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Magnetoelectric effects in ferromagnetic films on ferroelectric substrates

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Abstract
Theories for magnetoelectric (ME) effects in a bilayer consisting of magnetostrictive film on piezoelectric substrate are discussed. The ME coupling at low frequencies and at mechanical resonance due to acoustic modes have been estimated and applied to the specific case of a thin film of permendur or nickel ferrite on lead zirconate titanate (PZT). Both ideal and non-ideal interface coupling are considered. The theory predicts strong ME coupling for magnetic films on piezoelectric substrates. At low frequency, the ME coefficient is maximum when PZT is 2–4 times as thick as the magnetic film. The ME coefficient, for ideal coupling, shows resonance enhancement at a single frequency. For non-ideal interface coupling, enhancement is expected at two frequencies corresponding to coupled oscillations in magnetic and piezoelectric layers.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The magnetoelectric (ME) effect is the dielectric polarization of a material in an applied magnetic field or an induced magnetization in an external electric field. The induced polarization **P** is related to the magnetic field **H** by the expression, $\mathbf{P} = \alpha\mathbf{H}$, where α is the second rank ME-susceptibility tensor. The effect was first observed in antiferromagnetic Cr_2O_3 [1]. But most single phase compounds show weak ME interactions [2]. A sample of piezomagnetic–piezoelectric phases is expected to be magnetoelectric since $\alpha = \delta P/\delta H$ is the product of the piezomagnetic deformation $\delta z/\delta H$ and the piezoelectric charge generation $\delta Q/\delta z$ [3]. For an ac magnetic field δH applied to a biased sample, one measures the induced voltage δV. The ME voltage coefficient $\alpha_E = \delta V/t\delta H$ and $\alpha = \varepsilon_o\varepsilon_r\alpha_E$ where t is the composite thickness and ε_r is the relative permittivity.

The strongest ME coupling is expected in a layered structure due to (i) the absence of leakage current and (ii) ease of poling to align the electric dipoles and strengthen the piezoelectric effect. Systems studied so far include ferrite, metals or alloys for the magnetic phase and lead zirconate titanate (PZT) or lead magnesium niobate-lead titanate (PMN-PT) for the piezoelectric phase [3–10]. Several composites are found to show a giant ME coupling at low frequencies and a resonance enhancement in the coupling strength when the electrical sub-system shows a resonant behavior, i.e., electromechanical resonance (EMR) for PZT [3].

The nature of ME interactions in nanocomposites is of interest for an understanding of the phenomenon and for useful applications [3–5]. We recently modeled the low-frequency ME effects in nanobilayers, nanopillars and nanowires of nickel ferrite (NFO) and lead zirconate titanate (PZT) on MgO substrates or templates [11]. The clamping effect of the substrate was considered in determining the ME voltage coefficient. It was shown that the coupling strength decreases with increasing substrate thickness. There have been some recent experiments on ME effects in nanobilayers. Systems studied so far, for examples, nickel ferrite–barium titanate (BTO) heterostructure on strontium titanate, show weak ME coupling due to substrate clamping [4]. A much stronger coupling was reported for a system free of a free of any substrate, i.e., cobalt ferrite on BTO [5].

This work is on modeling of ME interactions at low frequencies and at EMR in a bilayer of nanometer thick ferromagnetic phase deposited directly onto a piezoelectric substrate. The model is applied to two systems: permendur, a highly magnetostrictive alloy of Fe and Co, deposited onto PZT substrates and for nickel ferrite on PZT. The model predicts strong ME interactions. Such structures

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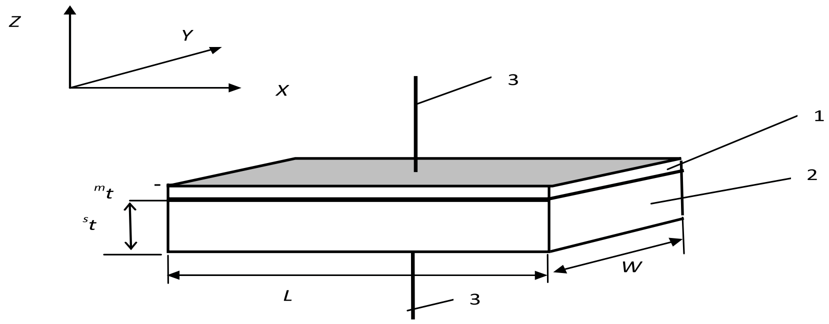

