Time-decaying magnetoelectric effects in multiferroic fibrous composites with a viscous interface

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This paper addresses the time-dependent magnetoelectroelastic responses of multiferroic fibrous composites with a viscous interface. First, the problem of an isolated multiferroic fiber embedded in an infinite multiferroic matrix is rigorously solved. It is observed that the internal magnetoelectroelastic field such as stresses, electric displacements, and magnetic inductions inside an isolated multiferroic fiber is uniform but time dependent. The Mori–Tanaka mean-field method is then utilized to derive an extremely concise expression of the time-dependent effective moduli of the multiferroic fibrous composite. The numerical results demonstrate that the viscosity of the interface will cause a time-decaying magnetoelectric effect of the BaTiO3–CoFe2O4 fibrous composite. As the time approaches infinity the magnetoelectric effect will approach zero due to the fact that a viscous interface will finally evolve into a free-sliding one which does not sustain shear stress. This interesting feature should be particularly important to the analysis and design of multiferroic composites where the interface is utilized to enhance the magnetoelectric effect.

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I. INTRODUCTION

Recently magnetoelectric (ME) multiferroic materials, which simultaneously possess both ferroelectric and ferromagnetic order in the same phase, have attracted significant attention from the scientific community due to their ME effect, dielectric polarization of a material under magnetic field, or induced magnetization under an electric field. Particularly multiferroic composites made of ferromagnetic and ferroelectric phases can exhibit a strong extrinsic ME effect through the product property, which is absent in the constituents and which can be a few orders larger than the intrinsic ME effect observed in natural single-phase multiferroic materials. It has been identified that the interface in multiferroic composites is critical in achieving the ME effect, and that any kind of imperfection at the interface would lead to a reduction in the ME effect.

At elevated working temperatures exceeding about one-third of the homologous temperature, mass transport becomes important along high diffusivity path such as an interface or grain boundary. Experimental data of Srinivasan et al. have demonstrated that the interfacial diffusion of metal ions between manganites and PZT will degrade the ME effect in multilayer composites. Raj and Ashby and Ashby suggested that the microscopically mass diffusion-controlled mechanism on a length scale comparable to the size of the asperity of the interface can be macroscopically described by the linear law for a viscous interface: \( \dot{\delta} = \tau / \eta \), where \( \dot{\delta} \) is the sliding velocity (i.e., the differentiatation of the relative sliding with respect to time \( t \)), \( \tau \) is the interfacial shear stress, and \( \eta \) is the interfacial viscosity which can be determined experimentally and theoretically.

Motivated by the importance of the interface in the ME effect of multiferroic composites, we propose, in this paper, a theoretical framework to study how the interfacial diffusion characterized by a viscous interface influences the ME effect in the multiferroic composite. For simplicity, the possible contribution of the current density due to the flow of ions at the interface is not considered, which renders only a scalar magnetic potential into the governing equations. This paper is organized as follows: In Sec. II, the problem of an isolated fiber in an infinite matrix is solved in detail; in Sec. III, we derive the time-dependent effective moduli of the multiferroic composite based on the Mori–Tanaka mean-field method; a numerical example of the ME effect is given in Sec. IV; and conclusions are drawn in Sec. V.

II. ISOLATED FIBER IN AN INFINITE MATRIX

We first consider an isolated multiferroic fiber with a circular cross section (phase 2) of radius \( R \) centered at the origin embedded in an infinite multiferroic matrix (phase 1) (Fig. 1). Both the fiber and matrix are 6 mm material symmetry about the fiber axis. At infinity, the matrix is subjected to the antiplane shear stresses \( \sigma_{xy}^m \) and \( \sigma_{yz}^m \), and the in-plane electric displacements \( D_x^m \) and \( D_y^m \), and magnetic fluxes \( B_y^m \) and \( B_z^m \). Thus the two-phase composite system is in a state of antiplane deformation described by

\[
\begin{align*}
    u_x &= u_y = 0, \\
    u_z &= w(x, y, t),
\end{align*}
\]

