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Theoretical and experimental investigation of magnetoelectric effect for bending-tension coupled modes in magnetostrictive-piezoelectric layered composites

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In this paper, we discuss a theoretical model with experimental verification for the resonance enhancement of magnetoelectric (ME) interactions at frequencies corresponding to bending-tension oscillations. A dynamic theory of arbitrary laminated magneto-elasto-electric bars was constructed. The model included bending and longitudinal vibration effects for predicting ME coefficients in laminate bar composite structures consisting of magnetostrictive, piezoelectric, and pure elastic layers. The thickness dependence of stress, strain, and magnetic and electric fields within a sample are taken into account, as such the bending deformations should be considered in an applied magnetic or electric field. The frequency dependence of the ME voltage coefficients has obtained by solving electrostatic, magnetostatic, and elastodynamic equations. We consider boundary conditions corresponding to free vibrations at both ends. As a demonstration, our theory for multilayer ME composites was then applied to ferromagnetic-ferroelectric bilayers, specifically Metglas-PZT ones. A theoretical model is presented for static (low-frequency) ME effects in such bilayers. We also performed experiments for these Metglas-PZT bilayers and analyzed the influence of Metglas geometry (length and thickness) and Metglas/PZT volume fraction on the ME coefficient. The frequency dependence of the ME coefficient is also presented for different geometries (length, thickness) of Metglas. The theory shows good agreement with experimental data, even near the resonance frequency. © 2012 American Institute of Physics. [http://dx.doi.org/10.1063/1.4732130]

I. INTRODUCTION

Active materials with coupled electric, magnetic, and elastic parameters that simultaneously display ferroelectricity, ferromagnetism, and ferroelasticity can be called multiferroics. Multiferroics are a special class of materials that have attracted much attention because of their potential for enhanced functionality in sensors and other devices. 1–3 Magnetoelectric (ME) effects are defined as an induced polarization in an applied magnetic field, or an induced magnetization in an applied electric field. The ME effect is studied by subjecting the sample to a magnetic (electric) bias field and an ac magnetic field. One then measures the resulting ac electric (magnetic) field produced in the material. Over the past fifty years, ME materials have evolved from single phase compounds, to particulate composites, and finally to laminate composites. 1–4 Remarkably higher ME effects observed in laminate composites are produced by mechanically coupling continuous magnetostrictive and piezoelectric layers. While several models for laminate ME composites exist, these significantly over predict the experimental results. 1–4 Here, we provide an explanation for this discrepancy, and a corresponding analytical model that corrects these differences which is validated with experimental results.

Prior modeling investigations of laminate ME composites have sometimes made questionable assumptions: for example, a homogeneous distribution of field functions (stress, strain, and electromagnetic) through the layers. Based on these approaches, many authors 1–4 have provided an analytical foundation for static ME laminate composites. However, these theoretical approaches have yielded a huge disagreement with experiments, because of an assumption that all field functions are homogeneously distributed throughout the composite. To eliminate disagreement between analysis and experiments, various authors 2–5 have proposed a model that was free of the above mentioned assumption, where field functions were allowed to vary along (only) the longitudinal direction of the laminates (i.e., no bending). While this provided an approach to better correlate theoretical analysis with experimental data, the approach was applicable only to symmetric laminated composite structures. However, for nonsymmetrical structures, this model remained questionable. For example, ferromagnetic-ferroelectric bilayers cannot be modeled only with interactions along the longitudinal direction: in this particular case, bending-tension modes always couple together and the field functions vary not only along the longitudinal direction but also through the thickness as well. Please note that the mechanism of ferromagnetic-ferroelectric interactions in this case is quite complicated and that simplified approaches to describe them often do not provide adequate conclusions.

Since the ME coupling in composites is directly produced by the mechanical stress, one would expect orders of magnitude stronger coupling when the frequency of the ac field is tuned to acoustic mode frequencies in the sample, relative to non-resonance ones. Many recent experiments and modeling efforts have dealt with ME interactions at
electromechanical resonances in layered composites. However, a key drawback of these studies of ME resonance effects was that the resonance frequencies were quite high, on the order of hundreds of kilohertz for nominal samples of normal dimensions. In order to reduce the resonance frequency, researchers have increased the laminate size, which is disadvantageous for many applications. An alternative approach to achieving strong ME coupling at lower frequencies is resonance enhancement using bending modes of the composite. The frequency of AC fields that must be applied to the composite for such bending oscillations is much lower than that of the longitudinal acoustic modes. Recent investigations have shown giant ME effects at bending oscillations followed: the thickness dependence of stress, strain, and magnetic layers; in this case, the well-known Kirchhoff’s hypotheses are used for the mechanical quantities. The hypotheses for the electrical quantities are generally taken without considering the electrical boundary conditions of the faces of the piezoelectric-piezomagnetic layers.

As a validation, the theory that we developed for multilayer ME composites was applied to ferromagnetic-ferroelectric bilayers. We considered boundary conditions corresponding to free vibrations at both ends. Frequency dependent longitudinal and transverse ME voltage coefficients were obtained using the simultaneous solutions to the electrostatic, magnetostatic, and elastodynamic equations. The ME voltage coefficients were estimated from known material parameters and were then compared to experimental data for bilayers. The model is applied to the specific case of Metglas-PZT bilayers. A theoretical model is presented for static (low-frequency) ME effects in bilayers as a particular case. We analyzed the influence of Metglas geometry (length and thickness) and Metglas/PZT volume fraction on ME coefficient. We also performed experiments for Metglas-PZT bilayers. The frequency dependence of the ME coefficient is presented for different geometries (length and thickness) of Metglas. The theory shows good agreement with experimental data even around resonance frequency.