Figure 1. Schematic showing a thin magnetostrictive film (1) on a piezoelectric substrate (2) and electrodes (3). It is assumed that the piezoelectric is poled along z. A bias magnetic field and an ac magnetic field are along x.

offer an alternative to the case of ferromagnetic–ferroelectric nanobilayers on substrates.

2. Theory

A ferromagnetic film with thickness ${}^{\mathrm{m}}t$ on a piezoelectric substrate of width w, length L and thickness ${}^{\mathrm{s}}t$ is considered (figure 1). The sample is polarized along the normal to the contact planes (the Z axis) and subjected to a static magnetic field and an ac field with a frequency of f along the X axis (transverse field orientation). The magnetic field orientation corresponds to minimum demagnetizing field and maximum ME coupling. We assumed that the plate thickness and width are much smaller than its length, i.e., $({}^{\mathrm{m}}t + {}^{\mathrm{s}}t) \ll L, W \ll L$. The magnetic field induces oscillations of the ferromagnetic medium because of magnetostriction. These oscillations in turn induce longitudinal and flexural modes in the substrate and a piezoelectricity induced electrical field. The piezoelectric coefficients associated with the longitudinal modes are ${}^{\mathrm{p}}d_{31}$ and ${}^{\mathrm{p}}d_{32}$, and with the flexural mode is ${}^{\mathrm{p}}d_{35}$. For the PZT substrate the piezoelectric module ${}^{\mathrm{p}}d_{35} = 0$, therefore the flexural mode does not induce an electrical field directly and, therefore, we can neglect the flexural oscillations and take into account only the longitudinal mode.

Besides, the top and bottom surfaces of the plate are assumed to be free; therefore, the surface stresses are equal to zero. Since the plate is thin and narrow, the stress components T_2 and T_3 are assumed to be zero not only on the surfaces but throughout the volume. Thus, the only nonzero component is T_1. In addition, since the top and bottom faces of the plate represent equipotential surfaces, the only nonzero component of the electric field intensity vector is E_3. For the transverse field orientation, equations for the strain tensor ${}^{\mathrm{m}}S_i$ in the magnetic phase has the form

$$ {}^{\mathrm{m}}S_1 = {}^{\mathrm{m}}s_{11}{}^{\mathrm{m}}T_1 + {}^{\mathrm{m}}q_{11}H_1. \quad (1) $$

For the piezoelectric substrate the equation for the strain tensor ${}^{\mathrm{p}}S_i$ and the electric induction ${}^{\mathrm{p}}D_i$ have following form

$$ {}^{\mathrm{p}}S_1 = {}^{\mathrm{p}}s_{11}{}^{\mathrm{p}}T_1 + {}^{\mathrm{p}}d_{31}E_3 \quad (2) $$

$$ {}^{\mathrm{p}}D_3 = {}^{\mathrm{p}}\varepsilon_{33}E_3 + {}^{\mathrm{p}}d_{31}{}^{\mathrm{p}}T_1. \quad (3) $$

Expressing the stress components via the deformation components and substituting these expressions into the equation of the medium motion, we obtain a differential equation for the x projection of the displacement vector of the medium $u(x)$. A solution to this equation for the magnetic layer has the following form:

$$ {}^{\mathrm{m}}u(x) = A_1\cos(kx) + B_1\sin(kx), \quad (4) $$

where ${}^{\mathrm{m}}k = \omega({}^{\mathrm{m}}\rho{}^{\mathrm{m}}s_{11})^{1/2}$, ${}^{\mathrm{m}}\rho$ is the density of the magnetic.