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Thus the constitutive equations reduce to

\[
\begin{align*}
\sigma_{xx} &= c_{44} \frac{\partial w}{\partial x} + e_{15} \frac{\partial \phi}{\partial x} + q_{15} \frac{\partial \varphi}{\partial x}, \\
\sigma_{yy} &= c_{44} \frac{\partial w}{\partial y} + e_{15} \frac{\partial \phi}{\partial y} + q_{15} \frac{\partial \varphi}{\partial y}, \\
D_x &= e_{15} \frac{\partial w}{\partial x} - e_{11} \frac{\partial \phi}{\partial x} - \alpha_{11} \frac{\partial \varphi}{\partial x}, \\
D_y &= e_{15} \frac{\partial w}{\partial y} - e_{11} \frac{\partial \phi}{\partial y} - \alpha_{11} \frac{\partial \varphi}{\partial y}, \\
B_x &= q_{15} \frac{\partial w}{\partial x} - \alpha_{11} \frac{\partial \phi}{\partial x} - \mu_{11} \frac{\partial \varphi}{\partial x}, \\
B_y &= q_{15} \frac{\partial w}{\partial y} - \alpha_{11} \frac{\partial \phi}{\partial y} - \mu_{11} \frac{\partial \varphi}{\partial y},
\end{align*}
\]

where \(\sigma_{kk}, D_k, B_k, (k=x,y), c_{44}, e_{15}, q_{15}, \alpha_{11}, \) and \(\mu_{11}\) are the stresses, electric displacements, magnetic fluxes (i.e., magnetic inductions), elastic modulus, piezoelectric coefficient, piezomagnetic coefficient, dielectric permittivity, ME coefficient, and the magnetic permeability, respectively.

Under this antiplane deformation, the governing equations are simplified to

\[
\begin{align*}
c_{44} \nabla^2 w + e_{15} \nabla^2 \phi + q_{15} \nabla^2 \varphi &= 0, \\
e_{15} \nabla^2 w - e_{11} \nabla^2 \phi - \alpha_{11} \nabla^2 \varphi &= 0,
\end{align*}
\]

where \(\nabla^2 = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\) is the two-dimensional Laplace operator. In Eq. (3) we have ignored the inertia effect in the fiber and matrix.

The general solution to Eq. (3) can be given by

\[
\begin{align*}
\mathbf{u} &= [w \quad \phi \quad \varphi]^T = \mathbf{f}(z,t),
\end{align*}
\]

where \(z=x+iy=r \exp(i\theta)\) is the complex variable. The appearance of the real time variable \(t\) in Eq. (4) comes from the influence of the viscous interface.

Furthermore, the strains \(\gamma_{xx}\) and \(\gamma_{yy}\), electric fields \(E_x\) and \(E_y\), and the magnetic fields \(H_x\) and \(H_y\) in Eq. (5) are related to \(w, \phi\) and \(\varphi\) through

\[
\begin{align*}
\gamma_{xx} &= w_{,xx}, & \gamma_{yy} &= w_{,yy}, \\
E_x &= -\phi_{,x}, & E_y &= -\phi_{,y}, \\
H_x &= -\varphi_{,x}, & H_y &= -\varphi_{,y}.
\end{align*}
\]

The boundary conditions on the viscous interface (or time-dependent sliding interface) between the fiber and matrix can be written as

\[
\begin{align*}
\sigma^{(1)}_{xx} &= \sigma^{(2)}_{xx}, & D^{(1)}_x &= D^{(2)}_x, & B^{(1)}_x &= B^{(2)}_x, \\
\phi^{(1)} &= \phi^{(2)}, & \varphi^{(1)} &= \varphi^{(2)}, & r &= R \text{ and } t > 0
\end{align*}
\]

where the overdot denotes differentiation with respect to time \(t\).