II. MODEL AND CONSTITUTIVE EQUATIONS FOR A MULTI-LAYER ME LAMINATE COMPOSITE

Consider an arbitrary laminated structure of length $2L$ and $N$ layers. In this case, there is no middle plane of the bar that can serve as a plane of symmetry. For simplicity, we will assume that the multilayer structure is two dimensional (i.e., a bar structure). In this case, the field functions depend only on the spatial coordinates $x_1$ and $x_3$. The geometry and the magnetic loads are represented schematically in Fig. 1. The $x_1$ axis in Cartesian coordinates is directed along the bar length, the $x_2$ axis is directed across the width, and the $x_3$ axis is orthogonal to them both. It is assumed that the piezoelectric layers are poled in the $x_1$ direction. It should be mentioned that the proposed theory can be successfully applied to multilayer structures when the polarization direction of the piezoelectric layers is along the $x_3$ direction, or when some of them are along $x_1$ and along $x_3$ or $x_2$.

We then assumed that the total thickness of the multilayers can be given by

$$h = \sum_{k=1}^{N} h_k = \sum_{k=1}^{N_p} h_{kp} + \sum_{k=1}^{N_m} h_{km} + \sum_{k=1}^{N_e} h_{ke},$$

(1)

where $N = N_p + N_m + N_e$ is the total number of layers; $N_p$ is the number of ferroelectric layers; $N_m$ is the number of ferromagnetic layers; $N_e$ is the number of pure elastic ones; $h_k (k = 1, N)$ is the thickness of the kth layer; $h_{kp} (k = 1, N_p)$ is the thickness of the ferroelectric kth layer; $h_{km} (k = 1, N_m)$ is the thickness of the ferromagnetic kth layer; and $h_{ke} (k = 1, N_e)$ is the thickness of the elastic kth layer. Later, we will use the following notations as well: $H_p = \sum_{k=1}^{N_p} h_{kp}$ which is the total thickness of the piezoelectric layers, $H_m = \sum_{k=1}^{N_m} h_{km}$ which is the total thickness of the piezomagnetic layers, $H_e = \sum_{k=1}^{N_e} h_{ke}$ which is the total thickness of the elastic layers, and $H = h = H_p + H_m + H_e$ which is the total thickness of the composite (Fig. 2).

A. Basic assumptions and restrictions

We then make the following assumptions and restrictions:

(I) The layers are perfectly bonded together (i.e., no cracks or other type of imperfections).

(II) The material of each layer is linearly elastic.

(III) Each layer is of uniform thickness.

FIG. 1. Magneto electric multilayer laminated composite. The arrows indicate the direction of polarization in piezoelectric layer, applied magnetic field and induced magnetic field in ferromagnetic layer.
(IV) The strains and displacements are small.
(V) The length of the composite is much greater than the total thickness, i.e., \( L \gg h \).
(VI) The transverse shear stresses on the top and bottom surfaces of the laminate are zero.
(VII) Bernoulli’s (Kirchhoff’s) hypothesis are valid for any layer, i.e., the displacements in \( x_1 \) and \( x_3 \) directions can be given as
\[
\begin{align*}
  u_1(x_1, x_3) &= u(x_1) - x_3 \frac{\partial w}{\partial x_1}, \\
  u_3(x_1, x_3) &= w(x_1)
\end{align*}
\]
where \( u(x_1) \) and \( w(x_1) \) are plates midplane displacements in \( x_1 \) and \( x_3 \) directions, respectively.

We also assume that the specimen was poled along the longitudinal (\( L \)) direction (i.e., \( x_1 \)-axis). The dc and ac magnetic fields were then applied along the \( L \) direction and across the planes of the contacts (\( \vec{H}_{\text{appl}} \)). The magnetostrictive and piezoelectric layers were assumed to be bonded together with an elastic bonding material-layer of a finite thickness.

Due to magnetostriction, an alternating magnetic field induces vibrations in the magnetostrictive layers, which propagates both across and along the specimen. Mechanical vibrations of the ferrite medium are transferred to the piezoelectric component, via mechanical bonding, wherein an electric field is induced by the piezoelectric effect. Our further considerations will be a case of coupled bending and longitudinal vibrations of this multilayer structure.

With the above assumptions of (I) to (VII), the equations of motions and Maxwell’s electro-magneto static equations, the elastic, magnetic and electro elastic layers can be written as follows (see Refs. 17–20)

\[
T_{ij}^{(k)} = \rho_k \frac{\partial^2 u_i^{(k)}}{\partial t^2} \quad (k = 1, 2, \ldots, N) \quad (3)
\]
\[
B_{i}^{(k)} = 0, \quad e_{ij}^{(k)}H_{j,m}^{(k)} = 0 \quad (k = 1, 2, \ldots, N) \quad (4)
\]
\[
D_{ij}^{(k)} = 0, \quad e_{ij,m}E_{j,m}^{(k)} = 0, \quad (k = 1, 2, \ldots, N); \quad (5)
\]
where \( F_s = \frac{\partial}{\partial t} \); \( e_{ij,m} \) is the permutation symbol with \( e_{ij,1} = 1 \) or \(-1\) depending on whether the indices are in cyclic or anti-cyclic order, respectively, and \( e_{ij,0} = 0 \); otherwise, the superscript in parentheses denotes the number of the layer. The constitutive equations for the magnetostrictive, piezoelectric and pure elastic layers can then be expressed in the following forms (see Refs. 6, 18–20):

**Piezoelectric media**

\[
\begin{align*}
  S_{ij}^{(k)} &= S_{ij}^{(k)} + D_{ij}^{(k)}E_{n,m}^{(k)} \quad (k = 1, 2, \ldots, N; i, j = 1, \ldots, 6; m, n = 1, 2, 3); \\
  B_{ij}^{(k)} &= B_{ij}^{(k)} + E_{n,m}^{(k)}H_{n,m}^{(k)} \quad (k = 1, 2, \ldots, N; i, j = 1, \ldots, 3; m, n = 1, 2, 3).
\end{align*}
\]

