MAGNETOELECTRIC COUPLING AND INTERFACE EFFECTS OF MULTIFERROIC COMPOSITES UNDER STRESS-PRESCRIBED BOUNDARY CONDITION

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Abstract. In multiferroic composites which typically consist of a piezoelectric and a piezomagnetic phase, magnetoelectric coupling is the most intriguing feature of their properties. In such composites, each constituent phase has either the electroelastic or the magnetoelastic coupling, but neither possesses the magnetoelectric coupling. The composite, however, has it. The magnitude of coupling is strongly dependent on the interface condition and the boundary condition. On the mechanical side this issue has been traditionally studied under the strain-prescribed boundary condition. In this paper we examine how magnetoelectric coupling arises under the stress-prescribed boundary condition, and with an imperfect interface. We examine a class of multiferroic composites containing aligned spheroidal inclusions, in which the matrix may be piezoelectric or piezomagnetic, and inclusions piezomagnetic or piezoelectric. We demonstrate how the magnetoelectric coupling coefficients depend on the phase properties, volume concentration, inclusion shape, and interface condition.

1. INTRODUCTION

Multiferroic composites are a class of composite materials which typically consist of a piezoelectric phase and a piezomagnetic phase. The former phase has the electroelastic coupling between the elastic and electric fields, while the latter has the magnetoelastic coupling between the elastic and magnetic fields. Many kinds of polycrystalline materials can exhibit such coupled behavior. For example, barium titanate (BaTiO₃, or in short, BTO) and lead zirconate titanate (commonly known as PZT) are two typical piezoelectric materials, while cobalt ferrite (CoFe₂O₄, or in short, CFO) is notably piezomagnetic. When these two types of materials are combined together into a composite, a new unique magnetoelectric coupling effect will emerge. It signifies the interaction between electric and magnetic fields, that is, an applied electric field can induce magnetization, and an applied magnetic field can generate electric polarization. This is a consequence of the electroelastic and magnetoelastic coupling in each phase, and the compatibility of their commonly possessed elastic strain. Since neither single piezoelectric nor piezomagnetic phase possesses such coupling characteristic, this is a so-called “0+0→1” product effect. This newly produced coupling represents the most intriguing property of multiferroic composites. It is often considered to be the figure of merit. When used for information stor-
age, this coupling allows data to be written electronically and read magnetically, thus avoiding the need to create a high local magnetic field to write.

The magneto-electric coupling effect is mathematically described by the magneto-electric coupling coefficient. Its determination involves many factors. In early studies it is recognized that the properties of each constituent phase, volume concentration, inclusion shape, and phase connectivity will influence this coefficient. To study the effect of each individual factor, it is necessary to develop a composite model for the overall multiferroic composites. To this end Nan [1] first developed a Green’s function approach to study BTO-CFO multiferroic composites, and later Benveniste [2] derived a set of explicit results for the effective properties of fibrous multiferroic composites. These are two important early contributions to the study of magneto-electric coupling and have important implications to the subsequent studies. It should be noted that the majority of subsequent studies are based on the micromechanical approach, in which a key issue is the availability of the components of Green’s function or Eshelby S-tensor [3] in the magneto-electro-elastic context. In classical micromechanics, this issue was limited to the pure elastic case. It was Chen [4], Dunn [5], Huang and Yu [6], and Dunn and Taya [7] who first extended the concept of Green’s function and Eshelby S-tensor to multiple coupled fields by studying the ellipsoidal inhomogeneity in piezoelectric composites. The procedures to derive the magneto-electric-elastic Eshelby S-tensor, as well as some explicit results for the S-tensor components of fibrous and lamellar composites, were later given by Huang and Kuo [8], Huang et al. [9], Li and Dunn [10], and recently by Wang et al. [11] and Wang and Weng [12]. These works represent some of the key developments in this area and have greatly facilitated the study of multiferroic composites.

Other important factors in the determination of magneto-electric coupling coefficients are the interface conditions and boundary conditions. The interface effect has always been an influential issue in the study of composite materials. It can significantly lower the overall properties of the composites. This issue has been extensively studied in the elastic context, as well as for thermal and electric conductivity. But for multiferroic composites, the issue of imperfect interface has only received very limited attention. Some notable works include those of Wang and Pan [13], Dinzart and Sabar [14], Kuo [15], and Yue and Xu [16], but all of them only focused on fibrous composites, not on general spheroidal inclusions. As to the issue of boundary conditions, it also has drawn only very limited attention. For example, Bichurin et al. [17] and Nan et al. [18] have compared the magneto-electric coupling of unclamped and rigidly clamped NiFe$_2$O$_4$-PZT bilayer composites, and found that there exists significant difference under these two different boundary conditions. So far there has been no general theoretical examination. As the elastic field always plays a crucial role in the mechanism of magneto-electric coupling, it can be expected that the influence of mechanical boundary conditions is noteworthy.

In retrospect, the effects of imperfect interface for a variety of inclusion shapes and phase connectivities have lately been investigated by Wang et al. [11] and Wang and Weng [19]. Even more recently the effect of strain- and stress-prescribed boundary conditions with a perfect interface has also been addressed by Wang and Weng [20]. However the influence of an imperfect interface under a stress-prescribed boundary condition has never been investigated. This is the major objective of this study. We will cover the entire range of inclusion shapes, from spheres, to fibers, to multilayers, and the general spheroidal shapes with prolate and oblate inclusions.

