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Author for correspondence:

George J. Weng

e-mail: gjweng@soe.rutgers.edu

Direct and converse nonlinear magnetoelectric coupling in multiferroic composites with ferromagnetic and ferroelectric phases

Juanjuan Zhang^{1,2,4}, Chao Fang^{3,4} and

George J. Weng⁴

¹Key Laboratory of Mechanics on Environment and Disaster in Western China, The Ministry of Education of China, and

²Department of Mechanics and Engineering Science, College of Civil Engineering and Mechanics, Lanzhou University, Lanzhou, Gansu 730000, People's Republic of China

³Department of Electrical and Electronic Engineering, Wuhan Polytechnic University, Wuhan, Hubei 430023, People's Republic of China

⁴Department of Mechanical and Aerospace Engineering, Rutgers University, New Brunswick, NJ 08903, USA

GJW, 0000-0003-1543-1535

In this paper, we develop a theoretical principle to calculate the direct and converse magnetoelectric (ME) coupling response of ferromagnetic/ferroelectric composites with 2–2 connectivity. We first present an experimentally based constitutive equation for Terfenol-D, and then build the mechanism of domain switch for the ferroelectric phase. In the latter, the change of Gibbs free energy, thermodynamic driving force and kinetic equations for domain growth are also established. These two sets of constitutive equations are shown to capture the experimental data of Terfenol-D and PZT, respectively, well. For the direct effect under an applied magnetic field, the induced electric field and the overall ME coupling coefficient are determined. For the converse effect under an applied electric field, the induced magnetization and the excited magnetic field are obtained. Both the induced electric field under direct effect and the excited magnetic field under converse effect are shown to display the hysteretic characteristics, and also in good agreement with experiments.

We conclude that the developed theory can both qualitatively and quantitatively reflect the essential features of nonlinear direct and converse ME coupling of the multiferroic composites.

1. Introduction

Multiferroic materials generally refer to the class of materials which are both ferroelectric and ferromagnetic, and in many cases they are also ferroelastic. A ferroelectric material is characterized by the presence of ionic dipoles after its paraelectric phase is cooled down below the Curie temperature, creating a polarization, P . Ferromagnetic behaviour on the other hand does not involve formation of ionic dipoles; it is caused instead by the ordered electron spins that generate magnetization, M . The electric polarization, P , and magnetization, M , are often referred to as the 'order' parameters in ferroic materials. Thus, a multiferroic material will simultaneously possess both P and M , but to have a strong magnetoelectric (ME) coupling, application of the magnetic field H must be able to cause the electric polarization P to switch, or, conversely, application of the electric field E must be able to cause magnetization M to switch. It is the characteristic of strong ME coupling that makes multiferroic materials special in a wide range of applications in electronic and magnetic devices. For instance, in information storage, this coupling allows the data to be written electronically and read magnetically, avoiding the problem of having to generate a locally high magnetic field to write.

However, ferroelectricity and ferromagnetism rarely coexist in a single-phase material, and even if they do, the ME coupling is generally weak. Indeed, only a very limited number of single-phase oxides, such as Cr_2O_3 , YMnO_3 , BiFeO_3 , BiMnO_3 , RMnO_3 and RMn_2O_5 (R : rare earth), have been reported to exhibit multiferroic phenomena. Due to the limited availability of single-phase multiferroics and their usually weak coupling effect, recent efforts have been directed toward production of multiferroic composites [1–4]. In such composites, the response of the magnetic (or electric) phase under a magnetic (or electric) field can be transmitted to the electric (or magnetic) phase through their common elastic deformation, leading to the ME coupling [5,6]. The strength of ME coupling depends strongly on the connectivity of the two phases. In a recent analysis on the linear magneto-electro-elastic coupling [7], it is proven that, among the 0–3 particulate, 1–3 fibrous and 2–2 lamellar connectivity, the 2–2 type possesses the highest in-plane ME coupling coefficient. Due to its simple structure, simple fabrication process and high ME coefficient at room temperature, the laminated ME composites have been widely applied in many smart devices, such as ME sensors, energy harvesters and transformers [8–15].

In addition to the connectivity, resonance is also a key for ME devices. Many ME devices work at the resonance state because the ME structures have larger ME coefficients. Resonance is a dynamic problem, and large deformation also tends to occur; its analysis is beyond the scope of this investigation. Besides, the choice of component materials is also important to achieve strong coupling strength [16–19]. To fully explore this characteristic, ferromagnetic and ferroelectric materials with strong nonlinear hysteretic characteristics under an applied magnetic and electric field, respectively, are frequently investigated [20–25]. Many experiments have shown that such nonlinearity could lead to stronger nonlinear ME coupling [26–31]. Such combination makes the multiferroic composites to have very complex nonlinear hysteretic characteristics, and, when properly tuned, there are many novel applications that the multiferroic composites could render. For example, Shen *et al.* [26] designed a memtransistor made of Ni/PMN-PT/Ni heterostructure, and found that this multiferroic composite exhibits a broader hysteresis loop and higher remanence at zero bias field. Shen *et al.* [27] demonstrated that a magnetoelectric hexaferrite (BSCFAO) exhibits a strong nonlinear ME effect at room temperature so that an applied magnetic field is able to change the electric polarization in a non-volatile way. Subhani *et al.* [28] studied the temperature-dependent behaviour of PZT/Terfenol-D composite, and found that the operating temperature and the volume fraction of the constituent phases have significant effects on the ME coefficient. To understand the temperature-frequency coupling effect, Gualdi

et al. [29] investigated the hysteretic behaviour of PMN-PT/CFO and PMN-PT/NFO composites at low temperature with different frequencies. To provide some insights into the dimensional effects, Burdin *et al.* [30] obtained the ME effect hysteresis in Ni-PZT structure. Kambale *et al.* [31] fabricated the asymmetric and symmetric laminated ME structures composed of piezoelectric macro-fibre and Ni, and then measured the ME coefficients along the length, width and thickness directions of the laminated structures.

