) into Eq. (18), reduces it to a dimensionless form,

$$\alpha_{3H}^* = \left[\frac{q_{31}}{q_{33}}(1 + \cos^2\theta) + \sin^2\theta\right]\cos\varphi + 2\frac{q_{15}}{q_{33}}\sin\theta\cos\theta\sin\varphi. \quad (24)$$

Plots of the change of dimensionless $|\alpha_{3H}^*|$ versus θ at different sets of q_{31} and q_{33} are presented in Fig. 2 for a particular case of parallel directions of the ac and dc magnetic fields. The figure demonstrates that $|\alpha_{3H}^*|$ reaches a maximum at $\theta = 0°$ in the case of $q_{31} > q_{33}$ (this case is corresponding to the transversely magnetized and transversely poled mode)[13] and at $\theta = 90°$ in the case of $q_{33} > q_{31}$ (this case is corresponding to the longitudinally magnetized and transversely poled mode[13]). For a special case of $q_{31} = q_{33}$, the angle dependency of ME coefficient vanishes. The results obtained here are consistent with the experimental report and prediction.[3,4] More interestingly, if the directions of $\boldsymbol{\delta}\mathbf{H}$ and $\mathbf{H}^0$ are not the same, then $\varepsilon_{ij}'^{m0}$ in Eq. (19) has off-diagonal components, which have contributions to the trace of the in-plane piezomagnetic strain tensor (the projection of $\alpha_{ik}\alpha_{jl}\varepsilon_{kl}'^{m0}$). In this case, the behavior of the orientation-dependent ME coefficient becomes more complex. Figure 3 shows the contour of $|\alpha_{3H}^*|$ with respect to θ and φ for the different sets of the constants, q_{15}, q_{31}, and q_{33}. Figure 3 also demonstrates that in the case of $q_{15} \gg q_{31}$, $q_{15} \gg q_{33}$, $|\alpha_{3H}^*|$ has a maximum at $\theta = 45°$ and $\varphi = 90°$. This would be in agreement with the observation of a significant enhancement in the magnitude of ME coefficient in the NCZF/0.9PZT-0.1PZN/NCZF trilayer system[7] if this was the case wherein piezomagnetic shear, q_{15}, is much higher than longitudinal coefficient, q_{33}.

The obtained results confirm the intuitive interpretation of the strain-mediated ME coupling of the layers. The strain field across the interfaces generated by the applied ac fields, in fact, is caused by the crystal lattice misfit (elastic incompatibility) along the interfaces between the layers induced by these fields. When the ME effect is characterized by a polarization induced by a magnetic field, the trace of the in-plane

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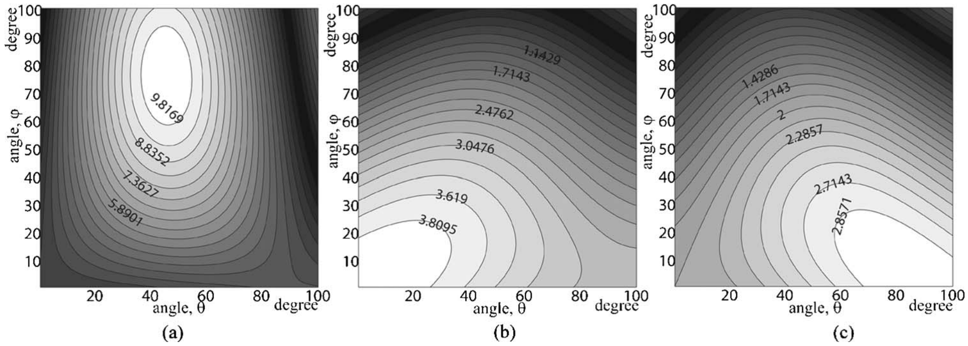

FIG. 3. The contour of the dimensionless ME_H coefficient, α^*_{3H}, with respect to the angle θ between the dc electric and magnetic fields, and the angle φ between the ac and dc magnetic fields for different sets of q_{15}, q_{31}, q_{33}; (a) $q_{15}=5q_{33}$, $q_{31}=0.5q_{33}$, (b) $q_{15}=q_{33}$, $q_{31}=2q_{33}$, and (c) $q_{15}=0.5q_{33}$, $q_{31}=0.5q_{33}$.

piezomagnetic strain tensor (the projection of $\alpha_{ik}\alpha_{jl}\varepsilon'^{m0}_{kl}$) is a measure of this incompatibility and thus is also a measure of the ME coupling. The obtained results demonstrate that the greater the trace of the magnetic-field induced in-plane strain on laminate interfaces the higher is the ME coupling. Therefore, the reason for the orientation dependence of ME coupling is that the trace of this in-plane strain does depend on configuration of the system, on a mutual orientation of the poling directions, and applied magnetic or electric fields.

III. CONCLUSION

In summary, the proposed model allows to characterize the ME effect in polycrystalline multiferroic laminates as a function of the orientations of applied electric/magnetic fields. The calculated ME coefficient, $\delta\mathbf{P}/\delta\mathbf{H}$, is found to be proportional to the trace of magnetically induced strain projected on laminate interfaces. This magnetically induced strain significantly depends on the orientation of applied magnetic fields with respect to the surface of the layers.

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