**Magnetoelectric media**

\[
\begin{align*}
  S_{ij}^{(k)} &= S_{ij}^{(k)} + D_{ij}^{(k)}E_{n,m}^{(k)} \quad (k = 1, 2, \ldots, N; i, j = 1, \ldots, 6; m, n = 1, 2, 3); \\
  B_{ij}^{(k)} &= B_{ij}^{(k)} + E_{n,m}^{(k)}H_{n,m}^{(k)} \quad (k = 1, 2, \ldots, N; i, j = 1, \ldots, 3; m, n = 1, 2, 3).
\end{align*}
\]

**Pure elastic media**

\[
E_{ij}^{(k)} = E_{ij}^{(k)} \quad (k = 1, 2, \ldots, N; i, j = 1, \ldots, 6). \quad (8)
\]

In Eqs. (6)–(8), we introduced the following notations: \( S_i \) and \( T_i \) are the strain and stress tensor components; \( E_m \) and \( D_m \) are the vector components of electric field and displacement; \( H_m \) and \( B_m \) are the vector components of magnetic field and induction; \( S_{ij}^{(k)} \), \( S_{ij}^{(k)} \), and \( S_{ij}^{(k)} \) are the compliance matrices of the piezoelectric, piezomagnetic, and pure elastic media, respectively; \( q_{im} \) and \( d_{im} \) are the piezomagnetic and piezoelectric coefficients; \( \varepsilon_{im} \) and \( \mu_{im} \) are the dielectric permittivity and magnetic permeability matrices, respectively.

The sub indexes \( P, M, \) and \( E \) correspond to the piezoelectric and piezomagnetic phases and substrate (i.e., pure elastic media), respectively, where the superscripts in the parentheses denote the number of the layer. If the material of the layer is not specified then for compliances we will use notation \( S_{ij}^{(k)} \). We assume that the symmetry of the piezoelectric phase is \( \infty \) and that of the piezomagnetic one is cubic.

As in elastic bar theory, the stresses \( T_1 \) and \( T_3 \) in the constitutive relations can be neglected compared to the stress \( T_1 \), and it is assumed that the magneto-electro-elastic state does not depend on the \( x_2 \) coordinate. Within the scope of Bernoulli’s (Kirchhoff’s) hypothesis of beam bending theory, only the strain \( S_1 \) is induced in a beam, given as

FIG. 2. Piezomagnetic-piezoelectric bi-layer in an applied magnetic field.
\[ S_i = \frac{\partial n_i(x_1, x_3)}{\partial x_1} = \frac{\partial u(x_1)}{\partial x_1} - x_3 \frac{\partial^2 w}{\partial x_1^2} = \varepsilon - x_3 \kappa; \quad (9) \]

where \( \varepsilon = \frac{\partial u(x_1)}{\partial x_1} \) is a strain along the neutral axis and \( \kappa = \frac{\partial^2 w}{\partial x_1^2} \) is a bending of the neutral axis. Equation (9) denotes the linear behavior of the strain \( S_i \) over the entire cross section of the bending beam, whereas \( x_3 \) defines the distance from the neutral axis.

**B. Boundary conditions on the interfaces of layers**

We next provide the most frequently used boundary conditions for the electrical quantities. If there are no electrodes on the surfaces of the bar and the layer on their surfaces is in contact with a non-conducting medium (i.e., insulating glue or a vacuum or air), the component of the electric induction vector \( D_3 \) normal to these surfaces equals zero:

\[ D_3 = 0. \quad (10) \]

For the electrical field, magnetic field, and magnetic induction components, the following boundary conditions should be satisfied

\[ E^{(k)}_1 \bigg|_{x_3 = z_k} = E^{(k+1)}_1 \bigg|_{x_3 = z_k}; \quad H^{(k)}_1 \bigg|_{x_3 = z_k} = H^{(k+1)}_1 \bigg|_{x_3 = z_k}; \quad (11) \]

where \( k = 0, 1, \ldots N + 1 \) and the components with “0” or “N + 1” superscript in parentheses indicate electromagnetic components with surrounding area to the laminate. If the electrodes are in open-circuit conditions, then the following integral condition for the current is satisfied

\[ I = \int_S \frac{dD_1}{dt} ds = i \omega \int_S D_1 ds = 0, \quad (12) \]

where the integral is evaluated over the surface \( S \) of electrodes.

The mechanical load on the surface is given that on the bar faces, namely

\[ T_6^{(N)} \bigg|_{x_3 = z_N} = q^+_1; \quad T_6^{(N)} \bigg|_{x_3 = z_0} = q^-_1; \quad (13) \]

\[ T_3^{(N)} \bigg|_{x_3 = z_N} = q^+_3; \quad T_3^{(N)} \bigg|_{x_3 = z_0} = q^-_3; \quad (14) \]

where \( q^\pm \) are the forces applied on the surfaces \( x_3 = z_N \) and \( x_3 = z_0 \). The boundary conditions on the composite edges should also be included as well. We will provide these conditions later in a paragraph related to vibration of multilayer composites.