At the initial moment the interface is a perfect one on which

\[
\begin{align*}
\sigma^{(1)}_{xx} &= \sigma^{(2)}_{xx}, & D^{(1)}_x &= D^{(2)}_x, & B^{(1)}_x &= B^{(2)}_x, \\
w^{(1)} &= w^{(2)}, & \phi^{(1)} &= \phi^{(2)}, & \varphi^{(1)} &= \varphi^{(2)}, & r &= R \text{ and } t = 0
\end{align*}
\]

In view of the above initial conditions, Eq. (8) can also be expressed more concisely as

\[
\begin{align*}
\sigma^{(1)}_{xx} = \sigma^{(2)}_{xx}, & D^{(1)}_x = D^{(2)}_x, & B^{(1)}_x = B^{(2)}_x, \\
w^{(1)} - w^{(2)} = R\mathbf{A}D^{(2)}_x, & \phi^{(1)} - \phi^{(2)} = R\mathbf{A}D^{(2)}_x, & B^{(1)}_x = B^{(2)}_x.
\end{align*}
\]
where \( \lambda \) is a \( 3 \times 3 \) diagonal matrix defined by

\[
\lambda = \frac{1}{\eta R} \text{diag}[1 \ 0 \ 0].
\]

The interface conditions in Eq. (10) can also be expressed in terms of two analytic function vectors, \( \mathbf{f}_1(z, t) \) defined in the matrix and \( \mathbf{f}_2(z, t) \) defined in the fiber, as follows

\[
L_2 \mathbf{f}_2'(z, t) + L_2 \mathbf{f}_2''(z, t) = L_1 \mathbf{f}_1(z, t) + L_1 \mathbf{f}_1''(z, t), \quad (12a)
\]

\[
\mathbf{f}_1'(z, t) - \mathbf{f}_1''(z, t) = \lambda \mathbf{f}_1(0, t) + \frac{z}{z} \mathbf{f}_2''(z, t),
\]

\[
L_1 \mathbf{f}_2''(z, t) + L_1 \mathbf{f}_2'(z, t) = \lambda L_2 \mathbf{f}_2(z, t) - \mathbf{k} - \mathbf{k} \mathbf{R}^2 / z, \quad (13)
\]

where the vector \( \mathbf{k} \) is related to the remote loading through the following:

\[
\mathbf{k} = L_1^{-1} \begin{bmatrix} \sigma_x^0 + i \sigma_x^0 \\ \sigma_y^0 + i \sigma_y^0 \\ \sigma_z^0 + i \sigma_z^0 \end{bmatrix}. \quad (14)
\]

Substituting Eq. (13) into Eq. (12b) and eliminating \( \mathbf{f}_1'(z) \) and \( \mathbf{f}_1''(z) \), we finally arrive at the following set of first-order partial differential equations:

\[
z \mathbf{A} L_2 \mathbf{f}_2''(z, t) + \mathbf{H} \mathbf{L}_2 \mathbf{f}_2'(z, t) = 0, \quad (|z| < R), \quad (15)
\]

where \( \mathbf{H} \) is a real and symmetric matrix given by

\[
\mathbf{H} = \mathbf{H}^T = \begin{bmatrix} H_{11} & H_{12} & H_{13} \\ H_{12} & H_{22} & H_{23} \\ H_{13} & H_{23} & H_{33} \end{bmatrix} = \frac{1}{\eta R} \text{diag}[1 \ 0 \ 0]. \quad (16)
\]

In order to solve Eq. (15), we first consider the following eigenvalue problem:

\[
(\lambda \mathbf{H} - \mathbf{A}) \mathbf{v} = \mathbf{0}, \quad (17)
\]

The three eigenvalues \( \lambda_i \) \( (i=1-3) \) of this eigenvalue problem can be explicitly determined as

\[
\lambda_1 = \frac{H_{22}H_{33} - H_{23}^2}{\eta R} > 0, \quad \lambda_2 = \lambda_3 = 0. \quad (18)
\]

The eigenvectors associated with these eigenvalues are

\[
\mathbf{v}_1 = \begin{bmatrix} H_{22}H_{33} - H_{23}^2 \\ H_{12}H_{33} - H_{13}H_{23} \\ H_{13}H_{22} - H_{12}H_{23} \end{bmatrix}, \quad \mathbf{v}_2 = \begin{bmatrix} 0 \\ 1 \\ 0 \end{bmatrix}, \quad \mathbf{v}_3 = \begin{bmatrix} 0 \\ 0 \\ -H_{22} \end{bmatrix}. \quad (19)
\]

