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Enhanced magnetoelectric effects in laminate composites of Terfenol-D/Pb(Zr,Ti)O₃ under resonant drive

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We have found that laminate composites consisting of longitudinally magnetized magnetostrictive Terfenol-D and longitudinally poled piezoelectric Pb(Zr,Ti)O₃ layers have dramatically enhanced magnetoelectric effects when driven near resonance. The maximum induced magnetoelectric voltage at resonance was ~ 10 Vp/Oe, which is $\sim 10^2$ times higher than previous reports at subresonant frequencies. © 2003 American Institute of Physics. [DOI: 10.1063/1.1631756]

The magnetoelectric (ME) effect is a polarization P response to an applied magnetic field H.¹ Recently, a number of studies concerning the ME effect in piezoelectric/magnetostrictive laminate composites have been published.^{2–11} Investigations have focused on subresonant operation conditions, where the ME effect has a flat response over a specified bandwidth to the magnitude of an applied ac magnetic field $H_{\rm ac}$. Terfenol-D/Pb(Mg_{1/3}Nb_{2/3})O₃-PbTiO₃ (PMN-PT) laminates have a large ME voltage coefficient of \sim 0.11 V/Oe, and a limit of magnetic field sensitivity of \sim 10^{–11} T.^{10,11} However, for laminates driven under subresonant conditions, it has proven difficult to further enhance the ME voltage coefficient by an additional, significant margin.

We note that both piezoelectric and magnetostrictive resonators have high coupling (electromechanical and magnetomechanical, respectively) effects. 12,13 Accordingly, it is obvious to suppose that ME resonators driven near their resonance frequency should also have a significantly higher ME coupling, relative to laminates driven subresonantly. Here, we report the development of a resonant type of ME laminate of Terfenol-D and Pb(Zr,Ti)O₃ (PZT) piezoceramic that has a much higher ME voltage coefficient.

laminates consisted of Terfenol-D two $(Tb_{1-x}Dy_xFe_{2-y})$ layers magnetized in their length (or longitudinal) directions, and two hard PZT piezoelectric ceramic layers also poled longitudinally. The piezoelectric elements had a cross-sectional area half that of the Terfenol-D ones. The two PZT elements were then laminated as a single layer between the two Terfenol-D layers, where the polarization direction of the two piezoelements were arranged in reverse orientations within the piezolayer, as shown in Fig. 1(a). We designate this laminate configuration as (L-L). The length of the laminate was 60 mm and the total cross-sectional area A was $10 \times 6 \text{ mm}^2$. This long configuration intensifies the principal vibration along the longitudinal axis. The induced ME voltage was then measured across the longitudinal axis of the PZT layer, where the middle electrode separating the two reversely poled piezoelements of the layer acted as electrical ground. When the laminate was operated in its longitudinal resonance state, maximum ME coupling effects were obtained. This configuration and operational mode are significantly different than previous ones.^{2–11}

First, we designed the laminate using an equation of motion to couple the piezomagnetic and piezoelectric constitutive equations. Because the piezomagnetic and piezoelectric layers are mutually coupled via strain S(z) and stress T(z), application of H along the length direction of the magnetostrictive layer puts the piezoelectric one into forced oscillation in the longitudinal axis, generating a voltage between the end and middle electrodes. At the first longitudinal resonance frequency, this laminate is a half-wavelength ($\lambda/2$) ME resonator. A node line is located at the middle position of the laminate, where the vibration velocity (i.e., mechanical current) is zero. Assuming a symmetric vibration of the laminate, and that the polarization of the piezoelectric layer is symmetric about the node line, the ME voltages ($\alpha_{\rm ME}$) induced across both piezoelements in the layer are equivalent. In addition the magnetoelastoelectric equivalent circuits are