In the meantime, some theoretical studies have started to emerge. Some representative works are carried out [32–34]. For example, Zhang & Gao [32] established a nonlinear dynamic hysteretic model for giant magnetostrictive/piezoelectric composites, and implemented a finite-element analysis to investigate the influences of hysteresis and temperature on the ME effects of M/P/M and P/M/P structures (M: magnetic; P: piezoelectric). Combining a nonlinear hysteretic magnetostrictive model and a structural vibration theory, Xu *et al.* [33] studied the frequency dependence of the harmonic hysteretic ME effect in a multiferroic composite. In addition, Zhou *et al.* [34] built a hysteretic nonlinear ME theoretical model for the symmetric tri-layered structure. It must be pointed out that, in these nonlinear theoretical studies of ME coupling, only the nonlinear constitutive equations of the ferromagnetic phase are considered. For the ferroelectric phase, only the linear constitutive equations are implemented, usually patterned after that of a piezoelectric material. These works represent some key developments in this area and have greatly facilitated the theoretical study of ME composites.

In order to simultaneously consider the influence of nonlinear characteristics of ferroelectric materials and ferromagnetic materials on the ME effect for ME composites, some studies have been carried out [35–38]. More specifically, Lu *et al.* [35] established a model by using thermodynamic theory combined with the Eshelby's inclusion theory to study the electric, magnetic and ME properties of the nano-structured multiferroic composites. Avakian *et al.* [36,37] applied the physically motivated constitutive models for ferromagnetic and ferroelectric phases and investigated the ME coupling coefficients in multifunctional composite consisting of a ferromagnetic inclusion in a ferroelectric matrix. Xu *et al.* [38] proposed a new probabilistic domain switching function considering the surface FM anisotropy and the interface charge-mediated effect and investigated the size-dependent electric tuning magnetization behaviour for the ME laminates at nanoscale.

Against the backdrops of these studies, we will establish a nonlinear theoretical model for ME composite by considering simultaneously the nonlinear characteristics of ferromagnetic and ferroelectric phases in this paper. The basic frameworks of the nonlinear constitutive equations of the ferromagnetic phase in the form of magnetostrictive materials, and those of the ferroelectric phase with domain switch, will be introduced in §2. In the latter case, the change of Gibbs free energy, the thermodynamic driving force, and kinetic equations for domain growth will also be covered. Then calculation of ME coupling coefficient through their commonly shared strain will be introduced. In §3, we will present and discuss the calculated results. The nonlinear characteristics of each component materials will be addressed first, and then the nonlinear ME effect of the multiferroic composites will be investigated. Along the way the change of free energy and the nature of domain growth in the ferroelectric layer will be examined during the magnetic and electric coupling process. Finally, some conclusions are drawn in §4.

2. The nonlinear theoretical model for laminated multiferroic composites

Consider the L-T (longitudinal-transverse) mode multiferroic composite composed of ferromagnetic and ferroelectric materials shown in figure 1. The applied magnetic field is along the length direction of the composite. Because of the magnetostriction effect, the ferromagnetic layer stretches along the applied magnetic field. We assume that the interface is perfect, then the ferroelectric layer is driven by the strain of ferromagnetic layer. The polarization electric field along the thickness direction is produced as a consequence of their elastic coupling.

Figure 1. Illustration of the L-T mode ME laminated composite. (Online version in colour.)

(a) The nonlinear constitutive equations of the ferromagnetic material

As a kind of ferromagnetic materials, Terfenol-D is attractive for many ME devices due to its larger magnetostriction when subjected to a magnetic field. Many experiments show that the relationship between stress and strain is nonlinear for Terfenol-D [20,21,39]. For example, for either Terfenol-D rods or thin films, the magnetization curves and the magnetostrictive strain curves not only have saturation feature, but also have the classical hysteresis characteristic. Due to its significance, many nonlinear models have been developed to describe the magneto-mechanical response for Terfenol-D and other ferromagnetic materials. Several multiscale models have been established by considering the micro-domain structures and the magneto-crystalline anisotropy of the materials [40–50]. Phenomenological models have also been presented based on the thermodynamic theory [51–54].

Without loss of generality, we express the stress–strain relationship of the ferromagnetic material as

$$\boldsymbol{\varepsilon}^m = \mathbf{s}^m \boldsymbol{\sigma}^m + \boldsymbol{\lambda} + \boldsymbol{\varepsilon}^{\text{initial}}, \quad (2.1)$$

where $\boldsymbol{\varepsilon}^m$ and $\boldsymbol{\sigma}^m$ are strain and stress tensors for the ferromagnetic material, $\mathbf{s}^m$ is the compliance matrix, and $\boldsymbol{\lambda}$ is the magnetostrictive strain. We have

$$\boldsymbol{\varepsilon}^m = [\varepsilon_1^m, \varepsilon_2^m, \varepsilon_3^m, \varepsilon_4^m, \varepsilon_5^m, \varepsilon_6^m], \quad (2.2)$$

$$\boldsymbol{\sigma}^m = [\sigma_1^m, \sigma_2^m, \sigma_3^m, \sigma_4^m, \sigma_5^m, \sigma_6^m] \quad (2.3)$$

and

$$\boldsymbol{\lambda} = [\lambda_1, \lambda_2, \lambda_3, \lambda_4, \lambda_5, \lambda_6]. \quad (2.4)$$

The initial strain $\boldsymbol{\varepsilon}^{\text{initial}}$ has the following relationship with the initial stress $\boldsymbol{\sigma}^{\text{initial}}$:

$$\boldsymbol{\varepsilon}^{\text{initial}} = \mathbf{s}^m \boldsymbol{\sigma}^{\text{initial}}. \quad (2.5)$$

The shear strains caused by the magnetic field are zero, that is $\lambda_4 = \lambda_5 = \lambda_6$, and λ_2 and λ_3 can be expressed $\lambda_2 = \lambda_3 = -\nu_s \lambda_1$, where ν_s is the secant Poisson's ratio. When the ferromagnetic material is in the degaussing state (external magnetic field and the initial stress are zero), the magnetic domains are disorderly arranged, and the directions of the spontaneous magnetization of the magnetic domains are random. So the ferromagnetic is not showing elongation or shortening in any directions, as shown in figure 2a.