2. THE THEORY

2.1. Magneto-electro-elastic constitutive equations and the associated Eshelby S-tensor

The magneto-electro-elastic constitutive equations for multiferroic composites involve the coupled relations between the mechanical stress $\sigma_{ij}$ and strain $\epsilon_{ij}$ for the elastic field, the electric displacement $D_i$ and electric field $E_i$ for the electric part, and the magnetic flux density $B_i$ and magnetic field $H_i$ for the magnetic component. The most common constitutive setting, which has been adopted in most literature so far, is to take $(\epsilon_{ij}, E_i, H_i)$ as the applied fields and their counterparts $(\sigma_{ij}, D_i, B_i)$ as the responses. This setting has the merit that all the quantities in applied and responded fields follow the same mathematical behavior. In the absence of body force, free charge, and magnetic poles, the mechanical stress, electric displacement and magnetic flux density must satisfy the mechanical equilibrium and the quasi-static form of the Maxwell equations as

$$\sigma_{ij} = 0, \quad D_i = 0, \quad B_{ij} = 0. \quad (1)$$
On the other hand, the mechanical strain, electric field, and magnetic field are all derivable from the gradients of displacement, \( u_j \), electric potential \( \phi \) and magnetic potential \( \phi \), respectively, as

\[
\varepsilon_{ij} = \frac{1}{2} (u_{ij} + u_{ji}), \quad E_j = -\phi_j, \quad H_j = -\phi_j.
\]

Due to Eq. (2), a magneto-electro-elastic Green function is well-defined in this setting, which serves as the start point to evaluate the magneto-electro-elastic Eshelby S-tensor. In tensor representation, this constitutive setting can be cast into the matrix form as

\[
\begin{bmatrix}
\sigma \\
D \\
B
\end{bmatrix} = \begin{bmatrix}
C_{E,H} & e_i^T & q_i^E \\
e_i & -\kappa_{i,j} & -\alpha_i \\
q_i & -\alpha_{i} & -\mu_{i,E}
\end{bmatrix} \begin{bmatrix}
\varepsilon \\
E \\
H
\end{bmatrix}.
\]

The diagonal tensors, \( C_{E,H} \), \( \kappa_{i,j} \), and \( \mu_{i,E} \) are, respectively, the fourth-order elastic stiffness tensor (measured at constant electric and magnetic fields, as denoted by its subscript), second-order electric permittivity tensor (measured at constant strain and magnetic field), and second-order magnetic permeability tensor (measured at constant strain and electric field). The off-diagonal tensors, \( e_i \), \( q_i \), and \( \alpha_i \) are the third-order piezoelectric tensor (measured at constant magnetic field), third-order piezomagnetic tensor (measured at constant electric field), and second-order magnetoelectric coefficient tensor (measured at constant strain), respectively, with the superscript \( T \) denoting the transpose operation. For the convenience of calculation, Eq. (3) can be expressed in the Voigt and Nye contracted notations, as

\[
X^{(i)} = L^{(i)} Y^{(i)},
\]

where \( X^{(i)} \) and \( Y^{(i)} \) are both 12-dimensional vectors, such that

\[
X^{(i)} = [\sigma_1, \sigma_2, \sigma_3, \sigma_4, \sigma_5, D_1, D_2, D_3, B_1, B_2, B_3]^T,
\]

\[
Y^{(i)} = [e_1, e_2, e_3, e_4, e_5, e_6, -E_1 - E_2 - E_3 - H_1 - H_2 - H_3]^T,
\]

and \( L^{(i)} \) is the 12x12 magneto-electro-elastic moduli matrix, in which \( C_{E,H} \) is contracted to a 6x6 matrix, \( e_i \) and \( q_i \) to a 3x6 matrix, \( \kappa_{i,j} \) and \( \mu_{i,E} \) to a 3x3 matrix. To ensure that \( L^{(i)} \) can be cast in a diagonally symmetric form, we have chosen the negative values \( -E_i \) and \( -H_i \) in the \( Y^{(i)} \) vector. For a general transversely isotropic multiferroic composite with 3-direction as the symmetric axis and plane 1-2 isotropic, \( L^{(i)} \) carries the form

\[
L^{(i)} = \begin{bmatrix}
C_{11}^{E,H} & C_{12}^{E,H} & C_{13}^{E,H} & 0 & 0 & 0 & 0 & e_{31}^E & 0 & 0 & q_{31}^E \\
C_{12}^{E,H} & C_{12}^{E,H} & C_{13}^{E,H} & 0 & 0 & 0 & 0 & e_{32}^E & 0 & 0 & q_{32}^E \\
C_{13}^{E,H} & C_{13}^{E,H} & C_{13}^{E,H} & 0 & 0 & 0 & 0 & e_{33}^E & 0 & 0 & q_{33}^E \\
0 & 0 & 0 & C_{44}^{E,H} & 0 & 0 & 0 & e_{34}^E & 0 & 0 & q_{34}^E \\
0 & 0 & 0 & 0 & C_{44}^{E,H} & 0 & 0 & e_{35}^E & 0 & 0 & q_{35}^E \\
0 & 0 & 0 & 0 & 0 & C_{44}^{E,H} & 0 & 0 & e_{36}^E & 0 & 0 \\
0 & 0 & 0 & e_{34}^E & 0 & 0 & -\kappa_{11}^{E,H} & 0 & 0 & -\alpha_{1}^E & 0 \\
0 & 0 & 0 & e_{35}^E & 0 & 0 & 0 & -\kappa_{12}^{E,H} & 0 & 0 & -\alpha_{1}^E \\
0 & 0 & 0 & e_{36}^E & 0 & 0 & 0 & 0 & -\kappa_{13}^{E,H} & 0 & 0 & -\alpha_{1}^E \\
e_{31}^E & e_{32}^E & e_{33}^E & 0 & 0 & 0 & 0 & 0 & -\kappa_{11}^{E,H} & 0 & 0 & -\alpha_{1}^E \\
e_{32}^E & e_{32}^E & e_{33}^E & 0 & 0 & 0 & 0 & 0 & 0 & -\kappa_{12}^{E,H} & 0 & 0 & -\alpha_{1}^E \\
e_{33}^E & e_{33}^E & e_{33}^E & 0 & 0 & 0 & 0 & 0 & 0 & 0 & -\kappa_{13}^{E,H} & 0 & 0 & -\alpha_{1}^E \\
q_{31}^E & q_{32}^E & q_{33}^E & 0 & 0 & 0 & 0 & -\alpha_{1}^E & 0 & 0 & -\mu_{11}^{E} \\
q_{32}^E & q_{32}^E & q_{33}^E & 0 & 0 & 0 & 0 & 0 & -\alpha_{1}^E & 0 & 0 & -\mu_{11}^{E} \\
q_{33}^E & q_{33}^E & q_{33}^E & 0 & 0 & 0 & 0 & 0 & 0 & -\alpha_{1}^E & 0 & 0 & -\mu_{11}^{E}
\end{bmatrix}
\]