In order to construct a theory of ME bars, some additional assumptions regarding the electrical and magnetic quantities must be made. As in the theory of piezoelectric shells and plates, the content of the hypotheses assumed depends on the electrical conditions on the surfaces of the ME layers. For the piezoelectric layers, we will make the following assumptions, which were previously justified by an asymptotic method for single-layer electro elastic plates and shells:

- The electric field component \( E^{(k)}_1(x_1, x_3) \) is not a function of the thickness coordinate \( x_3 \), i.e.
  \[ E^{(k)}_1(x_1, x_3) = E^{(k)}_1(x_1) = E_0(x_1). \quad (15) \]
- The magnetic field component \( H^{(k)}_1(x_1, x_3) \) is not a function of the thickness coordinate \( x_3 \), i.e.
  \[ H^{(k)}_1(x_1, x_3) = H^{(k)}_1(x_1) = H_0(x_1). \quad (16) \]
- The third component of magnetic induction is zero, i.e.
  \[ B_3^{(k)}(x_1, x_3) = 0. \quad (17) \]

Note that due to assumptions (15)–(17) that the boundary conditions given in Eq. (11) are automatically fulfilled. These three assumptions can be derived by the asymptotic integration of Maxwell’s equations (4) and (5) in composite layers and surrounding areas. These assumptions are the first terms in an asymptotic representation. Any further foundation of assumptions (15)–(17) are outside of the scope of this paper. Analogous asymptotic integration of Maxwell’s equations for only piezoelectric layers was provided in Refs. 17–21.

### III. Resulting Tangential Force and Bending Moment

Due to representation (9), the induced stresses in the \( k \)th layers of the various phases can be expressed as

**Piezoelectric layer**

\[ T_{1P}^{(k)} = \frac{1}{S_{P11}} \left( \varepsilon - x_3 \kappa - d^{(k)}_{11} E^{(k)}_1 \right) (k = 1, 2 \ldots N_P). \quad (18) \]

**Piezomagnetic layer**

\[ T_{1M}^{(k)} = \frac{1}{S_{M11}} \left( \varepsilon - x_3 \kappa - q^{(k)}_{11} H^{(k)}_1 \right) (k = 1, 2 \ldots N_M). \quad (19) \]

**Elastic layer**

\[ T_{1E}^{(k)} = \frac{1}{S_{E11}} \left( \varepsilon - x_3 \kappa \right) (k = 1, 2 \ldots N_E). \quad (20) \]

By integrating the stresses over the thickness, we can obtain the resultant tangential force \( T_1 \) in the following form

\[ T_1 = \sum_{k=1}^{N} \int_{z_{k-1}}^{z_k} T_{1P}^{(k)}(x_1, x_3) dx_3 \]

\[ = \sum_{k=1}^{N} \int_{z_{k-1}}^{z_k} T_{1M}^{(k)}(x_1, x_3) dx_3 \]

\[ + \sum_{k=1}^{N} \int_{z_{k-1}}^{z_k} T_{1E}^{(k)}(x_1, x_3) dx_3. \]

Using Eqs. (18)–(20), the expression in Eq. (21) for \( T_1 \) results in
\[ T_1 = Ae - BK - A_{01}E_0 - A_{02}H_0, \]  

(22)

where

\[ A = \sum_{k=1}^{N_r} h_{kp} + \sum_{k=1}^{N_t} h_{km} + \sum_{k=1}^{N_e} h_{kp}, \]

\[ B = \sum_{k=1}^{N_r} \frac{1}{2s_{p11}} \left( (z_k^E)^2 - (z_k^M)^2 \right) + \sum_{k=1}^{N_t} \frac{1}{2s_{m11}} \left( (z_k^E)^2 - (z_k^M)^2 \right), \]

\[ D = \sum_{k=1}^{N_t} \frac{1}{2s_{m11}} \left( (z_k^E)^2 - (z_k^M) \right), \]

\[ A_01 = \sum_{k=1}^{N_r} \frac{d_{11}^{(k)} h_{kp}}{s_{p11}}, \]

\[ A_02 = \sum_{k=1}^{N_r} \frac{d_{11}^{(k)} h_{km}}{s_{m11}}. \]

\[ (23) \]

The resulting bending moment \( M_1 \) is calculated according to

\[ M_1 = \sum_{k=1}^{N_t} x_3 T_{1}^{(k)}(x_1,x_3)dx_3, \]

\[ = \sum_{k=1}^{N_r} \frac{1}{2s_{p11}} \left( x_3 T_{1}^{(k)}(x_1,x_3) dx_3 \right) + \sum_{k=1}^{N_t} \frac{1}{2s_{m11}} \left( x_3 T_{1}^{(k)}(x_1,x_3) dx_3 \right) + \sum_{k=1}^{N_e} \frac{1}{2s_{m11}} \left( x_3 T_{1}^{(k)}(x_1,x_3) dx_3 \right), \]

\[ (24) \]

Using Eqs. (18)–(20), the expression in Eq. (24) for \( M_1 \) results in

\[ M_1 = Be - DK - C_1 E_0 - C_2 H_0; \]

\[ (25) \]

where

\[ D = \sum_{k=1}^{N_r} \frac{1}{2s_{p11}} \left( (z_k^E)^2 - (z_k^M)^2 \right) + \sum_{k=1}^{N_t} \frac{1}{2s_{m11}} \left( (z_k^E)^2 - (z_k^M)^2 \right) + \sum_{k=1}^{N_e} \frac{1}{2s_{m11}} \left( (z_k^E)^2 - (z_k^M)^2 \right), \]

\[ (26) \]

\[ C_1 = \sum_{k=1}^{N_r} \frac{d_{11}^{(k)}}{2s_{p11}} \left( (z_k^E)^2 - (z_k^M)^2 \right), \]

\[ C_2 = \sum_{k=1}^{N_t} \frac{d_{11}^{(k)}}{2s_{m11}} \left( (z_k^E)^2 - (z_k^M)^2 \right). \]

\[ (27) \]