It can be proved that the following orthogonal relationships with respect to the two real and symmetric matrices \( \mathbf{H} \) and \( \lambda \) hold

\[
\mathbf{P}^T \mathbf{H} \mathbf{P} = \text{diag}[\delta_1 \ \delta_2 \ \delta_3],
\]

\[
\mathbf{P}^T \lambda \mathbf{P} = \lambda_1 \mathbf{v}_1 \mathbf{v}_1^T \quad (20a)
\]

and

\[
\delta_1 = \mathbf{v}_1^T \mathbf{H} \mathbf{v}_1 = \lambda_1^{-1} \mathbf{v}_1^T \lambda \mathbf{v}_1 = |\mathbf{H}|(H_{22}H_{33} - H_{23}^2),
\]

\[
\delta_2 = \mathbf{v}_2^T \mathbf{H} \mathbf{v}_2 = H_{22},
\]

\[
\delta_3 = \mathbf{v}_3^T \mathbf{H} \mathbf{v}_3 = H_{23}(H_{22}H_{33} - H_{23}^2). \quad (21b)
\]

We now introduce a new vector function \( \mathbf{\Omega}(z, t) = [\mathbf{\Omega}_1(z, t) \ \mathbf{\Omega}_2(z, t) \ \mathbf{\Omega}_3(z, t)]^T \) defined by

\[
\mathbf{L}_2 \mathbf{f}_2(z, t) = \mathbf{P} \mathbf{\Omega}(z, t), \quad (22)
\]

In view of Eqs. (20) and (22), the original coupled set of differential Eqs. (15) can be decoupled as follows:

\[
\mathbf{\Omega}_1(z, t) + \lambda_1 \mathbf{\Omega}_1'(z, t) = 0, \quad (|z| < R), \quad (23)
\]

\[
\mathbf{\Omega}_2(z, t) = 0, \quad (|z| < R), \quad (24)
\]

\[
\mathbf{\Omega}_3(z, t) = 0.
\]

It is of interest to observe that the two component functions \( \mathbf{\Omega}_2(z, t) \) and \( \mathbf{\Omega}_3(z, t) \) are in fact time independent. Due to the fact that at the time \( t=0 \) the interface is a perfect one, then we arrive at the following initial state of \( \mathbf{\Omega}(z, t) \):

\[
\mathbf{\Omega}(0) = \mathbf{P}^{-1} \mathbf{L}_2 \mathbf{f}_2'(0) = 2 \text{diag} \begin{bmatrix} 1/\delta_1 & 1/\delta_2 & 1/\delta_3 \end{bmatrix} \mathbf{v}_k z. \quad (25)
\]

During the above derivation we have utilized the first orthogonal relationship in Eq. (20) and the following expression:

\[
\mathbf{f}_2'(z, t) = 2 \mathbf{L}_2^{-1} \mathbf{H}^{-1} \mathbf{v}_k z. \quad (26)
\]

It follows from Eqs. (24) and (25) that the explicit solution of \( \mathbf{\Omega}(z, t) \) is
\[ \Omega(z,t) = 2 \text{diag} \left[ \frac{\exp(-\lambda_1 t)}{\delta_1}, \frac{1}{\delta_2}, \frac{1}{\delta_3} \right] \Phi^T \kappa z. \]  

(27)

Consequently \( f_1(z,t) \) defined in the matrix and \( f_2(z,t) \) defined in the fiber are given by

\[ f_1(z,t) = 2L_1^{-1} \Phi \text{diag} \left[ \frac{\exp(-\lambda_1 t)}{\delta_1}, \frac{1}{\delta_2}, \frac{1}{\delta_3} \right] \Phi^T \kappa z, \]

\[ f_2(z,t) = 2L_2^{-1} \Phi \text{diag} \left[ \frac{\exp(-\lambda_1 t)}{\delta_1}, \frac{1}{\delta_2}, \frac{1}{\delta_3} \right] \Phi^T \kappa z, \]

which indicates that the internal magnetoelastic field such as stresses, electric displacements and magnetic inductions inside the multiferroic fiber is uniform but time dependent.