with 17 independent material constants (note that \( C_{44}^{E,H} = (C_{11}^{E,H} - C_{12}^{E,H}) / 2 \)). But for a transversely isotropic piezoelectric phase, its property does not have piezomagnetic \( q_i \) terms, while a transversely isotropic
piezomagnetic phase has no piezoelectric $e_{ij}$ terms. Also neither of them has the magnetoelectric coupling coefficient $\alpha_{ij}$ terms.

The above constitutive setting is suitable for evaluating the overall properties of multiferroic composites when they are under strain-prescribed boundary conditions, but when we have stress-prescribed boundary condition, another setting with $(\sigma_{ij}, E_i, H_i)$ as the applied fields and $(\epsilon_{ij}, D_i, B_i)$ as responses is more appropriate, such that

$$
\begin{bmatrix}
\varepsilon
\end{bmatrix} =
\begin{bmatrix}
S_{E,H} & d_{ij} & p_{E}^T
\end{bmatrix}
\begin{bmatrix}
\sigma
\end{bmatrix},
$$

where the diagonal tensors, $S_{E,H}$, $\kappa_{\sigma,H}$ and $\mu_{\sigma,E}$ are, respectively, the fourth-order elastic compliance tensor (measured at constant electric and magnetic fields), second-order electric permittivity tensor (measured at constant stress and magnetic field), and second-order magnetic permeability tensor (measured at constant stress and electric field). The off-diagonal tensors, $d_{ij}$ and $p_{E}^+$ are another form of third-order piezoelectric tensor (measured at constant magnetic field) and piezomagnetic tensor (measured at constant electric field), and $\alpha_{ij}$ is the second-order magnetoelectric coupling coefficient tensor (measured at constant stress). In the Voigt and Nye contracted notations, this setting can be expressed as

$$
X^{(1)} = L^{(1)}Y^{(1)},
$$

where 12-dimensional $X^{(2)}$ and $Y^{(2)}$ hold the form

$$
X^{(2)} = 
\begin{bmatrix}
\epsilon_1 & \epsilon_2 & \epsilon_3 & \epsilon_4 & \epsilon_5 & \epsilon_6 & D_1 & D_2 & D_3 & E_1 & E_2 & E_3
\end{bmatrix}^T,
$$

and similarly, $L^{(2)}$ is another $12 \times 12$ magneto-electro-elastic moduli matrix, consisting of a $6 \times 6$ matrix $S_{E,H}^+$, a $3 \times 6$ matrix $d_{ij}^+$ and $p_{E}^+$, a $3 \times 3$ matrix $\kappa_{\sigma,H}^+$, $\mu_{\sigma,E}^+$ and $\alpha_{ij}^+$. The general transversely isotropic $L^{(2)}$ is given by

$$
L^{(2)} =
\begin{bmatrix}
S_{11}^{E,H} & S_{12}^{E,H} & S_{13}^{E,H} & 0 & 0 & 0 & 0 & d_{11}^{H} & 0 & 0 & p_{11}^{E}
S_{12}^{E,H} & S_{22}^{E,H} & S_{23}^{E,H} & 0 & 0 & 0 & 0 & d_{22}^{H} & 0 & 0 & p_{22}^{E}
S_{13}^{E,H} & S_{23}^{E,H} & S_{33}^{E,H} & 0 & 0 & 0 & 0 & d_{33}^{H} & 0 & 0 & p_{33}^{E}
0 & 0 & 0 & S_{44}^{E,H} & 0 & 0 & 0 & d_{44}^{H} & 0 & 0 & p_{44}^{E}
0 & 0 & 0 & S_{55}^{E,H} & 0 & 0 & 0 & d_{55}^{H} & 0 & 0 & p_{55}^{E}
0 & 0 & 0 & S_{66}^{E,H} & 0 & 0 & 0 & d_{66}^{H} & 0 & 0 & p_{66}^{E}
0 & 0 & 0 & 0 & S_{11}^{E,H} & 0 & 0 & 0 & \kappa_{11}^{H} & 0 & 0 & \mu_{11}^{E}
0 & 0 & 0 & 0 & S_{22}^{E,H} & 0 & 0 & 0 & \kappa_{22}^{H} & 0 & 0 & \mu_{22}^{E}
0 & 0 & 0 & 0 & S_{33}^{E,H} & 0 & 0 & 0 & \kappa_{33}^{H} & 0 & 0 & \mu_{33}^{E}
d_{11}^{H} & d_{22}^{H} & d_{33}^{H} & 0 & 0 & 0 & 0 & \kappa_{11}^{E} & 0 & 0 & \mu_{11}^{E}
d_{44}^{H} & d_{55}^{H} & d_{66}^{H} & 0 & 0 & 0 & 0 & \kappa_{44}^{E} & 0 & 0 & \mu_{44}^{E}
d_{11}^{E} & d_{22}^{E} & d_{33}^{E} & 0 & 0 & 0 & 0 & \kappa_{11}^{E} & 0 & 0 & \mu_{11}^{E}
0 & 0 & 0 & p_{11}^{E} & 0 & 0 & 0 & \alpha_{11}^{E} & 0 & 0 & \mu_{11}^{E}
0 & 0 & 0 & p_{22}^{E} & 0 & 0 & 0 & \alpha_{22}^{E} & 0 & 0 & \mu_{22}^{E}
0 & 0 & 0 & p_{33}^{E} & 0 & 0 & 0 & \alpha_{33}^{E} & 0 & 0 & \mu_{33}^{E}
\end{bmatrix},
$$