When an initial compression is applied, the magnetic domain will rotate, so that the ferromagnetic material has a pre-compression strain (figure 2b). At this time, since there is no magnetic field, the magnetostrictive strain is still zero. From the free state in (a), if we apply a certain magnetic field, the magnetic domains inside the ferromagnetic material will rotate toward the direction of the field, causing the material to elongate, as shown in figure 2c. When the applied magnetic field is large enough, the internal magnetic domains will completely turn

Figure 2. Schematic diagram of magnetic domain rotation under magnetic field and prestress. (Online version in colour.)

toward the field direction, reaching a saturation state as shown in figure 2d. It is noted that the magnetostrictive strain from (b) to (d) is larger than from (a) to (d). All of these features have been confirmed in the experiments [55] and must be captured for the FM phase. We have found that they can all be represented by the Wang–Zhou theory. The Wang–Zhou nonlinear magneto-elastic coupling model is [53]

$$\lambda_1 = -\frac{\tilde{B}}{M_s^2}(T - T_r)\tilde{M}^2 + \begin{cases} \frac{[1 - \tanh(\sigma_1^m/\sigma_s)]\lambda_s}{M_s^2}\tilde{M}^2 & \left(\frac{\sigma_1^m}{\sigma_s} \geq 0\right) \\ \frac{[2 - \tanh(2\sigma_1^m/\sigma_s)]\lambda_s}{2M_s^2}\tilde{M}^2 & \left(\frac{\sigma_1^m}{\sigma_s} \leq 0\right) \end{cases} \quad (2.6)$$

and

$$H_1 = H_{\text{eff}} - \tilde{\eta}\tilde{M}, \quad (2.7)$$

where T and T_r , respectively, are the experimental temperature and the spin reorientation temperature. With the unit in Kelvin, T_r is about 273 K (or 0°C) for Terfenol-D. H_1 is the applied magnetic field along the longitudinal direction and H_{eff} is the effective field. $\tilde{M}$ is magnetization, and $\tilde{\eta}$ can be expressed as

$$\tilde{\eta} = \eta - \frac{3\tilde{B}\sigma_1^m}{\mu_0 M_s^2}(T - T_r) + \begin{cases} \frac{3[1 - \tanh(\sigma_1^m/\sigma_s)]\lambda_s\sigma_1^m}{\mu_0 M_s^2} & \left(\frac{\sigma_1^m}{\sigma_s} \geq 0\right) \\ \frac{3[2 - \tanh(2\sigma_1^m/\sigma_s)]\lambda_s\sigma_1^m}{2\mu_0 M_s^2} & \left(\frac{\sigma_1^m}{\sigma_s} \leq 0\right) \end{cases}, \quad (2.8)$$

where $\tilde{B}$ is the slope determined by the saturation magnetostrictive strain-temperature cure. η is the Weiss molecular field coefficient. The vacuum permeability μ_0 is $4\pi \times 10^{-7} \text{ H m}^{-1}$. σ_s , λ_s

and M_s are the saturation stress, saturation magnetostrictive strain and saturation magnetization along the longitudinal direction, respectively.

The magnetization can be obtained through the following equation:

$$\begin{aligned} & \mu_0 \delta_m \frac{D^2}{2\rho\beta} \left(\frac{d\tilde{M}}{dt} \right)^2 + \mu_0^{1/2} \delta_m \left(\frac{G_0 S_0 H_0}{\rho} \right)^{1/2} \left(\frac{d\tilde{M}}{dt} \right)^{3/2} \\ & + \left[\varsigma K - \delta_m \tilde{\eta} (M_{\text{an}} - \tilde{M}) - \tilde{c} \varsigma K \tilde{\eta} \frac{dM_{\text{an}}}{dH_{\text{eff}}} \right] \frac{d\tilde{M}}{dt} \\ & - \left[\delta_m (M_{\text{an}} - \tilde{M}) + \tilde{c} \varsigma K \frac{dM_{\text{an}}}{dH_{\text{eff}}} \right] \frac{dH_1}{dt} = 0, \end{aligned} \quad (2.9)$$

in which

$$\delta_m = \begin{cases} 0, & \text{if } \dot{H}_1 < 0 \text{ and } M_{\text{an}} - \tilde{M} > 0, \\ 0, & \text{if } \dot{H}_1 > 0 \text{ and } M_{\text{an}} - \tilde{M} < 0 \\ 1, & \text{otherwise.} \end{cases} \quad (2.10)$$

In this equation, D and S_0 are the thickness and the cross-sectional area of the ferromagnetic layer, ς is a sign function, which is -1 when the magnetic field decreases and is 1 when the field increases. The parameters ρ and K are the resistivity and microstructural parameter, respectively. The reversibility coefficient $\tilde{c}$ is a dimensionless parameter. The dimensionless constant G_0 is 0.1356 . H_0 represents the internal potential experienced by domain walls, and β is a geometrical factor with $\beta = 6$ for laminations.

The relationship between the anhysteretic magnetization M_{an} and the effective magnetic field H_{eff} is

$$M_{\text{an}} = M_S \frac{(1 - T/T_c)^x}{(1 - T_r/T_c)^x} \left[\coth \left(\frac{3\chi_m(1 - T_r/T_c)^x}{M_S(1 - T/T_c)^x} H_{\text{eff}} \right) - \frac{M_S(1 - T/T_c)^x}{3\chi_m(1 - T_r/T_c)^x H_{\text{eff}}} \right]. \quad (2.11)$$

The Curie temperature T_c is 656.3 K (about 383.3°C), and $x = 0.5$ for Terfenol-D material. Parameter χ_m stands for the initial linear magnetic susceptibility.

(b) The nonlinear constitutive equations of ferroelectric material

Compared with ferromagnetic materials, there are obvious 90° and 180° domains inside the ferroelectric material. The domain switch leads to the nonlinear hysteresis behaviour of ferroelectric materials. There have been considerable studies on the nonlinear behaviours of ferroelectric single crystals and polycrystals [56–63]. Phase field simulations have further revealed more in-depth microstructures evolution [64–69]. In the present approach, we will take a thermodynamics-based micromechanics approach to study the evolution of ferroelectric domain for the ferroelectric layer. The basic frameworks of this approach can be found in [70–72].