### III. TIME-DEPENDENT EFFECTIVE MODULI

We assume that the aligned circular multiferroic fibers of same radius are randomly distributed in the \( x-y \) plane; then the fiber-reinforced multiferroic composite is transversely isotropic with the \( x-y \) plane being the isotropic plane. The overall constitutive law for the fibrous multiferroic composite can be represented by

\[ \begin{bmatrix} \langle \sigma_{yz} \rangle \\ \langle D_y \rangle \\ \langle B_y \rangle \end{bmatrix} = L_c \begin{bmatrix} \langle \gamma_{yz} \rangle \\ \langle -E_y \rangle \\ \langle -H_y \rangle \end{bmatrix}, \]

where \( \langle \cdot \rangle \) stands for the average value, and \( L_c \) is the effective moduli of the fibrous composite. In the following we will apply the Mori–Tanaka mean-field method to derive the expression for the effective moduli \( L_c \).

In order to describe the overall behavior of the multiferroic composite, we focus on a representative volume element (RVE). In addition we assume that the RVE is subjected to the loadings \( \sigma_{yz}^w, D_y^w, \) and \( B_y^w \). Then, the volume-averaged physical quantities within the RVE can be proved to be

\[ \begin{bmatrix} \langle \sigma_{yz} \rangle \\ \langle D_y \rangle \\ \langle B_y \rangle \end{bmatrix} = (1 - c_2) \begin{bmatrix} \langle \sigma_{yz} \rangle \langle D_y \rangle \langle B_y \rangle \end{bmatrix} + c_2 \begin{bmatrix} \langle \sigma_{yz} \rangle \langle D_y \rangle \langle B_y \rangle \end{bmatrix}, \]

(30)

\[ \begin{bmatrix} \langle \gamma_{yz} \rangle \\ \langle -E_y \rangle \\ \langle -H_y \rangle \end{bmatrix} = (1 - c_2) \begin{bmatrix} \langle \gamma_{yz} \rangle \langle -E_y \rangle \langle -H_y \rangle \end{bmatrix} + c_2 \begin{bmatrix} \langle \gamma_{yz} \rangle \langle -E_y \rangle \langle -H_y \rangle \end{bmatrix}, \]

and the surface integral in Eq. (31) can be carried out as follows:

\[ \frac{1}{\pi R^2} \begin{bmatrix} \int_l (w^{(1)} - w^{(2)}) \hat{n}_2 dl \\ 0 \\ 0 \end{bmatrix} \]

\[ = \frac{2[1 - \exp(-\lambda_1 t)]}{\lambda_1 \delta_1} \Lambda \Phi \text{diag} \left[ 1 \ 0 \ 0 \right] \]

(35)
In addition, in view of the orthogonal relations in Eq. (20), the following identity can be established:

\[
\begin{align*}
(1 - c_2)L_1 + 2c_2L_1L_2^{-1} & \Phi \left( \frac{\exp(-\lambda_1 t/\delta_1)}{\delta_1} \right. \\
& \left. + \frac{1}{\delta_2} \frac{1}{\delta_3} \right) \Phi^T \\
\end{align*}
\]

Consequently we can simplify Eq. (36) as

\[
\begin{align*}
\left[ \langle \gamma_{xy} \rangle, \langle -E_x \rangle, \langle -H_y \rangle \right] = L_1^{-1} \left( (1 + c_2)L_1 - 2c_2 \Phi \right)
\end{align*}
\]

\[
\times \left( (1 - c_2)L_1 + 2c_2 \Phi \right)
\]

\[
\times \left( \frac{\exp(-\lambda_1 t/\delta_1)}{\delta_1} \frac{1}{\delta_2} \frac{1}{\delta_3} \right) \Phi^T
\]

\[
\times \left[ \sigma_{xy}^\infty \right] D_y^\infty \left[ B_y^\infty \right].
\]