also with 17 independent material constants (note that $S_{66}^{E,H} = 2(S_{11}^{E,H} - S_{12}^{E,H})$). As has been discussed in our previous work with perfect interface, the magnetoelectric coupling coefficients under stress-prescribed boundary conditions, $\alpha_{ij}^+$ and $\alpha_{ij}^+$, are significantly different from those under strain-prescribed boundary conditions, $\alpha_{ij}^+$ and $\alpha_{ij}^+$. For a stress-controlled process, $\alpha_{ij}^+$ and $\alpha_{ij}^+$ of the composite are the figures of merits which we will explore now.

As pointed out in Wang and Weng [20], the two magneto-electro-elastic moduli matrices $L^{(1)}$ and $L^{(2)}$ are related. $L^{(2)}$ can be transformed from $L^{(1)}$, by
\[
\begin{bmatrix}
S_{E,H} & d_E^T & p_E^T \\
d_H & \kappa_{\alpha,H} & \alpha_\alpha \\
p_E & \alpha_\alpha & \mu_{\alpha,E}
\end{bmatrix} =
\begin{bmatrix}
I & 0 & 0 \\
e_H & -\kappa_{\alpha,H} & -\alpha_\alpha \\
q_E & -\alpha_\alpha & -\mu_{\alpha,E}
\end{bmatrix}^{-1}
\begin{bmatrix}
C_{E,H} & -e_H^T & q_E^T \\
e_H & -\kappa_{\alpha,H} & -\alpha_\alpha \\
q_E & -\alpha_\alpha & -\mu_{\alpha,E}
\end{bmatrix}
\begin{bmatrix}
I & 0 & 0 \\
-1 & 0 & 0 \\
0 & 0 & -1
\end{bmatrix},
\]

where \(I_3\) and \(I_6\) are 3- and 6-dimensional identity matrices, and 0 stands for zero matrix. Likewise, \(L^{(2)}\) can transform back to \(L^{(1)}\), in a similar way.

\[
\begin{bmatrix}
C_{E,H} & e_E^T & q_E^T \\
e_E & -\kappa_{\alpha,E} & -\alpha_\alpha \\
q_E & -\alpha_\alpha & -\mu_{\alpha,E}
\end{bmatrix} =
\begin{bmatrix}
I & 0 & 0 \\
-1 & 0 & 0 \\
0 & 0 & -1
\end{bmatrix}^{-1}
\begin{bmatrix}
S_{E,H} & d_E^T & p_E^T \\
d_E & \kappa_{\alpha,E} & \alpha_\alpha \\
p_E & \alpha_\alpha & \mu_{\alpha,E}
\end{bmatrix}
\begin{bmatrix}
I & 0 & 0 \\
-1 & 0 & 0 \\
0 & 0 & -1
\end{bmatrix}
\]

Each moduli matrix is associated with a magneto-electro-elastic Eshelby S-tensor. For convenience the evaluation of the associated S-tensor is presented in the Appendix.

### 2.2. The effective properties of multiferroic composites with a perfect and an imperfect interface

The microstructure of the present multiferroic composites is taken to be of the inclusion-matrix type, as depicted in Figs. 1a-1d, where piezomagnetic inclusions are embedded in a piezoelectric matrix, or vice versa. With a perfect interface, the effective properties of this class of composites can be most conveniently evaluated by the Mori-Tanaka method [21]. By taking the matrix as phase 0 and inclusions as phase 1, and denoting their volume concentrations as \(c_0\) and \(c_1\), and phase moduli as \(L_0\) and \(L_1\), respectively, the effective magneto-electro-elastic moduli matrix \(L_e\) can be calculated from [22,23]

\[
L_e = L_0 + c_1 (L_1 - L_0) \left[ I + c_0 S_{int} (L_1 - L_0) \right]^{-1},
\]

where \(S_{int}\) is the Eshelby S-tensor of the inclusion phase. It is dependent on the property of the matrix phase, and the shape of the ellipsoidal inclusions, which is governed by the aspect ratio (length-to-diameter ratio). In this study the moduli matrices and S-tensor are all in the form which has \((\sigma, E, H)\) as the applied fields.

With an imperfect interface, we first assume it to exist as a very thin layer of interface between the ellipsoidal inclusions and the matrix [24]. This thin layer and the piezoelectric (or piezomagnetic) inclusion are then taken to form a thinly-coated inclusion, in which the volume concentration of the interphase is denoted as \(c_{int}\), and that of the inclusion as \((1 - c_{int})\). We further assume that the interphase is amorphous, neither piezoelectric nor piezomagnetic, with an isotropic property, denoted by \(L_{int}\). The property of the transversely isotropic inclusion still has no magneto-electro-coupling. The effective property of this thinly-coated inclusion, denoted by \(L_{coat}\), then can be calculated from Eq. (13) again, as

\[
L_{coat} = L_{int} + (1 - c_{int}) (L_1 - L_{int}) \left[ I + c_{int} S_{int} (L_1 - L_{int})^{-1} (L_1 - L_{int}) \right]^{-1}.
\]

