Magnetoelectric effects at electromechanical resonance in laminates of lead-free piezoelectric bimorph and magnetostrictive component

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Abstract. The paper presents a theoretical model for magnetoelectric (ME) effect in multilayers of piezoelectric and magnetostrictive components. We assume the piezoelectric layer to be compositionally-stepped. Taking into account the toxicity of lead-based ceramic, piezoelectric component is made of a lead-free piezoelectric. Analytical expressions for ME voltage coefficients is obtained by substituting the expression for stress at bending mode into open electric circuit condition. Using the layered structure based on the compositionally-stepped piezoelectric components leads to an enhanced magnetoelectric coupling for specific component thickness ratio due to an increase in flexural deformation as compared with the bilayer based on homogeneous piezoelectric layer. For the laminate of lead-free piezoelectric langatate bimorph and magnetostrictive Permendur, ME coupling is stronger due to higher ratio of the piezoelectric coupling coefficient to the dielectric constant. In addition, an 80 % increase in magnetoelectric coupling strength due using the compositionally-stepped piezoelectric is obtained.

1. Introduction
The coupling between the magnetic and ferroelectric ordering in a multiferroic gives rise to magnetoelectric (ME) effect [1-4] due to mechanical deformations. Applied magnetic field $H$ produces the strains of magnetostrictive phase the strains are transferred to the piezoelectric layers. Electric polarization $P$ results from the piezoelectric coupling. ME coupling is described by expression: $P = \alpha H$, with $\alpha$ being the ME susceptibility. In contrast to single-phase materials, the interaction between piezoelectric and magnetic phases in ME composites leads to large values of ME coefficients. In particular, the obtained values of ME susceptibility are several orders of magnitude higher than in the known single-phase materials at room temperature. Thus ME composites are attractive for use in multifunctional devices, such as magnetoelectric converters, attenuators and sensors. Since the ME effect in composite materials is due to the mechanical coupling of the components, a significant enhancement in the ME coupling strength is observed in the electromechanical resonance (EMR) region. [1, 2]. For nominal dimensions of the sample, flexural vibrations occur at significantly lower frequencies than radial and thick oscillations, which makes bending modes preferable for practical applications. A bilayer of functionally graded piezoelectric and magnetostrictive components shows an increased ME coupling at bending mode [5].
The model of low-frequency ME coupling in laminates of piezoelectric bimorph and magnetostrictive layer was discussed recently [6]. Direct ME effect is characterized by the ME voltage coefficient which is equal to the ratio of the induced electric field to the applied magnetic field. It should be emphasized that the magnitude of the ME voltage is proportional to the ratio $d/e$, with $d$ and $e$, denoting the piezoelectric coefficient and dielectric constant for piezoelectric phase, respectively. As a result, the use of appropriate lead-free single-crystal piezoelectrics makes it possible to obtain a value of ME voltage coefficient which exceeds that for piezoceramics based composites containing lead oxide.

This study presents a theoretical model for direct ME effect in the EMR region in multilayers of lead-free single-crystal piezoelectric bimorph and magnetostrictive components. Lead-free composites and single crystal piezoelectrics are the most appropriate piezoelectric materials for environmental protection [7].

2. Theoretical modeling

In this article, we turn to the case of a layered structure with magnetostrictive layer of Permendur and two lead-free single crystal piezoelectric layers of $x$-cut langate with opposite directions of $x$-axis. To reduce demagnetising effects, the ac magnetic field is applied parallel to the length direction, $y$-axis. In addition, the dc magnetic field is applied to the sample for optimizing the piezomagnetic properties of magnetic layer. The sample is assumed to be a thin plate with thickness $t$ that is small compared to width $b$. In turn, $b$ is assumed to be small compared to length $L$. For this case, only the $y$-components of strain and stress can be taken into consideration.

In our model, the strain $S_2$ and electric displacement $D_1$ for two piezoelectric layers (with superscripts $i=p$ and $i=q$) are expressed in terms of stress components $T_2$ and electric field $E_i$ in two piezoelectric layers:

$$\begin{align*}
\rho^1 S_2 &= \rho s_{11} \ T_2 + \rho d_{11} \ E_1; \\
\rho^1 D_1 &= \rho d_{11} \ T_2 + \rho e_{11} \ E_1; \\
\rho^2 S_2 &= \rho s_{11} \ T_2 - \rho d_{11} \ E_1; \\
\rho^2 D_1 &= -\rho d_{11} \ T_2 + \rho e_{11} \ E_1;
\end{align*}$$

(1)

where $\rho d_{11}$, $\rho s_{11}$, and $\rho e_{11}$ are the piezoelectric coefficient, compliance coefficient at constant electric field, and permittivity of piezoelectric layers, correspondingly.