In the context of the above simplification, from the second equation in Eq. (6), the component of electric displacement \( D_1 \) can be expressed in a following form

\[ D_1 = \sum_{k=1}^{N_r} \frac{d_{11}^{(k)}}{2s_{p11}} \left( x_3 T_{1}^{(k)}(x_1,x_3) dx_3 \right), \]

\[ (28) \]

Using Eqs. (18)–(20), the expression (28) results in

\[ C_3 E_0 + C_4 e - C_5 k = D_1; \]

\[ (29) \]

\[ C_3 = \sum_{k=1}^{N_r} \frac{d_{11}^{(k)}}{2s_{p11}} \left( 1 - K_1^2 \right) h_{kp}, \]

\[ C_4 = \sum_{k=1}^{N_r} \frac{d_{11}^{(k)}}{2s_{p11}} \left( 1 - K_1^2 \right) h_{kp}, \]

\[ C_5 = \sum_{k=1}^{N_r} \frac{d_{11}^{(k)}}{2s_{p11}} \left( 1 - K_1^2 \right) h_{kp} + \sum_{k=1}^{N_t} \frac{d_{11}^{(k)}}{2s_{m11}} \left( 1 - K_1^2 \right) h_{km}, \]

\[ \rho = \sum_{k=1}^{N_r} \frac{1}{2s_{p11}} \left( z_k^E \right)^3 - \left( z_k^M \right)^3 + \sum_{k=1}^{N_t} \frac{1}{2s_{m11}} \left( z_k^E \right)^3 - \left( z_k^M \right)^3, \]

\[ \rho = \sum_{k=1}^{N_r} \frac{1}{2s_{p11}} \left( z_k^E \right)^3 - \left( z_k^M \right)^3 + \sum_{k=1}^{N_t} \frac{1}{2s_{m11}} \left( z_k^E \right)^3 - \left( z_k^M \right)^3. \]

\[ (32a) \]

\[ (32b) \]

\[ (32c) \]

\[ (33a) \]

\[ (33b) \]

\[ (33c) \]
V. VIBRATIONS OF MULTILAYER ME COMPOSITES

Next, based on Eqs. (34), we will discuss some issues related to the bending-tension vibration of multilayer composites. The dynamic ME coefficient will be derived for free-free boundary condition at the edges of the composite. As a particular case, bi-layer composites made of ferromagnetic-ferroelectric will be considered.

As an example, consider the harmonic vibrations of a multilayer bar. The top and bottom surfaces are free of stresses, i.e., \(q_i = 0\). We will assume that the system of equations (34) can be decomposed in to two systems of equations:

(A) The equations of the plane problem

\[
\begin{align*}
A \frac{\partial^2 u_0}{\partial x^2} - \rho L^2 \frac{\partial^2 u_0}{\partial t^2} &= 0 \\
\frac{A}{L} \frac{\partial u_0(x,t)}{\partial x} - A_01E_0 - A_02H_0,
\end{align*}
\]

and (B) the equations of the bending problem

\[
\begin{align*}
0 = -D \frac{\partial^2 w_0}{\partial x^2} + C_1E_0 - C_2H_0 \\
M_{10} - \frac{D}{L^2} \frac{\partial^2 w_0(x_1,t)}{\partial x^2} + C_1E_0 - C_2H_0
\end{align*}
\]

We next have to examine the following boundary conditions applied to the edges of the laminated composite. Both ends are free of stresses, i.e., at the edges, \(x = \pm 1\) should satisfy

\[
T_{10} = 0, \quad Q_0 = 0 \quad \text{and} \quad M_{10} = 0.
\]

Using boundary conditions (37), the general solutions of Eqs. (35) and (36) can be obtained in the following form:

\[
u_0(x,t) = e^{i\omega} \frac{A_{01}E_0 + A_{02}H_0}{A} \sin(\beta x) \cos(\gamma x),
\]

\[
w_0(x,t) = e^{i\omega} \frac{C_1E_0 + C_2H_0}{D} \frac{1}{\cos(\beta x) + \sin(\beta x) \cosh(\beta x)} \sinh(\gamma x) \\
\times \left[ \cos(\beta x) \sinh(\beta x) - \sin(\beta x) \cosh(\beta x) \right].
\]

For strain \(\varepsilon = \frac{\partial u_0(x,t)}{\partial x}\) and bending \(\kappa = \frac{\partial w_0(x_1,t)}{\partial x}\), we can obtain the following representations

\[
\varepsilon = \tilde{\varepsilon}(x) e^{i\omega} \frac{A_{01}E_0 + A_{02}H_0}{A}, \quad \kappa = \tilde{\kappa}(x) e^{i\omega} \frac{C_1E_0 + C_2H_0}{D},
\]

where

\[
\tilde{\varepsilon}(x) = \frac{\cos(\beta x)}{\cos(\gamma x)},
\]

\[
\tilde{\kappa}(x) = \frac{1}{\cos(\beta x) + \sin(\beta x) \cosh(\beta x)} \times \left[ -\cos(\beta x) \sinh(\beta x) + \sin(\beta x) \cosh(\beta x) \right].
\]

A. ME COEFFICIENT

Using open circuit conditions (12), along with solutions (38) and (39), we can derive the following compact expression for the ME coefficient \(x_{ME}\)

\[
x_{ME} \equiv x_{ME}(\omega) = \frac{-H}{H} \frac{\Delta_{01}}{\Delta_{02}}.
\]

where

\[
\Delta_{01} = \frac{A_{02}C_4 \tan(\beta L)}{D} + \frac{2C_2C_5}{D} \Delta_0(\beta L),
\]

\[
\Delta_{02} = C_3 + \frac{A_{01}C_4 \tan(\beta L)}{D} + \frac{2C_2C_5}{D} \Delta_0(\beta L),
\]

\[
\Delta_0(\beta L) = \frac{\sinh(\beta L)}{\sin(\beta L) \cosh(\beta L) + \sin(\beta L) \cosh(\beta L) \beta L}.
\]