Since \(L_{int}\) is isotropic, it has only 4 independent material constants (2 elastic constants, 1 electric permittivity and 1 magnetic permeability). In the \((\varepsilon, E, H)\) constitutive setting, its moduli matrix \(L_{int}\) depends on \((C_{E,H}^{int}, \sigma_{E,H}^{int}, \kappa_{\alpha,H}^{int}, \mu_{\alpha,E}^{int})\) and has the following nonzero components: 

\[
\begin{align*}
L_{11}^{int} & = L_{22}^{int} = L_{66}^{int} = C_{11,H}^{int}, \\
L_{14}^{int} & = L_{44}^{int} = L_{66}^{int} = C_{44,H}^{int}, \\
L_{12}^{int} & = L_{23}^{int} = L_{32}^{int} = L_{13}^{int} = C_{12,H}^{int} = 2C_{44,H}^{int}, \\
L_{22}^{int} & = L_{33}^{int} = L_{44}^{int} = L_{66}^{int} = \kappa_{\alpha,H}^{int}, \\
L_{11}^{int} & = L_{11}^{int} = L_{22}^{int} = L_{33}^{int} = L_{44}^{int} = L_{55}^{int} = L_{66}^{int} = -\kappa_{\alpha,H}^{int}, \\
L_{11}^{int} & = L_{11}^{int} = L_{22}^{int} = L_{33}^{int} = -\mu_{\alpha,E}^{int}.
\end{align*}
\]

While in the \((\sigma, E, H)\) constitutive setting, \(L_{int}\) is determined by \((S_{11,H}^{int}, S_{44,H}^{int}, \sigma_{\alpha,H}^{int}, \kappa_{\alpha,H}^{int}, \mu_{\alpha,E}^{int})\) and its nonzero components are:

\[
\begin{align*}
L_{11}^{int} & = L_{22}^{int} = L_{33}^{int} = S_{11,H}^{int}, \\
L_{14}^{int} & = L_{44}^{int} = L_{66}^{int} = S_{44,H}^{int}, \\
L_{12}^{int} & = L_{23}^{int} = L_{32}^{int} = L_{13}^{int} = S_{12,H}^{int} = 2S_{44,H}^{int}, \\
S_{11,H}^{int} & = S_{44,H}^{int} = S_{66,H}^{int} = S_{12,H}^{int} = S_{23,H}^{int} = S_{32,H}^{int} = S_{13,H}^{int} = S_{24,H}^{int} = S_{34,H}^{int} = S_{66,H}^{int} = \kappa_{\alpha,H}^{int} = \mu_{\alpha,E}^{int}. \\
S_{11}^{int} & = \mu_{\alpha,E}^{int} = L_{11}^{int} = L_{22}^{int} = L_{33}^{int} = L_{44}^{int} = L_{55}^{int} = L_{66}^{int} = -\kappa_{\alpha,H}^{int} = -\mu_{\alpha,E}^{int}.
\end{align*}
\]

It should be noted that \(\kappa_{\alpha,H}^{int}\) and \(\mu_{\alpha,E}^{int}\) are equal, so we have \(\kappa_{\alpha,H}^{int} = \mu_{\alpha,E}^{int} = \kappa_{\alpha,E}^{int}\). Similarly we also have \(\mu_{\alpha,H}^{int} = \mu_{\alpha,E}^{int} = \mu_{\alpha,H}^{int}\).

The S-tensor \(S_{int}\) of the interphase can also be evaluated by the method given in the Appendix.

### 3. RESULTS AND DISCUSSION

#### 3.1. The effect of imperfect interface and inclusion concentration dependence

In numerical calculations the multiferroic composite is taken to consist of BTO as the piezoelectric phase and CFO as the piezomagnetic phase. Both CFO-in-BTO (matrix: BTO; inclusion: CFO) and
BTO-in-CFO (matrix: CFO; inclusion: BTO) configurations are considered. The aspect ratio of inclusions (length-to-diameter ratio) is denoted by scalar, \( \alpha \), which is not to be confused with tensor \( \alpha \) in bold face that means the magnetoelectric coefficient tensor. Moreover, \( \alpha \) with super and subscript represents the magnetoelectric coupling coefficient. The material constants of BTO and CFO are listed in Table 1, which have been widely used by Huang and Kuo [8], Li and Dunn [10], and others. These

**Table 1.** Material constants of BTO and CFO phase used in numerical calculations.

<table>
<thead>
<tr>
<th>Data source</th>
<th>Huang and Kuo (1997) and Li and Dunn (1998)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Phase</td>
<td>BTO</td>
</tr>
<tr>
<td>( C_{11}^{E,H} ) GPa</td>
<td>166</td>
</tr>
<tr>
<td>( C_{12}^{E,H} ) GPa</td>
<td>77</td>
</tr>
<tr>
<td>( C_{13}^{E,H} ) GPa</td>
<td>78</td>
</tr>
<tr>
<td>( C_{33}^{E,H} ) GPa</td>
<td>162</td>
</tr>
<tr>
<td>( C_{44}^{E,H} ) GPa</td>
<td>43</td>
</tr>
<tr>
<td>( e_{31}^{H} ) C/m²</td>
<td>-4.4</td>
</tr>
<tr>
<td>( e_{33}^{H} ) C/m²</td>
<td>18.6</td>
</tr>
<tr>
<td>( e_{45}^{H} ) C/m²</td>
<td>11.6</td>
</tr>
<tr>
<td>( q_{31}^{E} ) N/(A·m)</td>
<td>0</td>
</tr>
<tr>
<td>( q_{33}^{E} ) N/(A·m)</td>
<td>0</td>
</tr>
<tr>
<td>( q_{15}^{E} ) N/(A·m)</td>
<td>0</td>
</tr>
<tr>
<td>( \kappa_{11}^{E,H} ) C²/(N·m²)</td>
<td>( 11.2\times10^9 )</td>
</tr>
<tr>
<td>( \kappa_{33}^{E,H} ) C²/(N·m²)</td>
<td>( 12.6\times10^9 )</td>
</tr>
<tr>
<td>( \mu_{11}^{E} ) N/A²</td>
<td>( 5\times10^6 )</td>
</tr>
<tr>
<td>( \mu_{33}^{E} ) N/A²</td>
<td>( 10\times10^6 )</td>
</tr>
</tbody>
</table>

Fig. 1. The schematic plot of the microstructure of piezoelectric-piezomagnetic multiferroic composites: (a) the general prolate inclusions (aspect ratio \( \alpha >1 \)), (b) the general oblate inclusions (0< \( \alpha <1 \)), (c) the fibrous composites (\( \alpha =\infty \)), (d) the multilayers (\( \alpha =0 \)).
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The CFO inclusion concentration dependence of effective magnetoelectric coupling coefficient $\alpha_3^{\sigma}$ under (a) a perfect interface, and (b) an imperfect interface.