Due to its electromechanical coupling, the nonlinear constitutive equations can be most conveniently cast in the coupled form. Using $\mathbf{X}$ to represent the electromechanical load and $\mathbf{Y}$ the response, it can be written in Voigt's notations as

$$\mathbf{Y} = \mathbf{MX} + \mathbf{Y}^{sp}, \quad (2.12)$$

where $\mathbf{X}$ comprises the stress σ^p and the electric field E as

$$\mathbf{X} = [\sigma_1^p, \sigma_2^p, \sigma_3^p, \sigma_4^p, \sigma_5^p, \sigma_6^p, E_1, E_2, E_3], \quad (2.13)$$

and $\mathbf{Y}$ is made up with the strain ε^p and electric displacement D as

$$\mathbf{Y} = [\varepsilon_1^p, \varepsilon_2^p, \varepsilon_3^p, \varepsilon_4^p, \varepsilon_5^p, \varepsilon_6^p, D_1, D_2, D_3]. \quad (2.14)$$

Figure 3. Transversely isotropic materials (a): 1–2 plane is isotropic (0° domain), (b) 2–3 plane is isotropic (90° domain). (Online version in colour.)

The superscript ‘ p ’ signifies the piezoelectric (ferroelectric) phase. The last term $\mathbf{Y}^{sp}$ is composed of the spontaneous strain $\boldsymbol{\varepsilon}^{sp}$ and spontaneous polarization $\mathbf{P}^{sp}$, as

$$\mathbf{Y}^{sp} = [\varepsilon_1^{sp}, \varepsilon_2^{sp}, \varepsilon_3^{sp}, \varepsilon_4^{sp}, \varepsilon_5^{sp}, \varepsilon_6^{sp}, P_1^{sp}, P_2^{sp}, P_3^{sp}]. \quad (2.15)$$

In addition, $\mathbf{M}$ is the electromechanical compliance tensor, which consists of three parts as

$$\mathbf{M} = \begin{bmatrix} \mathbf{s}^p & \mathbf{d}^T \\ \mathbf{d} & \mathbf{k} \end{bmatrix}, \quad (2.16)$$

in which $\mathbf{s}^p$, $\mathbf{d}$ and $\mathbf{k}$ are the elastic compliance, piezoelectric compliance and dielectric permittivity tensors, respectively. The electromechanical moduli tensor $\mathbf{L}$ is the inverse of $\mathbf{M}$, given by $\mathbf{L} = \mathbf{M}^{-1}$.

During domain switch, $\mathbf{L}$ and $\mathbf{M}$ represent the effective electromechanical moduli and compliances, respectively, of the 2-domain ferroelectric phase. It depends on the volume concentrations of the parent and product domains, c_0 and c_1 , respectively, as well as their compliance tensors, $\mathbf{M}_0$ and $\mathbf{M}_1$. Based on the homogenization theory of a 2-phase material, the effective compliance $\mathbf{M}$ can be calculated from

$$\mathbf{M} = (c_0 \mathbf{M}_0 + c_1 \mathbf{M}_1 \mathbf{B}_1) \mathbf{Q}, \quad (2.17)$$

where $\mathbf{B}_1$ is the electromechanical field concentration tensor of the product phase, whose expression is shown in appendix A, and $\mathbf{Q} = (c_1 \mathbf{B}_1 + c_0 \mathbf{I})^{-1}$. In the present L-T mode, the two different domain orientations shown in figure 3 will be considered.

(i) Gibbs free energy

For the ferroelectric material, different domain states correspond to different energy levels. As the domain state changes due to domain switch, the Gibbs free energy will change. As shown in [71], the change of Gibbs free energy can be expressed as

$$\Delta G = -\frac{c_1}{2} (\bar{\mathbf{X}}^T \mathbf{A} \bar{\mathbf{X}} + \bar{\mathbf{X}}^T \mathbf{B} \mathbf{Y}^{ds} + \mathbf{Y}^{dsT} \mathbf{C} \mathbf{Y}^{ds}) = -(\Delta G_A + \Delta G_B + \Delta G_C). \quad (2.18)$$

It consists of three components characterized by the three matrices, $\mathbf{A}$, $\mathbf{B}$ and $\mathbf{C}$, whose expressions are shown in appendix A. Here an overbar signifies that it is an averaged field for the 2-domain ferroelectric phase, so that $\bar{\mathbf{X}}$ is the average field of $\mathbf{X}$. The superscript ‘ ds ’ signifies that the quantity is related to ‘domain switch’, i.e. the change of the indicated quantity from the parent to the product phase. Furthermore, the average field $\bar{\mathbf{X}}$ can be evaluated from the average response $\bar{\mathbf{Y}}$ through

$$\bar{\mathbf{Y}} = \mathbf{M}_0 \bar{\mathbf{X}} + c_1 (\mathbf{Y}^{ds} + \mathbf{Y}^*) + \mathbf{Y}_0^{sp}, \quad \text{or conversely, } \bar{\mathbf{X}} = \mathbf{M}_0^{-1} [\bar{\mathbf{Y}} - c_1 (\mathbf{Y}^{ds} + \mathbf{Y}^*) - \mathbf{Y}_0^{sp}]. \quad (2.19)$$

In this way, if $\bar{\mathbf{Y}}$ (such as the magnetostrictive strain transmitted from the ferromagnetic phase) is known, $\bar{\mathbf{X}}$ (such as electric field, E_3) can be determined. Here $\bar{\mathbf{Y}}$ is the average of $\mathbf{Y}$ over both domains and $\mathbf{Y}_0^{sp}$ is the spontaneous strain and spontaneous polarization of the parent domain.