For the case of ferromagnetic-ferroelectric bi-layer composites, the ME coefficient \(x_{ME}\) can be simplified as

\[
x_{ME} \equiv x_{ME}(\omega) = q_1 d_{11} \frac{\gamma}{\gamma_0} \frac{1}{\gamma + \frac{\gamma}{2}} \frac{\Delta_{01}}{\Delta_{02}}.
\]

where

\[
\tilde{\Delta}_{01} = \frac{\gamma_0}{\gamma_0 + 1} \tan(\beta L) - \frac{3}{2} \frac{\gamma_1}{\gamma_2 + 1} \Delta_0(\beta L),
\]

\[
\tilde{\Delta}_{02} = 1 - K_1^2 + K_2^2 \frac{1}{\gamma_0 + 1} \tan(\beta L) + \frac{3}{2} K_1^2 \frac{1}{\gamma_2 + 1} \Delta_0(\beta L).
\]
\[ \dot{\omega}_T = \sqrt{\frac{D}{A}}, \quad \dot{\omega}_B = \sqrt{\frac{D}{A}}, \quad \omega_0 = \frac{s_{p11}h_m}{s_{m11}h_p}, \]
\[ \gamma_1 = \frac{s_{p11}}{s_{m11}} \left( \frac{h_m}{h_p} \right)^2, \quad \gamma_2 = \frac{s_{p11}}{s_{m11}} \left( \frac{h_m}{h_p} \right)^3, \]
\[ K_1 = \frac{d_{11}^2}{s_{p11}h_p}. \]

1. Some notes and definitions

The frequency at which the ME coefficient \( \gamma_M \rightarrow \pm \infty \) is called the resonance frequency; whereas, the frequency at which \( \gamma_M \rightarrow 0 \) is called the anti-resonance one. The resonance frequency can be determined from the equation \( \Delta_{01} = 0 \). The anti-resonance frequency can be determined from the equation \( \Delta_{0L} = 0 \). The number of resonance and anti-resonant frequencies are discrete and infinite: i.e., the transcendental equations (42) and (43) (or (45) and (46) for bi-layer) have an infinite and discrete number of zeros.

2. Special cases

(A) Static ME coefficient. If we assume \( \omega \rightarrow 0 \) in Eqs. (41)–(43) or (44)–(46) for bi-layers, we can arrive to
\[ \gamma_M = \gamma_M^S = - \frac{C_Aa_02}{A} + \frac{C_2C_5}{D} \frac{h_p}{C_3 + \frac{A_1C_4}{A} + \frac{C_1C_5}{D}} \frac{H_p}{H}; \] (47)
or to
\[ \gamma_M = - \frac{q_{11}d_{11}}{s_{p11}e h_p + h_m} \left\{ \begin{array}{l} \gamma_0 \left( \frac{\gamma_0}{\gamma_0 + 1} + 1 \right) + 1 \left( -K_1^2 - K_1^2 \frac{1}{\Delta_{01}} \left( \frac{\gamma_0}{\gamma_0 + 1} \right) \right) \\
+ \left( -K_1^2 - K_1^2 \frac{1}{\gamma_0 + 1} \right) \end{array} \right\} \] for bi-layers \( \Delta_{01} = \left( \frac{\gamma_0}{\gamma_0 + 1} \right)^2 - \frac{4}{3} \left( \frac{\gamma_0}{\gamma_0 + 1} \right) \). (48)

Formulae (47) is consistent to the model developed by Refs. 3–5 for static ME coefficient. In other words, assuming the vibration frequency approaches zero, from the formulæ for dynamic ME coefficients, we can derive the static ME coefficient as a particular case.

(B) Pure longitudinal mode: If \( D \rightarrow \infty \) in Eq. (41), we can obtain the dynamic ME coefficient for the pure longitudinal mode
\[ \gamma_M \equiv \gamma_M(\omega) = - \frac{H_p A_0C_4}{H - \frac{A_0C_4 \tan(\omega_L)}{\omega_L}} \left\{ \frac{C_3 + \frac{A_0C_4 \tan(\omega_L)}{\omega_L}}{A - \frac{A_0C_4 \tan(\omega_L)}{\omega_L}} \right\}^{-1}. \] (49)

Furthermore, similar formulae can be derived for bi-layer composites: i.e., if in Eq. (44) \( \gamma_2 + 1 \rightarrow \infty \), then one obtains a pure longitudinal mode
\[ \gamma_M \equiv \gamma_M(\omega) = - \frac{q_{11}d_{11}}{s_{p11}e h_p + h_m} \left( \frac{\gamma_0}{\gamma_0 + 1} + 1 \right) + \frac{1}{\Delta_{01}} \left( \frac{\gamma_0}{\gamma_0 + 1} \right) \]
\[ \times \left\{ 1 - K_1^2 + K_1^2 \frac{1}{\gamma_0 + 1} \frac{\tan(\omega_L)}{\omega_L} \right\}^{-1}. \] (50)

This case is consistent to the model developed in Refs. 22 and 23.