Oscillations of the magnetic displacement ${}^{\mathrm{m}}u(x)$ excited by the magnetic field are transmitted to the substrate via the interface. Since the mechanical contact at the interface in the general case is non-ideal, the x-components of the displacement vector in the ferrite-piezoelectric composite and substrate are related as

$$ {}^{\mathrm{p}}u_x(x) = \beta{}^{\mathrm{m}}u_x(x) + (1-\beta){}^{\mathrm{p}}u_x^{(0)}(x) \quad (5) $$

where β is a parameter describing mechanical coupling between the magnetic layer and the substrate $0 < \beta \leqslant 1$ and ${}^{\mathrm{p}}u_x^{(0)}(x)$ is the displacement of the piezoelectric substrate. From the motion equation of the piezoelectric medium, for the case of no coupling between the two phases, we get the expression for ${}^{\mathrm{p}}u_x^{(0)}(x)$ in following form

$$ {}^{\mathrm{p}}u_x^{(0)}(x) = \frac{{}^{\mathrm{p}}d_{31}E_3}{{}^{\mathrm{p}}k\cos({}^{\mathrm{p}}\kappa)}\sin({}^{\mathrm{p}}kx), \quad (6) $$

where ${}^{\mathrm{p}}k = \omega({}^{\mathrm{p}}\rho{}^{\mathrm{p}}s_{11})^{1/2}$, ${}^{\mathrm{p}}\rho$ is the density of the substrate, and ${}^{\mathrm{p}}\kappa = {}^{\mathrm{p}}k_L/2$ is dimensionless parameter. For the strain-free side surface of the plate at $x = 0$ and $x = L$, the condition of equilibrium for mechanical stresses at the boundary yields

$$ {}^{\mathrm{m}}t{}^{\mathrm{m}}T_1 + {}^{\mathrm{s}}t{}^{\mathrm{p}}T_1 = 0. \quad (7) $$

Using equations (5) and (6) and the boundary condition equation (7), we obtain an expression for magnetic displacement in the form

$$ {}^{\mathrm{m}}u(x) = \frac{({}^{\mathrm{m}}q_{11}H_1 + \beta\gamma\delta^{s}d_{31}E_3)}{{}^{\mathrm{m}}k(1+\beta\gamma\delta)}\frac{\sin({}^{\mathrm{m}}kx)}{\cos({}^{\mathrm{m}}\kappa)}, \quad (8) $$

where $\gamma = \frac{{}^{\mathrm{m}}s_{11}}{{}^{\mathrm{p}}s_{11}}$, $\cdot\delta = \frac{{}^{\mathrm{s}}t}{{}^{\mathrm{m}}t}$ and ${}^{\mathrm{m}}\kappa = {}^{\mathrm{m}}kL/2$ are dimensionless parameters. Substituting equation (8) into equation (5) and take into account equation (6) we get for the substrate displacement the following expression

$$ {}^{\mathrm{p}}u(x) = \beta \frac{({}^{\mathrm{m}}q_{11}H_1 + \beta\gamma\delta\, {}^{\mathrm{p}}d_{31}E_3)}{{}^{\mathrm{m}}k(1+\beta\gamma\delta)} \frac{\sin({}^{\mathrm{m}}kx)}{\cos({}^{\mathrm{m}}\kappa)} + (1-\beta)\frac{{}^{\mathrm{p}}d_{31}E_3}{{}^{\mathrm{p}}k\cos({}^{\mathrm{p}}\kappa)}\sin({}^{\mathrm{p}}kx). \quad (9) $$