Fig. 2. The CFO inclusion concentration dependence of effective magnetoelectric coupling coefficient $\alpha_3^{\sigma}$.
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3.1. The CFO inclusion concentration dependence of effective magnetoelectric coupling coefficient $\alpha_{11}^E$ under (a) a perfect interface, and (b) an imperfect interface.

Fig. 3. The CFO inclusion concentration dependence of effective magnetoelectric coupling coefficient $\alpha_{11}^E$ under (a) a perfect interface, and (b) an imperfect interface.

since CFO has very weak electric permittivity. This phenomenon can also be observed in Figs. 5a and 5b for the $\alpha_{11}^E$. The weakening effect of an imperfect interface is also evident in these two sets of figures.

For BTO-in-CFO composites, the corresponding results are shown in Figs. 6-9. For $\alpha_{11}^E$ and $\alpha_{11}^H$, their behaviors are quite similar to those in Figs. 2 and 3. While for $\alpha_{33}^E$ and $\alpha_{33}^H$, the highest magnitudes shift to lower $c_1$ region, since now BTO is the inclusion phase and lower BTO concentration means smaller overall $\kappa_{33}^{H}$ and $\kappa_{33}^{H}$ value.

3.2. The effect of an elastically stiff and soft imperfect interface

As mentioned above, the elastic constants of the interphase does have an influence on the overall magnetoelectric coupling. To further reveal its effect, we choose an elastically stiff interface with $C_{11,\text{int}}^{E,H} = 20000$ GPa and $C_{44,\text{int}}^{E,H} = 5000$ GPa, and an elastically soft interface with $C_{11,\text{int}}^{E,H} = 3$ GPa and $C_{44,\text{int}}^{E,H} = 1$ GPa, to investigate to what extent will the effective magnetoelectric coupling coefficients be lowered. Here we take a CFO-in-BTO composite with inclusion aspect ratio $\alpha_{11}^E$ as an example. All the other parameters except for $C_{11,\text{int}}^{E,H}$ and $C_{44,\text{int}}^{E,H}$ remain the same as those in the previous calculation. It turns out that the weakening effect of elastically stiff and soft interface on the magnetoelectric coupling coefficients under strain- and stress-prescribed boundary conditions are totally opposite. As seen from Figs. 10a and 10b, under stress-prescribed boundary conditions, an elastically soft interface can significantly lower their magnitudes. On the other hand, under strain-prescribed boundary conditions, the influence of an elastically soft interface instead becomes greater than that of an elastically stiff interface, as shown in Figs. 11a and 11b. We keep on investigating this issue by letting the interface continuously change from elastically soft to stiff. Here the Young's modules and Poisson’s ratio of the interphase are taken as $E_{\text{int}}$ and $\nu_{\text{int}}$, so that they are related to the elastic constants by

$$
E_{\text{int}} = \frac{C_{11,\text{int}}^{E,H} (1-\nu_{\text{int}})}{1+\nu_{\text{int}} (1-2\nu_{\text{int}})},
$$

$$
C_{44,\text{int}}^{E,H} = \frac{E_{\text{int}}}{2(1+\nu_{\text{int}})}.
$$

(15)
Our focus is on varying $E_{\text{int}}$ with $v_{\text{int}}$ kept as a constant at 1/3. Since the elastic constants of BTO and CFO phase are mostly on the order of 100 GPa, this value is taken to be the reference for $E_{\text{int}}$. We then introduce a parameter $N$, to signify how many orders of magnitude that $E_{\text{int}}$ is greater or smaller than the reference value 100 GPa, so that $N = E_{\text{int}} / 100$ GPa. For a CFO-in-BTO composite with inclusion aspect ratio $\alpha = 10$, we calculate that when CFO volume concentration $c_1 = 0.5$, the ratio of effective magnetoelectric coupling coefficients under an imperfect interface to those under a perfect interface, at different $N$ values from $10^{-6}$ to $10^6$ (from very soft to very stiff). The results are displayed in Figs. 12a and 12b, for the strain and stress-prescribed boundary conditions, respectively. It can be clearly seen that around $N=1$ the weakening effect of an imperfect interface is very limited. When the interface is extremely soft (such as a void or crack) or extremely stiff, the effective magnetoelectric coupling coefficients will both drop to almost zero. But the behaviors for strain- and stress-prescribed boundary conditions are different. Under the stress-prescribed boundary condition, the magnetoelectric coupling coefficients drop faster with an elastically stiff interface, while under strain-prescribed boundary conditions the magnetoelectric coupling coefficients drop faster with an elastically interface. This comparison clearly points out that, the weakening effect of an imperfect interface depends on both its elastic constants and the prescribed boundary conditions.

4. CONCLUSION

In this paper we have developed a composite model to study the magnetoelectric coupling effect of multiferroic composites. In particular, we emphasize on the effect of imperfect interface under the stress-prescribed boundary condition on the effective magnetoelectric coupling coefficients $\alpha_{33}^a$ and $\alpha_{11}^a$, as
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Fig. 7. The BTO inclusion concentration dependence of effective magnetoelectric coupling coefficient $\alpha_{11}^\sigma$ under (a) a perfect interface, and (b) an imperfect interface.

Fig. 8. The BTO inclusion concentration dependence of effective magnetoelectric voltage coefficient $\alpha_{33}^E$ under (a) a perfect interface, and (b) an imperfect interface.