The expression for the domain switch response, $\mathbf{Y}^{\text{ds}}$, represents the difference of the spontaneous response between the product and the parent one, as

$$\mathbf{Y}^{\text{ds}} = \mathbf{Y}_{(1)}^{\text{sp}} - \mathbf{Y}_{(0)}^{\text{sp}}, \quad (2.20)$$

where $\mathbf{Y}_{(0)}^{\text{sp}}$ and $\mathbf{Y}_{(1)}^{\text{sp}}$ are the spontaneous strain and spontaneous polarization of the parent and the product domains, respectively. For example, if the parent domain is the 90° domain and the product one the 0° domain, then $\mathbf{Y}_{(0)}^{\text{sp}} = \mathbf{Y}_{90^\circ}^{\text{sp}}$ and $\mathbf{Y}_{(1)}^{\text{sp}} = \mathbf{Y}_{0^\circ}^{\text{sp}}$. The spontaneous strain and spontaneous polarization in 0° and 90° domains can be written as

$$\mathbf{Y}_{0^\circ}^{\text{sp}} = \begin{bmatrix} \frac{a}{a_0} - 1, & \frac{a}{a_0} - 1, & \frac{c}{a_0} - 1, & 0, & 0, & 0, & 0, & 0, & P_s \end{bmatrix} \quad (2.21)$$

and

$$\mathbf{Y}_{90^\circ}^{\text{sp}} = \begin{bmatrix} \frac{c}{a_0} - 1, & \frac{a}{a_0} - 1, & \frac{a}{a_0} - 1, & 0, & 0, & 0, & P_s, & 0, & 0 \end{bmatrix}. \quad (2.22)$$

Here a_0 , a and c are the lattice constant of ferroelectric material. a_0 is independent of temperature. a and c are related to temperature. P_s is the spontaneous polarization. The express of $\mathbf{Y}^*$ is shown to follow

$$\mathbf{Y}^* = (\mathbf{M}_1 - \mathbf{M}_0)\mathbf{B}_1\mathbf{Q}[\bar{\mathbf{X}} - c_0\mathbf{L}_0(\mathbf{I} - \mathbf{S})\mathbf{Y}^{\text{ds}}], \quad (2.23)$$

which represents the Eshelby-type equivalent strain and polarization to replace the electromechanical moduli of the product phase by the parent one, and $\mathbf{S}$ is the Eshelby electromechanical $\mathbf{S}$ -tensor [73]. For detailed calculation of $\mathbf{S}$ involving transversely isotropic matrix, one may refer to Li [74].

(ii) Thermodynamic driving force and kinetic equations

The thermodynamic driving force f_{drive} can be determined from $f_{\text{drive}} = -\partial\Delta G/\partial c_1 - f_s$, where f_s is the resistance force due to the depolarization field [72]. It can be calculated as

$$f_s = \frac{1}{2}\mathbf{Y}^{\text{dsT}}\mathbf{C}_s\mathbf{Y}^{\text{ds}}, \quad (2.24)$$

Following equation (2.16), we arrive at

$$f_{\text{drive}} = \frac{1}{2}(\bar{\mathbf{X}}^T\mathbf{A}_f\bar{\mathbf{X}} + \bar{\mathbf{X}}^T\mathbf{B}_f\mathbf{Y}^{\text{ds}} + \mathbf{Y}^{\text{dsT}}\mathbf{C}_f\mathbf{Y}^{\text{ds}}) - \frac{1}{2}\mathbf{Y}^{\text{dsT}}\mathbf{C}_s\mathbf{Y}^{\text{ds}}, \quad (2.25)$$

where $\mathbf{A}_f$, $\mathbf{B}_f$, $\mathbf{C}_f$ and $\mathbf{C}_s$ are shown in appendix A. Noting that, at $c_1=0$, this quantity is not necessary zero. It follows that the effective thermodynamic driving force for the growth of product domain can be identified as

$$f_{\text{drive}}^e = f_{\text{drive}} - f_{\text{drive}}|_{c_1=0}. \quad (2.26)$$

Under the action of the driving force, the product domain will continue to evolve at the expense of the parent one. Its growth can be described by a kinetic equation. During the 0° – 90° switch, the kinetic equation can be expressed in a Lorentzian-like function [71]

$$\frac{dc_1}{df_{\text{drive}}} = \frac{\eta}{(1 - f_{\text{drive}}/\lambda_a)^2 + (f_{\text{drive}}/\lambda_b)^2}, \quad (2.27)$$

where η , λ_a and λ_b are material constants that control the growth of c_1 with respect to the thermodynamic driving force. The driving force increase as the applied electric field or strain increases.

(c) Magnetoelectric coupling effect

Assuming a perfect bonding interface, the ferromagnetic and ferroelectric layers undergo the same displacement, so the magnetostrictive and the ferroelectric layers have the same strain, with $\varepsilon_1^m = \varepsilon_1^p$, $\varepsilon_2^m = \varepsilon_2^p$. In the thickness direction, the stress is continuous ($\sigma_3^m = \sigma_3^p$). Because the magnetic field is applied along the length direction, the shear strains are zero. Without the constraint of the ferroelectric layer, the strain in the magnetic phase, ε^m , can be calculated directly

Figure 4. The elongation of the magnetostrictive material. (Online version in colour.)

from equation (2.1). It is equal to λ under a magnetic field. Compared with the elongation of a free ferromagnetic layer, the elongation of the ME composite is actually shorter due to the constraint from the ferroelectric phase. For example, if the elongation of a single magnetostrictive layer is Δl_1 under the magnetic field H_1 (figure 4*a,b*), the elongation is Δl_2 in the ME composite (figure 4*a,c*). As both ferromagnetic and ferroelectric layers are nonlinear, the determination of the final length is more complicated in comparison with two pure elastic phases. But to provide some insights, we can consider the ME structure as a nonlinear mass-spring-damper mechanical system. Then the difference $\Delta l_1 - \Delta l_2$ is caused by the damping of the structure. This damping signifies the interfacial resistance between the two phases. To illustrate it further, we take the elongation of the composite structure Δl_2 to be equivalent to that of a single ferromagnetic material subjected to a pair of resistance force F_c , as shown in figure 4*d*.