From equations (3) and (9), we obtain an equation for the normal component of the electric induction vector

$$ {}^{\mathrm{p}}D_3 = {}^{\mathrm{p}}\varepsilon_{33}E_3 + \frac{{}^{\mathrm{p}}d_{31}}{{}^{\mathrm{p}}s_{11}}\left(\frac{\beta({}^{\mathrm{m}}q_{11}H_1 - \beta\gamma\delta\, {}^{\mathrm{p}}d_{31}E_3)}{(1+\beta\gamma\delta)}\frac{\cos({}^{\mathrm{m}}kx)}{\cos({}^{\mathrm{m}}\kappa)} + \frac{(1-\beta)\, {}^{\mathrm{p}}d_{31}E_3\cos({}^{\mathrm{p}}kx)}{\cos({}^{\mathrm{p}}\kappa)} - d_{31}E_3\right). \quad (10) $$

For the electric current we get

$$ I = \int_0^W \mathrm{d}y \int_{-L/2}^{L/2} \frac{\partial D_3}{\partial t}\,\mathrm{d}x. \quad (11) $$

Substituting equation (10) into equation (11) and integrating we obtain an equation for the electric current in the following form

$$ I = \mathrm{i}\omega W\left({}^{\mathrm{p}}\varepsilon_{33}E_3L + \frac{{}^{\mathrm{p}}d_{31}}{{}^{\mathrm{p}}s_{11}} \times \left(\frac{\beta({}^{\mathrm{m}}q_{11}H_1 - \beta\gamma\delta\, {}^{\mathrm{p}}d_{31}E_3)}{{}^{\mathrm{m}}k(1+\beta\gamma\delta)}2\tan({}^{\mathrm{m}}\kappa) + \frac{(1-\beta)\, {}^{\mathrm{p}}d_{31}E_3}{{}^{\mathrm{p}}k}2\tan({}^{\mathrm{p}}\kappa) - {}^{\mathrm{p}}d_{31}E_3L\right)\right). \quad (12) $$

Using the open-circuit condition $I = 0$ for the strength of the electric field induced in the composite medium we obtain the following equation

$$ E_3 = \beta\frac{{}^{\mathrm{p}}d_{31}\,{}^{\mathrm{m}}q_{11}}{{}^{\mathrm{p}}s_{11}\,{}^{\mathrm{p}}\varepsilon_{33}}\frac{1}{\Delta_a}\left(\frac{1}{(1+\beta\delta\gamma)}\frac{\tan({}^{\mathrm{m}}\kappa)}{{}^{\mathrm{m}}\kappa}\right)H_1, \quad (13) $$

where

$$ \Delta_a = 1 - K_{\mathrm{p}}^2\left(1 - (1-\beta)\frac{\tan({}^{\mathrm{p}}\kappa)}{{}^{\mathrm{p}}\kappa} + \frac{\beta^2\gamma\delta}{(1+\beta\gamma\delta)}\frac{\tan({}^{\mathrm{m}}\kappa)}{{}^{\mathrm{m}}\kappa}\right) $$

and

$$ K_{\mathrm{p}}^2 = {}^{\mathrm{p}}d_{31}^2/({}^{\mathrm{p}}\varepsilon_{33}\,{}^{\mathrm{p}}s_{11}) $$

is the electromechanical coupling coefficient for the planar oscillations. The ME voltage coefficient for the structure is defined as

$$ \alpha_{E,T} = E_{\mathrm{av}}/H_1, \quad (14) $$

where $E_{\mathrm{av}} = U/({}^{\mathrm{m}}t + {}^{\mathrm{s}}t)$ is the average value of the electric field strength in the structure and U is the potential difference between electrodes. Using equation (14) we obtain for the ME voltage coefficient

$$ \alpha_{E,T} = \beta\frac{{}^{\mathrm{p}}d_{31}\,{}^{\mathrm{m}}q_{11}}{{}^{\mathrm{p}}s_{11}\,{}^{\mathrm{p}}\varepsilon_{33}}\frac{1}{\Delta_a}\left(\frac{1}{(1+\beta\delta\gamma)}\frac{\tan({}^{\mathrm{m}}\kappa)}{{}^{\mathrm{m}}\kappa}\right)\frac{\delta}{(1+\delta)}. \quad (15) $$

It is clear from equation (15) that the frequency dependency of $\alpha_{E,T}$ has resonant character. At the so-called antiresonance frequency when $\Delta_a = 0$, the ME coefficient sharply increases and is expected to show a resonance.