Fig. 9. The BTO inclusion concentration dependence of effective magnetoelectric voltage coefficient $\alpha_{11}^E$ under (a) a perfect interface, and (b) an imperfect interface.

Fig. 10. The influence of an elastically soft and stiff interface on the effective magnetoelectric coupling coefficient (a) $\alpha_{33}^\sigma$, and (b) $\alpha_{11}^\sigma$, for a CFO-in-BTO composite.
well as on the magnetoelectric voltage coefficients \( \alpha_{33}^{E} \) and \( \alpha_{11}^{E} \). We have shown that an imperfect interface can generally lower the magnitudes of these quantities. Furthermore we have also found out that an elastically stiff and soft interface will have different weakening effect under the strain- and the stress-prescribed boundary conditions. An elastically stiff interface has stronger weakening effect under the stress-prescribed boundary condition, whereas an elastically soft interface has greater influence under the strain-prescribed boundary condition.

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APPENDIX

A. The calculation of magneto-electro-elastic Eshelby S-tensor for \((\varepsilon,E,H)\) constitutive setting

The magneto-electro-elastic S-tensor has been studied by Li and Dunn, Huang et al., and several others. Here we briefly summarize the procedures to calculate this S-tensor for the \((\varepsilon,E,H)\) constitutive setting. This procedure can be applied to multiferroic composites with ellipsoidal inclusions embedded in transversely isotropic matrix, and the aspect ratio of inclusions can range from 0 to \( \infty \). It also requires that the symmetric axis of ellipsoidal inclusions must coincide with the symmetric axis of the transversely isotropic property of the matrix.

First, we define a pseudo material constant “tensor” (which is not a real tensor by rigorous definition) \( L_{iJMn} \) for the matrix phase of multiferroic composites, with subscript \( i,n=1-3 \) and \( J,M=1-5 \),

\[
L_{iJMn} = \begin{cases} 
C_{iJMn}, & J, M = 1, 2, 3, \\
e_{iJn}, & J = 1, 2, 3, M = 4, \\
q_{iJn}, & J = 1, 2, 3, M = 5, \\
e_{iMn}, & J = 4, M = 1, 2, 3, \\
q_{iMn}, & J = 5, M = 1, 2, 3, \\
-\varepsilon_{iJn}, & J = 4, M = 4, \\
-\varepsilon_{iJn}, & J = 4, M = 5 & J = 5, M = 4, \\
-P_{iJn}, & J = 5, M = 5. 
\end{cases} \tag{A1}
\]
With \( L_{\text{meff}} \) we introduce a 5\( \times \)5 matrix \( K_{\text{eff}} \)
\[
K_{\text{eff}} = L_{\text{meff}} X^T X,
\]
where \( x_i = [x_i, x_2, x_3]^T \). Then we define another pseudo tensor \( J_{\text{meff}} \)
\[
J_{\text{meff}}(x_1, x_2, x_3) = x_i x_j K_{\text{eff}}^{-1},
\]
so that it is a function of \( x_1, x_2, \) and \( x_3 \). Next we integrate \( J_{\text{meff}} \) over the volume of an ellipsoidal inclusion \( \Omega: x_1^2/a_1^2 + x_2^2/a_2^2 + x_3^2/a_3^2 \leq 1 \). When this spheroidal inclusion is symmetric about 3-direction, it satisfies \( a_1 = a_2, \alpha = a_3/a_1 \), where \( \alpha \) is the aspect ratio of inclusion. Hence the volume integral of \( J_{\text{meff}} \) can be written as
\[
H_{\text{meff}} = \int_{\Omega} J_{\text{meff}}(x_1/a_1, x_2/a_2, x_3/a_3) dV
= \int_{\Omega} J_{\text{meff}}(x_1, x_2, x_3/\alpha) dV,
\]
where the second equality is based on the fact that \( J_{\text{meff}} \) is a homogeneous function of order zero, thus multiplying all the variables by \( a \) will not affect the integral. The third equality is given by applying a change of variables from \( x_1, x_2, x_3 \) to
\[
y_1 = \sqrt{1-\tau^2} \cos \theta,
\]
\[
y_2 = \sqrt{1-\tau^2} \sin \theta,
\]
\[
y_3 = \tau,
\]
with \( \tau \in [-1,1] \) and \( \theta \in [0,2\pi] \). Finally the S-tensor is determined by
\[
S_{\text{meff}} = \begin{cases}
\frac{1}{8\pi} L_{\text{meff}} \left( J_{\text{meff}} + H_{\text{meff}} \right), & M = 1 - 3, \\
\frac{1}{4\pi} L_{\text{meff}} H_{\text{meff}}, & M = 4,
\frac{1}{4\pi} L_{\text{meff}} H_{\text{meff}}, & M = 5.
\end{cases}
\]

Still the magneto-electro-elastic S-tensor is a pseudo tensor. For convenience it is generally converted into a 12\( \times \)12 matrix by the Voigt and Nye contracted notations.

The integral in Eq. (A.4) can be analytically evaluated only when aspect ratio \( \alpha = 0 \) or \( \infty \). For a general spheroid, it can be numerically carried out by using Gaussian quadrature, which turns the definite integral into a weighted sum of function values at specified points within the domain of integration. Thus Eq. (A.4) can be rewritten as
\[
H_{\text{meff}} = \int_{\Omega} J_{\text{meff}}(y_1, y_2, y_3/\alpha) d\Omega
= \sum_{i=1}^{n} w_i f(\tau_i),
\]
where \( f(\tau) = \int_{\Omega} J_{\text{meff}}(y_1, y_2, y_3/\alpha) d\Omega \), \( n \) is the total number of specified points (usually \( n=20 \) or more will provide enough accuracy), \( w_i \) is the weight at each specified point which can be constructed by different kinds of weight function. Up to this point the magneto-electro-elastic S-tensor for constitutive setting \((\sigma,E,H)\) is determined.