(C) Pure bending mode: If \( A \rightarrow \infty \) in Eq. (41), we arrive at the dynamic ME coefficient for the pure bending mode, i.e.,
\[ \gamma_M \equiv \gamma_M(\omega) = - \frac{H_p 2C_5}{H \frac{A}{D} - \frac{A_0C_4 \tan(\omega_L)}{\omega_L}} \left\{ \frac{2C_1C_5}{\omega_L} \frac{\sin(\omega_L)}{\sin(\omega_L) \frac{\tan(\omega_L)}{\omega_L}} \frac{\cos(\omega_L)}{\omega_L} \right\}^{-1} \]
\[ \times \left\{ 1 - K_1^2 + K_1^2 \frac{1}{\gamma_0 + 1} \frac{\tan(\omega_L)}{\omega_L} \right\}^{-1}. \] (51)

and if \( \gamma_0 + 1 \rightarrow \infty \) in Eq. (44), one has a pure bending mode for bi-layer structure, i.e.,
\[ \gamma_M \equiv \gamma_M(\omega) = - \frac{q_{11}d_{11}}{s_{p11}e h_p + h_m} \left( \frac{\gamma_0}{\gamma_0 + 1} + 1 \right) \]
\[ \times \left\{ 1 - K_1^2 + K_1^2 \frac{1}{\gamma_0 + 1} \frac{\tan(\omega_L)}{\omega_L} \right\}^{-1}. \] (52)

VI. NUMERICAL RESULTS

To demonstrate the validity of the theory, we will consider bi-layer laminate composites. In the calculations, we will use the following material parameters for Metglas-PZT composite bilayers given in Table I. For numerical illustrations, the Metglas-PZT bilayer composite was chosen, as a model system, because it has higher piezoelectric and piezomagnetic coefficients.

A. Theoretical validations and experimental verifications

1. Sensor fabrication

To check the theoretical developments, we performed two experiments using Metglas-PZT bi-layer composites. In the first experiment, we assumed a laminate length of \( 2L = 8 \times 10^{-2} \text{ m} \) and in the second \( 2L = 4 \times 10^{-2} \text{ m} \). The maximum number of Metglas layer was chosen as 21. To fabricate bi-layer Metglas-PZT laminates, we obtained commercially available PZT fibers from Smart Materials (Florida, USA) and Metglas foils from Vitrovac Company (Germany). First of all, 5 pieces of 180 \( \mu \text{m} \) thick PZT fibers were oriented

| TABLE I. Material parameters (compliance coefficient’s (10^{-12} m^2/N), piezomagnetic coupling q (10^{-12} m^3/A), piezoelectric coefficient d (10^{-12} m/V), density \( \rho \) (kg/m^3), permeability \( \mu \), and permittivity e for lead zirconate titanate (PZT) and Metglass. |
| --- | --- | --- | --- | --- | --- |
| Material | \( s_{11} \) | \( s_{12} \) | \( s_{13} \) | \( q_{11} \) | \( q_{13} \) | \( d_{11} \) | \( d_{13} \) | \( \mu \rho \) | \( \varepsilon \) |
| PZT | 15.3 | -5 | -7.22 | 17.3 | -175 | 400 | 1 | 1750 | 7600 |
| Metglass | 10 | 50000 | 45000 | 7180 |
FIG. 3. ME voltage coefficient as a function of $f = \omega/2\pi$. Results are for a PZT-Metglas bilayer. Length of the composite $2L = 4 \cdot 10^{-2}$ m; Thickness of PZT is $h_P = 1.8 \cdot 10^{-5}$ m; thickness of one layer of Metglas is $h_M = 2.5 \cdot 10^{-5}$ m. (b) ME voltage coefficient as a function of $f = \omega/2\pi$. Results are for a PZT-Metglas bilayer. Length of the composite $2L = 4 \cdot 10^{-2}$ m; Thickness of PZT is $h_P = 1.8 \cdot 10^{-4}$ m; thickness of one layer of Metglas is $h_M = 2.5 \cdot 10^{-3}$ m. (c) ME voltage coefficient as a function of $f = \omega/2\pi$. Comparison of ME voltage coefficient based on the Eq. (44) for bending-tension coupled model (blue line) and on the Eq. (52) for pure longitudinal mode (purple line). Results are for a PZT-Metglas bilayer. Number of Metglas layers $N = 4$. Length of the composite $2L = 4 \cdot 10^{-2}$ m; Thickness of PZT is $h_P = 1.8 \cdot 10^{-4}$ m; (d) ME voltage coefficient as a function of $f = \omega/2\pi$. Comparison of ME voltage coefficient based on the Eq. (44) for bending-tension coupled model (blue line) and on the Eq. (52) for pure longitudinal mode (purple line). Results are for a PZT-Metglas bilayer. Number of Metglas layers $N = 12$. Length of the composite $2L = 8 \cdot 10^{-2}$ m; Thickness of PZT is $h_P = 1.8 \cdot 10^{-4}$ m; thickness of one layer of Metglas is $h_M = 2.5 \cdot 10^{-5}$ m. Dots are experimental results.

The ME voltage coefficients $\alpha_{ME}$ for bending mode laminates with different layers of Metglas were measured as a function of frequency of ac magnetic driving field. A commercial lock-in amplifier (SR-850) was used to drive a pair of Helmholtz coils, which can generate an ac magnetic field of $H_{ac} = 0.1$ Oe over a frequency range of $10$ Hz $< f = \omega/2\pi < 2$ kHz. The induced voltage from the ME laminates was measured by this amplifier as well. Figure 3(a) shows how $\alpha_{ME}$ was affected by the number of Metglas layers. The peak positions exhibited significant tenability on changing the number (N) of Metglas layers. For 4 cm (Metglas length), the resonant frequency shifted from 325 Hz to 790 Hz with N = 1 to N = 8. Bending mode laminates with N = 4 showed a maximum value of $\alpha_{ME} > 100$ V/cm-Oe. In Figures 3(a) (experiment) and 3(b) (predicted, based on formulae (44)), the ME voltage coefficient $\alpha_{ME}$ is shown as a function of ac magnetic field frequency $f = \omega/2\pi$. The length of the composite was $2L = 4 \cdot 10^{-2}$ m; the thickness of PZT was $h_P = 1.8 \cdot 10^{-4}$ m; and the thickness of each Metglas layer was $h_M = 2.5 \cdot 10^{-5}$ m.