Here $F_c = 0$ represents an ideal condition that the structure is undamped, or without the mutual constraint. The bigger the absolute value of F_c , the larger the damping of the structure. This F_c will provide a prestress σ_0 on the ME structure, that is $\sigma_0 = F_c / A_m$, A_m being the cross-sectional area of the magnetostrictive layer. By denoting the magnetostrictive strains in figure 4*b* and *d* as $\lambda_1|_{\sigma_0=0}$ and $\lambda_1|_{\sigma_0 \neq 0}$, respectively, the relationship between F_c and λ can be established as

$$E_s \lambda_1|_{\sigma_0=0} A_m - F_c = E_s \lambda_1|_{\sigma_0 \neq 0} A_m, \quad (2.28)$$

where E_s is the secant modulus of the ferromagnetic material at the current state of stress. Then σ_0 can be expressed as $|\sigma_0| = E_s |\lambda_1|_{\sigma_0=0} - \lambda_1|_{\sigma_0 \neq 0}|$. The quantity, σ_0 , in general can be used to analyse the influence of the interfacial resistance between the ferromagnetic and ferroelectric layers on various characteristics of ME coupling of the multiferroic composites. Then following equation (2.6), the magnetostrictive strain after considering this resistance is calculated as

$$\lambda_1|_{\sigma_0} = -\frac{\tilde{B}}{M_s^2} (T - T_r) \tilde{M}^2 + \begin{cases} \frac{[1 - \tanh(\sigma_0/\sigma_s)] \lambda_s}{M_s^2} \tilde{M}^2 & \left(\frac{\sigma_0}{\sigma_s} \geq 0 \right) \\ \frac{[2 - \tanh(2\sigma_0/\sigma_s)] \lambda_s}{2M_s^2} \tilde{M}^2 & \left(\frac{\sigma_0}{\sigma_s} \leq 0 \right) \end{cases}, \quad (2.29)$$

With this magnetostrictive strain transmitted to the ferroelectric phase, the induced electric field, E_3 , can be calculated from equation (2.19). The magnetization $\tilde{M}$ is directly related to the applied magnetic field, H_1 , in equation (2.9). Then the ME field coefficient α_E , can be obtained by $\alpha_E = \delta E_3 / \delta H_1$.

Table 1. The material parameters for Terfenol-D.

constants	λ_s (ppm)	M_s (A m ⁻¹)	σ_s (MPa)	$\tilde{B}$ (°C ⁻¹)	η	χ_m	$\tilde{c}$	ρ (Ωm)	K (A m ⁻¹)	H_0 (A m ⁻¹)	ν_s
values	1300	8.5×10^5	200	2.5×10^{-6}	0.03	18	0.1	4.3×10^{-6}	6000	0.03	0.3

3. Results and discussion

In order to illustrate the validity of the theoretical model and to reflect the domain change in the ferroelectric layers, we choose Terfenol-D as the ferromagnetic material and PZT as the ferroelectric material to compare with existing experiments [33,75]. Because the experiments were carried out at room temperature, we let $T = 30^\circ\text{C}$. In all calculations, we also adopted the open circuit boundary condition $D_3 = 0$ to be in line with the experiments.

We first discuss the nonlinear properties of magnetostrictive and ferroelectric phases individually, and then examine the calculated ME coupling effects for the multiferroic composites. The influence of internal resistance as characterized by σ_0 is investigated at the end.

(a) The nonlinear properties of constituent materials

To comply with the experimental conditions [75], the material parameters of Terfenol-D are given in table 1. Figure 5*a* shows the magnetization as a function of the applied magnetic field with $\sigma_0 = 0$. It can be seen that the magnetization first increases then tends to be stable with the increase of the magnetic field. The magnetization curve has strong nonlinear hysteretic characteristics. Then figure 5*b* displays the comparison of magnetostrictive strain hysteresis loops between experimental measurements and theoretical predictions. Here $\sigma_0 = 0$ corresponds to the free boundary condition for the single Terfenol-D layer in experiments [75]. As is evident in figure 5*b*, the nonlinear constitutive equations of ferromagnetic material adopted in this paper can accurately trace out the experimental hysteresis loops. This shows that selected calculation parameters are reasonable for Terfenol-D.

For ferroelectric materials, the material parameters of PZT are shown in table 2 [72]. The electromechanical compliance tensor **M** changes when the domain changes from one state to another. Now we discuss the components of **M** as a function of the volume concentration, c_1 , of the product phase. Here we discuss two domain switches, one from 180° to 90° and the other from 90° to 0° , as shown in figure 6. The domain wall is 45° for 180° – 90° domain switch and is -45° for 90° – 0° domain switch.

Figure 7 shows the variations of the nine independent components of the effective electromechanical compliance tensor as a function of c_1 in 180° – 90° and 90° – 0° domain switch processes for PZT. Here figure 7*a*–*c* are the flexibility coefficients, figure 7*d*–*f* are the dielectric coefficients, and figure 7*g*–*i* are the piezoelectric coefficients. Such variations reflect the continuous change of the electromechanical properties of the ferroelectric material in the course of two domain switch. Obviously, the material parameters change nonlinearly when the domain grows. The trends of change are not the same for different material parameters. For example, when the domains transform from 90° to 0° , M_{11} decreases first and then increases. It is the opposite for M_{13} , but for M_{91} , it decreases nonlinearly.

Next, we present the relationship between the electric displacement and strain of ferroelectric materials with applied electric field by using the thermodynamics-based micromechanics approach. In order to describe the trend of the hysteresis loop of ferroelectric materials, we compare the predicted results with Viola *et al.*'s experiments for PZT [76]. The applied electric field is along 3-direction.

Figure 8*a* shows the calculated D_3 versus E_3 results and the experimental data. Figure 8*b* shows the calculated strain along the 3-direction versus E_3 results and the experimental data, which display a butterfly shape. Both the electrical displacement vector and the strain exhibit hysteresis characteristics. The theoretical results agree well with the experimental results; it indicates that the

Figure 5. (a) Theoretical predictions of magnetic hysteresis loop. (b) Comparison of magnetostrictive strain hysteresis loops between experimental measurements and theoretical predictions with $\sigma_0 = 0$. (Online version in colour.)

Figure 6. (a) 180°–90° and (b) 90°–0° domain switch in ferroelectric materials. (Online version in colour.)