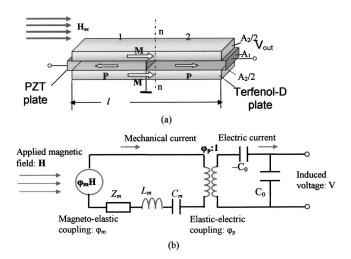


FIG. 1. (a) Configuration of our (*L-L*) mode magnetostrictive/piezoelectric laminate composite. Arrows *M* and *P* designate the magnetization and polarization directions, respectively. (b) Magnetoelastoelectric equivalent circuit of (*L-L*) ME laminate resonator, where $R_m = \pi Z_0/4Q_m$, $L_m = \pi Z_0/4\omega_s$, and $C_m = 1/\omega_s^2 L_m$ are the motional mechanical impedance, inductance, and capacitance, respectively; $\varphi_m = A_1 d_{33,m}/s_{33}^H$ is the magnetoelastic coupling factor; $\varphi_p = 2A_1 g_{33,p}/ls_{33}^D \bar{\beta}_{33}$ is the elastoelectric coupling factor; and $C_0 = 2A_1/l\bar{\beta}_{33}$ is the clamped capacitance of the piezoelectric layer.

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TABLE I. Material parameters for Terfenol-D and PMN-PT crystals.

	$d_{33} \text{ or } d_{33,p}$	d_{31} or $d_{31,p}$	S_{11}^H or	s_{33}^H or s_{33}^E	k ₃₃	k ₃₁	$\varepsilon_{33,T}$
Terfenol-D ^a PZT-8	1.2×10 ⁻⁸ Wb/N 300 pC/N	-5.8×10 ⁻⁹ Wb/N -125 pC/N	$125 \times 10^{-12} \text{ m}^2/\text{N}$ $11.8 \times 10^{-12} \text{ m}^2/\text{N}$	$40 \times 10^{-12} \text{ m}^2/\text{N}$ $17.4 \times 10^{-12} \text{ m}^2/\text{N}$	0.7 0.72	0.58	1250

aCited from Ref. 14.

the same for both parts of the laminate, and can be derived by using the piezoelectric and piezomagnetic constitutive equations, applying Newton's second law of motion to the laminate, and subsequently finding analogous electrical parameters. 12,13

Figure 1(b) shows the magnetoelastoelectric equivalent circuit in the region around the resonance frequency ω_s . This circuit is applicable to either section of the laminate about the nodal line. It is useful for understanding the ME coupling of the laminate under resonance drive conditions. An applied magnetic field H acts as a "mechanical voltage" (i.e., $\varphi_m H$) via a coupling factor φ_m . This excites a "mechanical current" through the magnetoelastic effect. In turn, this results in an electrical voltage V across the two ends of the piezoelectric layer due to electromechanical coupling. A transformer with a turn-ratio of φ_p can be used to represent the electromechanical coupling in the circuit. The ME voltage coefficient $\alpha_{\rm ME}(\omega)$ as a function of the ac magnetic-field frequency for the (L-L) ME laminate resonator can be derived from this equivalent circuit as

$$\alpha_{\rm ME}(\omega) = \frac{\partial V_{\rm ME}}{\partial H_3} = \left| \beta \frac{\varphi_p^2}{j\omega C_0} \frac{\varphi_m}{R_m + j\omega L_m + 1/j\omega C_m} \right|, \quad (1)$$

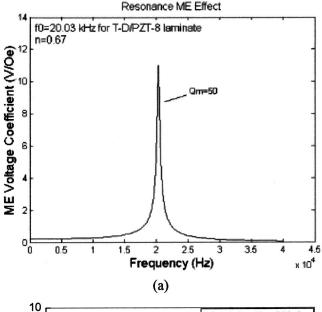
where R_m , L_m , and C_m are motional resistance, inductance, and capacitance of the laminate, respectively, and C_0 is the clamped capacitance. At a resonance frequency of $\omega_s = (1/L_m C_m)^{1/2}$, $\alpha_{\rm ME}(\omega)$ reaches a maximum value of

$$\alpha_{\text{ME},\omega_s} = \beta \frac{4Q_m \varphi_m \varphi_p^2}{n Z_0 \omega_s C_0},\tag{2}$$