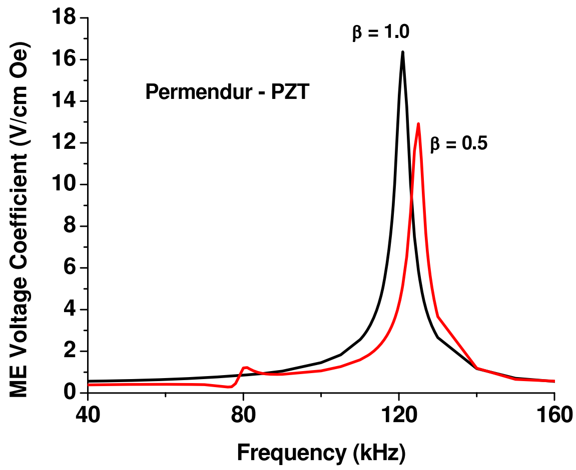

Figure 2. The frequency dependency of magnetoelectric voltage coefficient for a 100 μm thick permendur on 1 mm thick PZT. The estimates are for ideal ($\beta = 1$) and non-ideal ($\beta < 1$) interface coupling. The peak occurs at acoustic resonance modes in the sample.

3. Results and discussion

Next we apply the model to estimate the ME coupling for permendur deposited onto PZT. Permendur is a soft magnetic alloy consisting of 49% Fe, 49% Co and 2% V. It is an ideal material for studies on ME composites due to desirable low resistivity and high Curie temperature (1213 K) and magnetostriction (70 ppm). Lead zirconate titanate was chosen due to high ferroelectric Curie temperature and piezoelectric coupling constant. Figure 2 presents the frequency dependence of the ME voltage coefficient calculated using equation (15) for 100 μm thick permendur on a 1mm thick PZT substrate and the following values for material parameters [12]:

$$ {}^{\mathrm{m}}s_{11} = 5.5\times10^{-12}\ \mathrm{m^2\,N^{-1}}, \qquad q_{11} = 63.75\times10^{-10}\ \mathrm{m\,A^{-1}}, $$

$$ {}^{\mathrm{p}}s_{11} = 15\times10^{-12}\ \mathrm{m^2\,N^{-1}}, $$

$$ d_{31} = -175\times10^{-12}\ \mathrm{m\,V^{-1}}, \qquad {}^{\mathrm{p}}\varepsilon_{33}/\varepsilon_0 = 1750, $$

damping coefficient, $\chi = 20\,000\ \mathrm{s^{-1}}$; length of plate $L = 10$ mm. The estimates are for coupling coefficient β of 1 and 0.5. If the interface coupling is ideal ($\beta = 1$), figure 2 shows a resonant behavior with a single peak in $\alpha_{E,T}$. For non-ideal ($\beta < 1$), two peaks are seen in figure 2 corresponding to interconnected oscillations in the magnetic and piezoelectric layers. The resonance value of ME coefficient is a factor 5–6 smaller than for thick film composites [12].

It is evident from figure 2 that the low-frequency ME voltage coefficient is independent of frequency and, from equation (15), is given by

$$ \alpha_{E,T} = \beta\frac{{}^{\mathrm{p}}d_{31}\,{}^{\mathrm{m}}q_{11}}{{}^{\mathrm{p}}s_{11}\,{}^{\mathrm{p}}\varepsilon_{33}}\frac{1}{(1+\beta\gamma\delta)} \times \frac{1}{(1-K_{\mathrm{p}}^2)\beta(1+\beta\gamma\delta/(1+\beta\gamma\delta))}\frac{\delta}{(1+\delta)}. \quad (16) $$