Comparison of Eq. (29) with Eq. (38) immediately leads to the time-dependent effective moduli as

\[
L_t = L_t^T = L_1^{-1} \left( (1 + c_2)L_1 - 2c_2 \Phi \right)
\]

\[
\times \left( (1 - c_2)L_1 + 2c_2 \Phi \right)
\]

\[
\times \left( \frac{\exp(-\lambda_1 t/\delta_1)}{\delta_1} \frac{1}{\delta_2} \frac{1}{\delta_3} \right) \Phi^T
\]

\[
L_t(\tilde{t}) = L_1 \left[ (1 + c_2)L_1 - 2c_2(H^{-1} \exp(-\tilde{t})) + M \left[ 1 - \exp(-\tilde{t}) \right] \right] \times \left[ (1 - c_2)L_1 + 2c_2(H^{-1} \exp(-\tilde{t})) + M \left[ 1 - \exp(-\tilde{t}) \right] \right],
\]

where \( \tilde{t} = \lambda_1 t \) is a dimensionless time and

\[
M = M^T = \frac{1}{H_{22}H_{33} - H_{23}^2}
\]

\[
\begin{bmatrix}
0 & 0 & 0 \\
0 & H_{33} & -H_{23} \\
0 & -H_{23} & H_{22}
\end{bmatrix}.\]

IV. TIME-DECAYING ME EFFECTS OF BATO\(_3\) FIBERS REINFORCED IN COFE\(_2\)O\(_4\) MATRIX

As a numerical example, here we consider a typical multiferroic fibrous composite consisting of the magnetostrictive CoFe\(_2\)O\(_4\) matrix reinforced by the piezoelectric BaTiO\(_3\) fibers. The pertinent material properties of BaTiO\(_3\) are: \( c_{44} = 43 \times 10^9 \) N/m\(^2\), \( e_{15} = 11.6 \) C/m\(^2\), \( e_{11} = 11.2 \times 10^{-9} \) C\(^2\)/Nm\(^2\), \( \mu_{11} = 5 \times 10^{-6} \) Ns\(^2\)/C\(^2\); while those of CoFe\(_2\)O\(_4\) are: \( c_{44} = 45.3 \times 10^9 \) N/m\(^2\), \( q_{15} = 550 \) m/A, \( e_{15} = 0.08 \times 10^{-9} \) C\(^2\)/Nm\(^2\), \( \mu_{11} = 590 \times 10^{-6} \) Ns\(^2\)/C\(^2\). Figure 2 demonstrates the ME coefficient \( \alpha_{11} \) as a function of the BaTiO\(_3\) volume fraction \( c_2 \) at four different normalized time.
In this figure the dashed line indicates the trace of the locations of the transient maximum ME effect at different times. It is observed from Fig. 2 that: (i) the magnitude of $\alpha_{11}$ monotonically decreases as the time increases; (ii) the ME coefficient $\alpha_{11}$ is ignorable when the time $t$ is equal to or greater than three times of the relaxation time $t_0 = 1/\lambda_1$; (iii) the optimal value of the BaTiO$_3$ volume fraction, at which the transient maximum ME effect at a fixed time occurs, decreases as the time evolves.

V. CONCLUSIONS

The Mori–Tanaka mean-field approach is applied to obtain a closed-form expression of the time-dependent effective moduli of multiferroic fibrous composites with a viscous interface. This expression is considerably interesting since the important ME effect in multiferroic composites is based on the composite product property with the interface as a bridge. While at the initial moment the effective moduli are the largest, just as those for a perfect interface, they are reduced (or deteriorated) with time, and eventually decreased to those for a free-sliding interface as time approaches infinity. Our numerical results for the multiferroic composite of BaTiO$_3$ fibers reinforced in CoFe$_2$O$_4$ matrix further demonstrate that the viscosity of the interface will cause a time-decaying ME effect. We finally point out that while the responses of piezoelectric fibrous composites with a viscous interface were considered previously, a concise expression of the effective moduli as given in Eq. (40) has never been reported in the literature, not to mention the complex multiferroic composites presented in this article.

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