where $\beta \leqslant 1$, is a factor related to applied magnetic field bias $H_{\rm dc}$ and at the optimum $H_{\rm dc}$ it equals 1; Q_m is the effective mechanical quality factor of the laminate composite including contributions from the Terfenol-D and piezoelectric layers, and the bonding between the layers; and Z_0 is the characteristic mechanical impedance of the laminate. Equation (2) predicts at resonance that $\alpha_{\rm ME}$ is $8Q_m/\pi^2$ higher than at subresonant frequencies. Using the materials parameters for Terfenol-D and PZT-8 given in Table I, $\alpha_{\rm ME}$ was calculated as a function of the frequency of $H_{\rm ac}$, as shown in Fig. 2(a). Using the experimental thickness ratio for the Terfenol-D layers of n=0.67, and assuming Q_m =50, the maximum value of $\alpha_{\rm ME}$ was predicted to be 10.96 V/Oe at a resonance frequency of f_s =20.03 kHz (ω_s =2 πf_s).

The ME voltage induced across the end and middle (i.e., ground) electrodes of the PZT layer of the laminate was then measured as a function of frequency, using a lock-in amplifier method. Measurements were performed under a dc magnetic bias of $H_{\rm dc} = 200$ Oe, as the inflection point of the quadratic strain-magnetic-field ($\varepsilon - E$) response is known to be maximum near this bias level. A pair of Helmholtz coils was used to generate a small $H_{\rm ac}$, via an input ac current $I_{\rm coil}$. The frequency of $H_{\rm ac}$ was varied over the bandwidth of 1 to

40 kHz. The induced ME voltage of the (L-L) laminate was found to be maximum near a resonance frequency of f_s = 19.96 kHz, as shown in Fig. 2(b). This is consistent with the predictions of Eq. (2). The maximum value of $\alpha_{\rm ME}$ was 8.7 V using a drive field of $H_{\rm ac}$ =1 Oe, a drive frequency of 19.96 kHz, and a magnetic bias of $H_{\rm dc}$ =200 Oe. Correspondingly, the ME voltage coefficient $\alpha_{\rm ME}(\omega_s)$ at resonance was 8.7 V/Oe. From the ME voltage versus frequency data of Fig. 2(b), the 3 dB frequency bandwidth about ω_s was determined as $\Delta f = f_2 - f_1 \sim 0.4$ kHz. Accordingly, the effective mechanical quality factor of the laminate is $Q_m = f_s/\Delta f = 49.9$. Using this value of Q_m , the predicted maximum value of $\alpha_{\rm ME}$ from Eq. (2) agrees well with our mea-



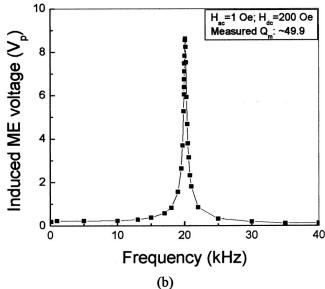


FIG. 2. Induced magnetoelectric voltage α_{ME} as a function of frequency. (a) Calculated using Eq. (2). (b) Measured using a lock-in amplifier method.

sured value. However, a higher Q_m value will clearly result in the laminate having higher values of a ME. For example, assuming that Q_m is equal to that for hard PZT (i.e., $Q_m = 1000$), the value of $\alpha_{\rm ME}$ can be estimated as 200 V/Oe.

It has been found that ME resonators have an extremely high $\alpha_{\rm ME}$. An enhancement in $\alpha_{\rm ME}$ of nearly two orders of magnitude over that of previous ME laminates^{2–11} has been achieved. This achievement opens significant possibilities for applications, due to large energy transduction abilities. In addition, with respect to passive applications as sensors, our ME laminate resonator offers dramatically higher magnetic field sensitivity over narrow bandwidths defined by ω_s . Furthermore, the bandwidth for sensor applications could be notably increased by using an array of resonators with different ω_s .

In summary, a colossal ME voltage coefficient has been found in a long resonance-type composite of PZT laminated between two Terfenol-D layers operated in a (*L-L*) mode. Analysis has shown that $\alpha_{\rm ME}$ at the resonance frequency is $8Qm/\pi^2$ higher than that at subresonant frequencies. Measured results confirm that $\alpha_{\rm ME}$ of our laminate resonator is 10^2 times higher under resonant drive than at subresonant frequencies.

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