Figure 3(a) shows the experimental results for different layers (number of layers = 5;7;8), and Figure 3(b) shows the theoretical validations using the same parameter choice. As can be clearly seen from these two figures, the experimental data and theoretical predictions are consistent not only qualitatively but also quantitatively. For example, the values of the resonant frequency from experiment differed only...
slightly with the theoretical predictions. In addition, the spectra of $x_{ME}$ computed using our model showed only discrete resonances (see Fig. 3(b)). Please bear in mind that real structures always exhibit losses, which are related primarily to nonideal contacts or introduction of dielectric and magnetic losses in the permittivity and permeability tensors (see Fig. 3(a)). These losses determine the resonance line width and limit the peak value of $x_{ME}$. In reality, such dissipation effects should be included in the modeling to quantitatively compare with experimental spectra. In this paper, we outlined qualitative effects in ME composites, and losses (damping effects) were not taken into consideration. However, losses could be taken into account in the modeling: for example, in formulas (51)–(52), the frequency $\omega$ could be represented as a complex quantity, then the imaginary part of $\omega$ would introduce the damping coefficient.

In Figures 3(c) and 3(d), we represent experimental and theoretical results in the same graph for $x_{ME}$ as a function of $f = \omega/2\pi$. Comparisons of $x_{ME}$ based on Eq. (44) for the bending-tension coupled model (blue line) and on Eq. (52) for the pure longitudinal mode (purple line) are shown, where dots are experimental results. The length of the composite $2L = 8 \cdot 10^{-2} \text{ m}$; thickness of PZT is $h_P = 1.8 \cdot 10^{-4} \text{ m}$; thickness of one layer of Metglas is $h_M = 2.5 \cdot 10^{-3} \text{ m}$. Dots are experimental results. (c) ME voltage coefficient as a function of number of layers of Metglas. Frequency value $f = 200 \text{ Hz}$. Comparison of ME voltage coefficient based on the Eq. (44) for bending-tension coupled model (blue line) and on the Eq. (52) for pure longitudinal mode (purple line). Results are for a PZT-Metglass bilayer. Length of the composite $2L = 8 \cdot 10^{-2} \text{ m}$; thickness of PZT is $h_P = 1.8 \cdot 10^{-4} \text{ m}$; thickness of one layer of Metglas is $h_M = 2.5 \cdot 10^{-3} \text{ m}$. Dots are experimental results. (d) ME voltage coefficient as a function of number of layers of Metglas. Frequency value $f = 280 \text{ Hz}$. Comparison of ME voltage coefficient based on the Eq. (44) for bending-tension coupled model (blue line) and on the Eq. (52) for pure longitudinal mode (purple line). Results are for a PZT-Metglass bilayer. Length of the composite $2L = 8 \cdot 10^{-2} \text{ m}$; thickness of PZT is $h_P = 1.8 \cdot 10^{-4} \text{ m}$; thickness of one layer of Metglas is $h_M = 2.5 \cdot 10^{-3} \text{ m}$. Dots are experimental results.
The data presented were for Metglas-PZT bilayers of length $L = 8$ cm for following frequency values: $f = 120$ Hz (Fig. 5(a)); $f = 150$ Hz (Fig. 5(b)); $f = 280$ Hz (Fig. 5(c)). It can be seen that $z_{ME}$ did not show a similar volume ratio effect as predicted by the theoretical model that only considered longitudinal deformations.\(^{4,14,23}\) Clearly from this figure, we can also see that the bending-tension coupled mode is in much better agreement with experimental data than longitudinal mode. In Fig. 5(d), we can see that overall the value of the ME coefficient increased upon approaching the first resonance frequency of the pure longitudinal mode $f_{1L}$: above which frequency, the overall behavior resembled that of the longitudinal mode (pure longitudinal mode is shown as a red dotted line). However, before approaching the first resonance frequency of the pure longitudinal mode $f_{1L}$, the ME coefficient exhibited resonances at the bending mode frequencies.

**VII. DISCUSSIONS**

A dynamic theory for arbitrary laminated magneto-electro-elastic bars was constructed. A theoretical model including both bending and longitudinal vibration effects was developed for predicting the magneto electric (ME) effects in laminated bar composite structures consisting of magnetostrictive, piezoelectric, and pure elastic layers. Analytical expressions indicate that the vibration frequency strongly influenced the strain distribution in the laminates and that these effects in turn strongly influenced the ME coefficients. As particular cases, the low frequency ME and dynamic coefficients were derived. Dynamic ME coefficients were also derived. As a demonstration, the developed theory for multilayer ME composites was applied to ferromagnetic-ferroelectric bilayers. The vibration of a two-layer magneto-electro-elastic bar was considered. The displacements, stresses, and magneto-electrical quantities were calculated, and the dependence of the coupling coefficient on the
vibration frequencies and the ferromagnetic/piezoelectric layer thickness studied. As a particular case, a theoretical model was presented for low-frequency ME effects in bilayers. We considered boundary condition corresponding to that which is free to vibrate at both ends.

As a demonstration, our theory for multilayer ME composites was then applied to ferromagnetic-ferroelectric bilayers. The model was applied to a specific case of Metglas-PZT bilayers. A theoretical model was presented for static (low-frequency) ME effects in bilayers as a particular case. We analyzed the influence of Metglas geometry (length, thickness) and Metglas/PZT volume fraction on the ME coefficient. We performed experiments for Metglas-PZT bilayers. The frequency dependence of the ME coefficient was also presented for different geometries (length, thickness) of Metglas. The theory shows good agreement with experimental data, even around the resonance frequency.

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