Similarly, the strain $S_2$ and magnetic field $H_1$ in magnetic layer are expressed in terms of stress components $T_2$ and magnetic induction $B_2$ for magnetic layer:

$$\begin{align*}
^m S_2 &= ^m s_{11} \ T_2 + ^m g_{11} \ B_2; \\
^m H_1 &= -^m g_{11} \ T_2 + \frac{1}{^m \mu_{11}} \ B_2;
\end{align*}$$

(2)

where $^m g_{11}$, $^m s_{11}$, and $^m \mu_{11}$ are the piezomagnetic coefficient, compliance coefficient at constant magnetic induction, and permeability of magnetic layer, respectively.

Bending vibrations of a thin plate are governed by following equation for deflection $w$ [8] (displacement in X-direction):

$$w = \frac{P}{64 \pi}$$
\[ V^2 \nabla^2 w + \frac{\rho t}{D} \frac{\partial^2 w}{\partial t^2} = 0 \]  

(3)

with \( \rho, t, D, \) and \( t \) denoting the average density, sample thickness, cylindrical stiffness of the plate, and time, correspondingly. The sample thickness \( t \) equals \( t_m + t_{p1} + t_{p2} \) with \( t_m, t_{p1}, \) and \( t_{p2}, \) and being thickness of magnetostrictive layer and two piezoelectric layers, correspondingly.

For solving (1), we use the boundary conditions for the sample with one end rigidly clamped (at \( y = 0 \)) and one end free (at \( y = L \)). Thus the deflection and derivative of deflection \( \partial w/\partial x \) equal zero at the rigidly clamped end and the turning moment \( M \) and transverse force \( V \) equal zero at free end. It should be noted that the lowest frequency is obtained for the sample fixed at one end and free at the other end.

The turning moment relative to \( z \)-axis can be expressed in terms of stresses as follows:

\[
M_z = \int_{A_{p1}} \rho^1 T_z dA + \int_{A_{p2}} \rho^2 T_z dA + \int_{A_m} \mu^m T_z dA
\]

(4)

where \( A_{p1}, A_{p2}, \) and \( A_m \) are cross-sections of piezoelectric and magnetostrictive layers.

The transverse force is known to be defined by

\[
V = \frac{\partial M_z}{\partial y}
\]

(5)

The following equations can be used to express the strains of both piezoelectric layers and magnetic layer in terms of deflection:

\[
\rho^2 S_2 = -x \frac{\partial^2 w}{\partial y^2}, \quad x_0 - t_{p1} - t_{p2} < x < x_0 - t_{p1},
\]

\[
\rho^1 S_2 = -x \frac{\partial^2 w}{\partial y^2}, \quad x_0 - t_{p1} < x < x_0,
\]

\[
\mu^m S_2 = -x \frac{\partial^2 w}{\partial y^2}, \quad x_0 < x < x_0 + t_m,
\]

(6)

where \( x_0 \) is the distance of sample middle plane from the interface between piezoelectric and magnetostrictive phases.

We are interested in the ME voltage coefficient \( a_E = E_1/H_2 \). The magnetic field induced voltage \( U \) can be estimated in the following manner:

\[
U = \int_{z_0 - \rho_{1t}}^{z_0} \rho^2 E_1 dx + \int_{z_0 - \rho_{1t} - \rho^2_{1t}}^{z_0} \rho^1 E_1 dx.
\]

(7)

Finding the electric field induced across the two piezoelectric layer is enabled by using the open circuit condition \( \int_{G} \rho^2 D_{s} dG = 0 \) for first piezoelectric layer and \( \int_{G} \rho^1 2D_{s} dG = 0 \) for second piezoelectric layer where \( G \) is the cross-sectional area of piezoelectric layers normal to the \( y \)-axis.
For harmonic vibrations, the deflection as a function of $x$ and $\tau$ is determined by expression

$$w(x, \tau) = w(x) \cdot \cos(\omega \cdot \tau)$$

with $\omega$ being the angular frequency. Assuming the electromechanical and magnetomechanical coupling coefficients to be small compared to unity, we obtain the following expression for ME voltage coefficient:

$$\alpha_{e31} = \frac{d_{31}}{\varepsilon_{11}} \cdot q_{11} \cdot (2y_0 + t_m) \frac{2y_0(t_{p_2} - t_{p_1}) + t_m(t_{p_1} - 2t_{p_2}) - t_{p_2}^2}{4s_11s_11DkL(t_{p_1} + t_{p_2} + t_m)[1 + \cosh(k L)\cos(k L)]}$$

where $k$ is the wave number, $k = \omega^{1/2} \left( \frac{\rho t}{D} \right)^{1/4}$.