Table 2. Elastic, dielectric and piezoelectric coefficients for PZT in 0° domain at room temperature (s in $10^{-12} \text{ m}^2 \text{ N}^{-1}$, $k_0 = 8.85 \times 10^{-12} \text{ F m}^{-1}$, d in $10^{-12} \text{ C N}^{-1}$ and P_s in C m^{-2}).

constants	s_{11}	s_{12}	s_{13}	s_{33}	s_{44}	s_{66}	k_{11}/k_0	k_{33}/k_0	d_{15}	d_{31}	d_{33}	a/a_0	c/a_0	P_s
PZT	16.4	−5.74	−7.22	18.8	47.5	44.3	1730	1700	584	−171	250	0.9985	1.0003	0.38

nonlinear constitutive model of ferroelectric materials used in this paper can perfectly describe the hysteresis characteristics of ferroelectric materials.

(b) The nonlinear ME effect of ME composites

Then we calculate the ME effect of the ME composite. For the L-T mode ME composites, the polarization electric field along the thickness direction is increased from zero. That is to say, the

Figure 7. The electromechanical compliance tensor as the function of c_1 for ferroelectric materials in 180° – 90° and 90° – 0° domain switch. (Online version in colour.)

Figure 8. (a) Comparison of electric displacement and (b) strain hysteresis loops between experimental measurements [76] and theoretical predictions for PZT. (Online version in colour.)

polarization along the 3-direction is zero at the initial moment. So, we set 90° domain as the original state and 0° domain as the product phase in the calculation. The materials parameters η , λ_a and λ_b from 90° to 0° domain are $5.32 \times 10^{-6} \text{ (J m}^{-3}\text{)}^{-1}$, $9.93 \times 10^5 \text{ J m}^{-3}$ and $1.29 \times 10^6 \text{ J m}^{-3}$.

First we discuss the change of Gibbs free energy and the volume concentrations of product phase in the process of ME coupling. The results are shown in figure 9. Figure 9a–c shows ΔG_A , ΔG_B and ΔG_C change with the applied magnetic field. It can be seen that ΔG_A and ΔG_B have nonlinear hysteretic characteristics under the action of periodic magnetic fields. The order of ΔG_A is about the same as that of ΔG_B , but due to the domain compatibility between

Figure 9. (a) ΔG_A , (b) ΔG_B , (c) ΔG_C , (d) f_{drive}^e and (e) c_1 versus the applied magnetic field. (Online version in colour.)

the parent and product domain, ΔG_C is clearly zero at an arbitrary magnetic field. So the change of Gibbs free energy mainly comes from ΔG_A and ΔG_B . ΔG_A and ΔG_B , respectively, are $c_1 \tilde{\mathbf{X}}^T \mathbf{A} \tilde{\mathbf{X}}/2$ and $c_1 \tilde{\mathbf{X}}^T \mathbf{B} \mathbf{Y}^{\text{ds}}/2$ which are related to the electromechanical heterogeneity and energy dissipation, respectively, in piezoelectric layers. Their variations show a similar trend as with the magnetostrictive strain (figure 5b). Figure 9d displays the relationship between the effective thermodynamic driving force and the applied magnetic field. Figure 9e displays the relationship between the volume concentration of 0° domain with the applied magnetic field. The overall growth rate of 0° domain appears to be quick initially and then shows a slower trend.

Now we turn to the polarization electric field and the ME coefficient. Figure 10a shows the curve of E_3 versus H_1 . It can be seen that the polarization electric field also exhibits the hysteretic characteristics. It increases linearly at first, and then stabilizes with the increase of the applied magnetic field. Figure 10b shows the comparison of the theoretically predicted values of ME coefficients with experiments. The experimental data shown here are taken from Zhang *et al.* [75]. It can be seen that the theoretical results are in close agreement with the experimental data. This serves to validate the proposed theory for the ME effect of the ME composite.

(c) The influence of resistances on the ME effect

After that, the influence of resistances on the ME effect are investigated. As mentioned in §2, σ_0 can be used to describe the interfacial resistance. Here we investigate its effects as σ_0 changes from 0 to -50 MPa. The bigger the absolute value of σ_0 is, the larger the interfacial resistance of the structure will be. Here the applied magnetic field is larger enough to ensure that Terfenol-D can reach the saturation condition under different resistances.

Figure 11a is the magnetization hysteresis loops of the Terfenol-D layer under different initial prestress. It can be seen that the magnetizations reach to the saturation value M_s when the applied magnetic field is larger enough. But the initial prestress makes the magnetization process more difficult along the length direction. Figure 11b shows the influence of initial prestresses on the magnetostrictive strain of the Terfenol-D layer. The magnetostrictive strain decreases with increasing compressive stress at the low magnetic field, but it reaches different saturation values at the larger magnetic fields. The crossover is caused by the different effect of the initial prestress at different levels of the applied field on Terfenol-D [55]. The polarization field of the PZT layer has similar variation trend with the magnetostrictive strain as the initial prestress changes, as

Figure 10. (a) E_3 versus H_1 and (b) comparison of ME coefficient between Zhang *et al.*'s experimental data [75] and theoretical predictions with $\sigma_0 = -5$ MPa. (Online version in colour.)

Figure 11. The influence of resistances on each physical quantity in Terfenol-D/PZT composite. (Online version in colour.)

shown in figure 11c. Figure 11d displays the influence of the interfacial resistance on α_E . It can be seen that the peak values of α_E decreases gradually with the increase of σ_0 . It is clear that the required magnetic field to achieve both peak values also increases. These results signify that the interfacial resistance tends to hinder the attainment of a strong ME coupling. It will require higher magnetic field to reach for the multiferroic composite.

(d) The nonlinear converse ME effect of ME composites

Finally, the converse ME effect of ME composites is studied. we compare the predicted results with experiments of Terfenol-D/PZT composites conducted by Xu *et al.* [38]. In their experiment,

Figure 12. Comparison of (a) electric displacement D_3 and (b) strain (ε_1^p) hysteresis loops between experimental measurements [38] and theoretical predictions for PZT. (Online version in colour.)