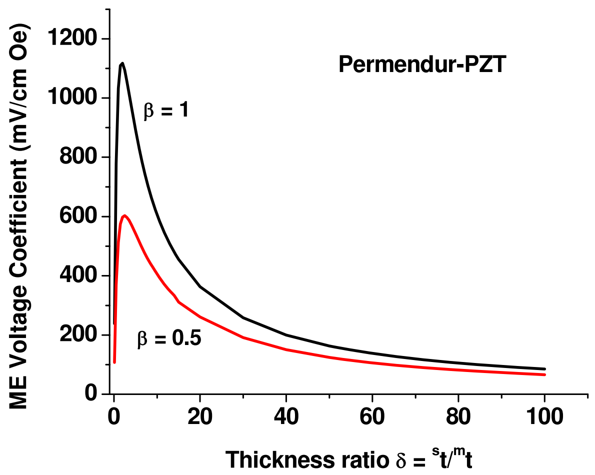

Figure 3. Dependence of the low-frequency magnetoelectric voltage coefficient on substrate-to-film thickness ratio δ for permendur film on PZT.

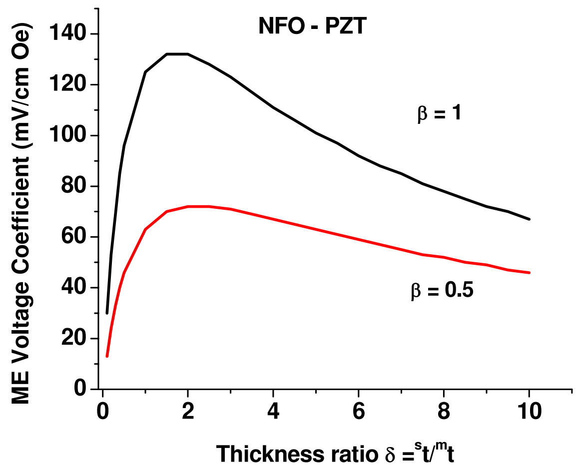

Figure 4. Results as in figure 3 for thin film of nickel ferrite (NFO) on PZT

The ME voltage coefficient depends on the ratio of substrate to film thickness $\delta = {}^{s}t/{}^{m}t$. This dependence has a maximum and can be estimated from equation (16):

$$\left(\frac{{}^{p}t}{{}^{m}t}\right)_{\max} = \sqrt{\frac{{}^{p}s_{11}(1-\beta K_{p}^{2})}{\beta\,{}^{m}s_{11}(1-2\beta^{2}K_{p}^{2})}}. \qquad (17)$$

Figure 3 presents the dependence of ME coefficient on δ for $f = 50$ kHz. For thick substrates, the voltage coefficient is quite small. It then increases with decreasing substrate thickness and reaches a peak value for $\delta \approx 4$. The ME voltage then shows a sharp drop with further decrease in the substrate thickness. The predicted ME coefficients in figure 3 are comparable to measured values in permendur-PZT-permendur trilayers [12].

Finally, a similar estimate of low-frequency ME voltage coefficients is shown in figure 4 for a thin film of nickel ferrite on PZT. The magnetostriction for NFO is smaller than for permendur and, therefore, the ME coefficient is expected to be a factor of 5–10 smaller in NFO-PZT than for permendur-PZT. Figure 4 shows a maximum in the ME voltage when the substrate is twice as thick as NFO and decreases gradually with increase PZT thickness. The ME coefficient is comparable to measured values in multilayers and bilayers of NFO-PZT [7].

4. Conclusions

A model has been developed for low-frequency and resonance ME effects for a structure consisting of a thin magnetostrictive film deposited onto a piezoelectric substrate. The theory is then applied to permendur deposited on PZT and nickel ferrite on PZT. The frequency dependence of α_E predicts resonance enhancement at mechanical resonance for the sample. A single resonance is expected for the case of ideal interface coupling. For non-ideal coupling, two resonances due to acoustic modes in permendur and PZT are predicted. The ME coupling at low frequencies is expected to be maximum when the substrate is four times as thick as the film. Similar ME characteristics are expected for NFO-PZT, but the strength of ME interactions will be smaller because of lower magnetostriction compared to permendur. The results presented here will likely encourage experimental work on the systems modeled here.

Acknowledgment

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