3. Results and Discussion

Equation (8) shows that ME voltage coefficient is proportional to piezomagnetic coefficient of magnetostrictive phase and ratio of piezoelectric coefficient to dielectric constant of piezoelectric phase. First we consider the ME coupling in a bilayer of magnetostrictive and lead-free single crystal piezoelectric components assuming the thickness of second piezoelectric layer equal to zero, $t_{p_2}=0$ in equation (8). We apply the model developed in the previous section to the bilayer of piezoelectric layer and Permendur, a ferromagnetic alloy (49% Fe, 49% Co and 2% V) with strong piezomagnetic coupling. Figure 1 shows the dependence of ME voltage coefficient on ratio of the piezoelectric coupling coefficient $d$ to the relative dielectric constant $\varepsilon_r$ for layered structure based on Permendur 0.2 mm thick and piezoelectric layer 0.5 mm thick. The following material parameters are used for theoretical estimates: $m_{s_{11}}=7.8 \cdot 10^{-12}$ m²/N, $s_{s_{11}}=9.8 \cdot 10^{-12}$ m²/N, $m_{q_{11}}=0.8 \cdot 10^{-8}$ m/A. The sample length equals 45 mm.
Note that the losses are taken into account by assuming the angular frequency $\omega$ to be complex:

$$\omega = 2\pi (1 + 0.005i) f$$

where $f$ is the applied ac magnetic field frequency.

Estimates in Figure 1 show that the peak ME voltage coefficient at bending mode increases with increasing ratio of the piezoelectric coupling coefficient $d$ to the relative dielectric constant $\epsilon_r$ for permendur-piezoelectric bilayer. Thus the layered structure based on Permendur and lead-free single crystal langatate (LGT) with $d/\epsilon_r=0.25$ enables one to obtain the approximately 2 times higher ME voltage coefficient compared to PZT based laminate containing lead oxide ($d/\epsilon_r=0.13$).

A further increase in ME coupling strength can be obtained for layered structure based on Permendur and piezoelectric bimorph. Next we assume that $t_{pl}=t_{p2}$ in equation (8). Figure 2 presents the frequency dependence of ME voltage coefficient for layered structure based on Permendur 0.2 mm thick and LGT single crystal piezoelectric bimorph (two layers 0.5 mm thick). We used the following material parameters for LGT: $^p d_{11}=-6.5\times10^{-12}$ m/V, $^p c_{11}/\epsilon_{0}=20$.

**Figure 2.** Frequency dependence of ME voltage coefficient for layered structure based on Permendur and LGT single crystal piezoelectric bimorph (1) and for bilayer of Permendur and LGT single crystal piezoelectric bimorph (2)

Data in figure 2 reveal an increase in ME voltage coefficient by 80 percents for the structure of langatate bimorph and permendur compared to the bilayer of permendur and homogeneous langatate. It is seen that the peak ME voltage coefficient reaches 230 V/(cm.Oe). The obtained enhancement of ME coupling strength arises due to an increase in the rotational moment and transverse force for piezoelectric bimorph based structure according to equations (4) and (5). The increase in ME voltage coefficient makes environmentally friendly samples of ME laminates potentially suitable for use in the design of magnetic field sensors and converters.

4. Conclusion

In summary, the explicit expression was derived for the ME coefficient in laminates of piezoelectric bimorph and magnetostrictive phase in the bending mode region. Direct ME effect for langate-
Permendur laminates is shown to be stronger compared to composites with lead-based ceramic ferroelectrics such as PZT.

Direct ME effect was estimated for lead-free piezoelectric langatate bimorph and magnetostrictive Permendur. ME voltage coefficient is approximately 1.8 times higher for the piezoelectric langatate bimorph based structure compared to the homogeneous langatate/Permendur bilayer. The obtained increase in ME coupling strength is due to redistribution of axial strains for compositionally stepped structure due to variation of flexural deformation of the sample. As a result, one obtains the like-sign voltage induced in both langatate layers with opposite x-axis direction. This investigation is of importance for the sensor and technological applications. Distinctive features of lead-free piezoelectric single crystals are the absence of ferroelectric hysteresis and pyroelectric losses. The lead-free composites and single crystal piezoelectrics are the most appropriate piezoelectric materials for environmental protection. The laminates investigated in the present work could potentially be used in ME effect-based devices such as magnetic sensors, energy harvesters etc.

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References

[1] Bichurin M I and Petrov V M 2014 Modeling of Magnetoelectric Effects in Composites (Dordrecht: Springer)
[2] Nan C W, Bichurin M I, Dong C, Viehland D, and Srinivasan G 2008 J. Appl. Phys. 103 031101
[3] Fiebig M 2005 J. Phys. D 38 R123
[4] Srinivasan G, Priya Sh, and Sun N X 2015 Composite Magnetoelectrics (Cambridge: Woodhead Publishing)
[5] Petrov V and Srinivasan G 2008 Phys. Rev. B 78 184421
[6] Petrov V M, Bichurin M I, Kovalenko D V 2017 Proc. of Progress In Electromagnetics Research Symp.-Spring (Saint Petersburg) (New York: IEEE) pp 39-41.
[7] Bichurin M, Petrov V, Zakharov A, Kovalenko D, Yang S. C., D. Maurya, V. Bedekar, and S. Priya 2011 Materials 4 651
[8] Weaver W, Timoshenko S P, and Young D H 1990 Vibration problems in engineering (New York: John Wiley & Sons)