Table 3. Elastic, dielectric and piezoelectric coefficients for PZT in 0° domain at room temperature (s in $10^{-12} \text{ m}^2 \text{ N}^{-1}$, $k_0 = 8.85 \times 10^{-12} \text{ F m}^{-1}$, d in $10^{-12} \text{ C N}^{-1}$ and P_s in C m^{-2}).

constants	s_{11}	s_{12}	s_{13}	s_{33}	s_{44}	s_{66}	k_{11}/k_0	k_{33}/k_0	d_{15}	d_{31}	d_{33}	a/a_0	c/a_0	P_s
PZT	16.4	−5.74	−7.22	18.8	47.5	44.3	1730	1300	584	−210	250	0	1.00025	0.35

Table 4. The material parameters for Terfenol-D.

constants	λ_s (ppm)	M_s (A m^{-1})	σ_s (MPa)	$\tilde{B}$ ($^\circ\text{C}^{-1}$)	η	χ_m	$\tilde{c}$	ρ (Ωm)	K (A m^{-1})	H_0 (A m^{-1})
values	890	6.2×10^5	200	2.5×10^{-6}	0.072	6	0.1	4.3×10^{-6}	6000	0.03

the direction of the applied electric field is along the thickness direction of the PZT layer. Under such an electric field, the ferroelectric material will be deformed. The material parameters of the PZT selected in the theoretical calculation are shown in table 3. Figure 12a shows the calculated D_3 versus E_3 results and the experimental data. Figure 12b shows the calculated in-plane strain ε_1^p versus E_3 results and the experimental data, which display a butterfly shape. The calculated nonlinear D_3 versus E_3 and ε_1 versus E_3 relations are in agreement with the test data.

The material parameters of the Terfenol-D selected in the theoretical calculation are shown in table 4. Figure 13a shows the calculated magnetization $\tilde{M}$ versus H_1 . Figure 13b shows the calculated magnetostrictive strain versus H_1 results and the experimental data. It can be seen that the theoretical calculation results are in agreement with the experimental results. These material properties of PZT and Terfenol-D are then used in the theory to calculate the converse ME effect. In this process, the deformation of Terfenol-D is transmitted through the interface layer of the ME composites. According to the converse ME effect (or Villarian effect) of the ferromagnetic material, the magnetization of the material will change when the ferromagnetic material is deformed,

Figure 13. (a) Theoretically predicted curve of $\tilde{M}$ versus H_1 . (b) Comparison of λ_1 versus H_1 between experimental measurements [38] and theoretical predictions for Terfenol-D. (Online version in colour.)

Figure 14. (a) Theoretically predicted curve of $\tilde{M}$ versus E_3 . (b) Comparison of H_1 versus E_3 between experimental measurements [38] and theoretical predictions for ME composites. (Online version in colour.)

thereby generating an excitation magnetic field [77]. Here, the calculation of magnetization along the longitudinal direction uses the following formula:

$$\tilde{M} = \begin{cases} \left[\frac{\lambda_1}{-\tilde{B}(T - T_r)/M_s^2 + \lambda_s/M_s^2} \right]^{1/2} & (\varepsilon_1^p \geq 0) \\ - \left[\frac{|\lambda_1|}{-\tilde{B}(T - T_r)/M_s^2 + \lambda_s/M_s^2} \right]^{1/2} & (\varepsilon_1^p < 0) \end{cases}, \quad (3.1)$$

and the excitation magnetic field is calculated by equation (2.7). Figure 14a,b displays the magnetization and the excited magnetic field caused by converse ME effect. It can be seen that the magnetization and the excited magnetic field have similar curve characteristics and have hysteresis characteristics. The theoretical calculation result can qualitatively reflect the converse ME effect of ME composites.

4. Concluding remarks

In this study, the ME coupling of multiferroic composites has been investigated by considering the nonlinear characteristics of ferromagnetic and ferroelectric materials. We started out by consideration of the constitutive behaviours of the ferromagnetic and ferroelectric phases. In the former, the magnetostrictive strain and magnetization are specified, and, in the latter, the change of free energy, the thermodynamic driving force and the kinetic equations of domain growth are discussed. The principle for the calculation of the ME coupling coefficient is then presented. The developed theory is then applied to study various characteristics of coupling effects for Terfenol-D/PZT composites. We have demonstrated the validity of the constitutive properties of Terfenol-D, and calculated the change of the electromechanical compliance tensor for the two ferroelectric materials when their domains change from one state to another. Then the polarization electric field and the ME coefficient are presented for Terfenol-D/PZT composites. The calculated ME coefficients have also been compared with experiments, and close agreement has been observed. Other features of the coupling process, such as the change of Gibbs free energy and the evolution of volume concentration of the 0° domain from the 90° one, have also been investigated. After that, the influence of interfacial resistance on the ME coupling effects are investigated. It shows that the interfacial resistance can hinder the attainment of large ME coupling coefficient, and it takes a higher applied magnetic field to reach their maximum values. Finally, the magnetization and the excited magnetic field caused by converse ME effect are calculated. Both of them have similar curve characteristics and have hysteresis characteristics. The theoretical calculation result can qualitatively reflect the converse ME effect of ME composites.

Data accessibility. All the data are displayed in the tables, figures and appendix.

Authors' contributions. J.Z. carried out the analytic modelling, analysed the data and drafted the manuscript; C.F. participated in data analysis and revised the manuscript; G.J.W. designed the analytic modelling, analysed the data and finalized the manuscript. All authors gave final approval for publication.

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Appendix A. The detailed expressions of B_1 , A , B , C , A_f , B_f , C_f and C_s

$$B_1 = [I + L_0(I - S)(M_1 - M_0)]^{-1}, \quad (A\ 1)$$

$$A = (M_1 - M_0)B_1Q, \quad (A\ 2)$$

$$B = I - c_0(M_1 - M_0)B_1QL_0(I - S) + (B_1Q)^T, \quad (A\ 3)$$

$$C = -c_0[B_1QL_0(I - S)]^T, \quad (A\ 4)$$

$$A_f = A - c_1(M_1 - M_0)B_1Q(B_1 - I)Q, \quad (A\ 5)$$

$$B_f = B + c_1\{-[B_1Q(B_1 - I)Q]^T + (M_1 - M_0)B_1Q[I + c_0(B_1 - I)Q]L_0(I - S)\}, \quad (A\ 6)$$

$$C_f = C + c_1[B_1Q[I + c_0(B_1 - I)Q]L_0(I - S)]^T, \quad (A\ 7)$$

$$C_s = [B_1QL_0(I - S)]^T. \quad (A\ 8)$$

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