**B. The calculation of magneto-electro-elastic Eshelby S-tensor for \((\sigma,E,H)\) constitutive setting**

With the magneto-electro-elastic Eshelby S-tensor \( S^{(1)} \) for constitutive setting \((\sigma,E,H)\), we can further construct the S-tensor for \((\sigma,E,H)\). First let \( L^{(3)} \) be the magneto-electro-elastic moduli matrix for \((\sigma,E,H)\), and \( L^{(3)} \) for \((\sigma,D,\beta)\), so that \( L^{(3)} = \mathbf{L}^{(3)} \). The S-tensor for \((\sigma,D,\beta)\) is denoted as \( S^{(3)} \). Let \( X = [\sigma,D,\beta]^T \) and \( Y = [\sigma,E,H]^T \). The generalized magneto-electro-elastic eigenstrain and eigenstress are denoted by \( ^* \). Therefore we have
\[
X = L^{(3)} \bigl(Y - Y^*\bigr), \quad Y = L^{(3)} \bigl(X - X^*\bigr),
\]
and
\[
Y = S^{(3)} Y^*, \quad X = S^{(3)} X^*.
\]
From Eq. (B.1) it can be concluded that
\[
X' = -L^{(3)} Y^*, \quad Y' = -L^{(3)} X^*.
\]
Then combine Eq. (B.1) to (B.3), \( S^{(3)} \) can be derived as
\[
S^{(3)} = L^{(3)} \left( I - S^{(3)} \right) L^{(3)}
\]
where \( I \) is the 12\( \times \)12 identity matrix. With \( S^{(1)} \) and \( S^{(3)} \), the S-tensor for constitutive setting \((\sigma,E,H)\), \( S^{(2)} \), can then be derived. First of all Eq. (B.1) can be rewritten as
\[
X = L^{(3)} \left(S^{(1)} - I\right) Y^*, \quad Y = L^{(3)} \left(S^{(1)} - I\right) X^*.
\]
By introducing two auxiliary tensors $T^{(1)}$ and $T^{(2)}$, with $T^{(1)} = L^{LS}(S^{(1)H})$ and $T^{(2)} = L^{(LS)I}$, we can rewrite Eq. (B.2) and (B.5) in block matrix form, as

\[
\begin{bmatrix}
\varepsilon \\
-E \\
-H
\end{bmatrix} =
\begin{bmatrix}
S^{(1)}_{11} & S^{(1)}_{12} & S^{(1)}_{13} \\
S^{(1)}_{21} & S^{(1)}_{22} & S^{(1)}_{23} \\
S^{(1)}_{31} & S^{(1)}_{32} & S^{(1)}_{33}
\end{bmatrix}
\begin{bmatrix}
\varepsilon \\
-E \\
-H
\end{bmatrix},
\]

\[\begin{bmatrix}
\sigma \\
D \\
B
\end{bmatrix} =
\begin{bmatrix}
S^{(1)}_{11} & S^{(1)}_{12} & S^{(1)}_{13} \\
S^{(1)}_{21} & S^{(1)}_{22} & S^{(1)}_{23} \\
S^{(1)}_{31} & S^{(1)}_{32} & S^{(1)}_{33}
\end{bmatrix}
\begin{bmatrix}
\sigma \\
D \\
B
\end{bmatrix},
\]

\[\begin{bmatrix}
\varepsilon \\
-E \\
-H
\end{bmatrix} =
\begin{bmatrix}
T^{(1)}_{11} & T^{(1)}_{12} & T^{(1)}_{13} \\
T^{(1)}_{21} & T^{(1)}_{22} & T^{(1)}_{23} \\
T^{(1)}_{31} & T^{(1)}_{32} & T^{(1)}_{33}
\end{bmatrix}
\begin{bmatrix}
\varepsilon \\
-E \\
-H
\end{bmatrix}.
\]

The definition of $S^{(2)}$ is given by

\[
\begin{bmatrix}
\sigma \\
E \\
H
\end{bmatrix} =
\begin{bmatrix}
S^{(2)}_{11} & S^{(2)}_{12} & S^{(2)}_{13} \\
S^{(2)}_{21} & S^{(2)}_{22} & S^{(2)}_{23} \\
S^{(2)}_{31} & S^{(2)}_{32} & S^{(2)}_{33}
\end{bmatrix}
\begin{bmatrix}
\sigma \\
E \\
H
\end{bmatrix}.
\]

When only $\sigma^*$ is applied in Eq. (B.6), we have

\[
\sigma = S^{(3)}_{11}\sigma^*, \quad E = -T^{(3)}_{21}\sigma, \quad H = -T^{(3)}_{31}\sigma^*.
\]

Likewise, when only $\sigma^*$ is applied in Eq. (B.7), we have

\[
\sigma = S^{(3)}_{11}\sigma^*, \quad E = S^{(3)}_{22}\sigma, \quad H = S^{(3)}_{33}\sigma^*.
\]

Comparing Eq. (B.8) with (B.9), it is easy to find

\[
S^{(2)}_{11} = S^{(3)}_{11}, \quad S^{(2)}_{22} = -T^{(3)}_{21}, \quad S^{(2)}_{33} = -T^{(3)}_{31}.
\]

In the same way, the expression of $S^{(2)}$ can be determined as

\[
S^{(2)} =
\begin{bmatrix}
S^{(3)}_{11} & -T^{(3)}_{12} & -T^{(3)}_{13} \\
-T^{(3)}_{21} & S^{(3)}_{22} & S^{(3)}_{23} \\
-T^{(3)}_{31} & S^{(3)}_{32} & S^{(3)}_{33}
\end{bmatrix}.
\]

Unlike the symmetric moduli tensor $L$, the $S$-tensor in general is not symmetric, but their product $LS$ and $S^{(L)}L^{-1}$ are always symmetric. This can be used as a guide to check the correctness of $S$-tensor.

REFERENCES