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Strong magnetoelectric charge coupling in stress-biased multilayer-piezoelectric/magnetostrictive composites

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A magnetoelectric composite consisting of a multilayer $Pb(Zr,Ti)O_3$ piezoelectric (d_{33}) stack and a magnetostrictive (Galfenol and/or Terfenol-D) rod assembled in a prestressed frame in a longitudinal-longitudinal configuration has been found to have a high magnetoelectric (ME) charge coupling to an applied magnetic field. Values of the ME charge coefficient as high as 16.4 nC/Oe (or 3.6×10^{-6} C/m^2 Oe) were found at quasistatic frequencies and 123 nC/Oe (or 2.8×10^{-5} C/m^2 Oe) under resonance drive, which are 100–1000 times higher than that previously reported for other ME laminates. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2748712]

INTRODUCTION

Giant magnetoelectric (ME) field coefficients of $\alpha_{ME} \sim 2$V/cm Oe have been reported in longitudinally magnetized transversely poled (LT) composites of piezoelectric $Pb(Zr,Ti)O_3$ (PZT) or $Pb(Mg_{1/3}Nb_{2/3})O_3-PbTiO_3$ (PMN-PT) layers laminated together with magnetostrictive $Tb_{1-x}Dy_xFe_{2-y}$,[1] Permendur,[2] Fe–Ga,[3] or $NiFe_2O_4$ (Ref. 4) ones. Such giant values occur when the magnetostrictive layer(s) are in a dc biased piezomagnetic state for $H_{dc}=300-500$ Oe. Various other laminate configurations have also been studied, including among others: (i) a longitudinally poled longitudinally magnetized or LL mode, which should have the highest ME effects;[5] (ii) a LT, consisting of a multilayer-piezoelectric stack,[6] offering enhanced dielectric capacitance due to N dielectric layers; and (iii) push-pull[7] and/or bimorph[8] ones, consisting of symmetrically poled piezoelectric layer(s) offering enhanced pyroelectric noise rejection. In such composites, a ME effect is produced under a dc magnetic field bias without an applied mechanical stress bias: the magnetostrictive phase produces a strain responding to applied magnetic fields (H) including a dc bias field and an ac exciting field via magnetostriction, which is then elastically coupled to the piezoelectric phase inducing an electric field (E) across it via piezoelectricity.

Theoretical investigations[5] have shown that the LL configuration should have by far the highest ME effects—about an order of magnitude. This is because in the LL configuration the piezomagnetic coefficient ($d_{33,m}$) and magnetoelastic coupling coefficient ($k_{33,m}$) of the magnetostrictive layer(s), and the piezoelectric coefficient ($d_{33,p}$) and electromechanical coupling coefficient ($k_{33,p}$) in the piezoelectric layers are maximum. However, experimental investigations[5] of ME effects in LL laminates have shown that α_{ME} for LL laminates are less than that for LT laminates made from the same material couple. Part of this discrepancy lies with the fact that a monolithic piezoelectric layer poled along its length direction has a low capacitance. The LL configuration produces a strong stress coupling between layers; however, the charge induced from the effect is small due to the low dielectric capacitance. Clearly, one has to be concerned with the potential for significantly distorted values of α_{ME}^{LL}, if calculated from the induced charge by $Q_{ME}^{LL}=\alpha_{ME}^{LL}C_0H$, simply due to parasitic capacitances. In addition, studies have shown under a suitable mechanically preload (or stress bias) that the grain-oriented magnetostrictive $Tb_xDy_{1-x}Fe_{2-y}$ exhibits a higher magnetostrictive coefficient due to a "jump effect."[9] This in turn favors a higher ME charge coefficient. When suitably stress biased, the piezoelectric phase in the ME composite may also have an enhanced piezoelectric coefficient[10] ($d_{33,p}$). High magnetoelectric charge couplings are important for developing applications, such as electromagnetic energy harvesting or magnetic-to-electric generators[11] and magnet field sensing based on a charge-detection method at quasistatic frequencies.[8]

Here, we report the finding of a stress-biased LL configuration with high capacitance. We report a dramatically *enhanced ME charge coupling*—a factor of 100–1000 times higher than in any previously reported laminate configuration.

STRESS-BIASED ME CONFIGURATION AND ME EQUIVALENT CIRCUIT

In this study, we bring together the high capacitance of a multilayer piezoelectric that is mechanically in series with a magnetostrictive element under a stress bias. In the said configuration, we can take advantage of the high-stress transfer intrinsically offered by the LL mode, while achieving high dielectric capacitance. We then show that the said LL configuration has enormous magnetoelectric charge coefficients ($\partial Q_{ME}/\partial H$), offering the potential to switch a notable fraction of the spontaneous polarization of the piezoelectric layer with moderate ac magnetic fields.

Figure 1(a) shows an illustration of our $Pb(Zr,Ti)O_3$ piezoelectric multilayer stacked together with a magnetostric-

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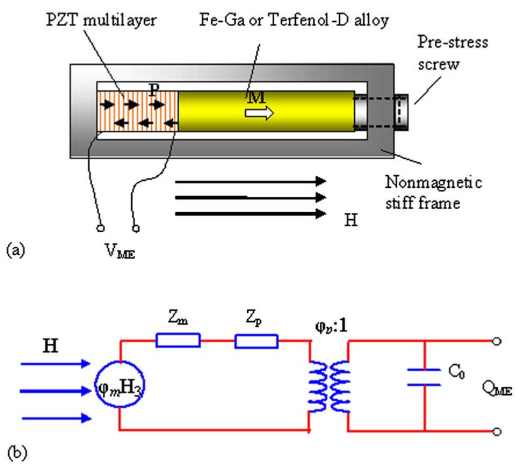

FIG. 1. (Color online) (a) Illustration of our longitudinally magnetized and longitudinally poled or LL configuration, magnetostrictive rod/piezoelectric PZT multilayer composite in a prestressed frame and (b) illustration of a magnetoelectric equivalent circuit for this configuration, where $\varphi_m = A_m d_{33,m}/s_{33}^H$, $Z_M = Z_{1m} + Z_{2m}$, $Z_{1m} = j\rho_m \nu_m A_m \tan(k_m l_m/2)$, $Z_{2m} = \rho_m \upsilon_m A_m / j \sin k_m l_m$, $\varphi_p = g_{33} C_{p0}/s_{33}^D$, $Z_P = Z_{1p} + Z_{2p}$, $C_0 = NC_{p0}$, $C_{p0} = A_p/\bar{\beta}_{33} l_p$, $Z_{1p} = j\rho_p \upsilon_p A_p \tan(Nk_{pe} l_p/2)$, $Z_{2p} = \rho_p \upsilon_p A_p / j \sin Nk_{pe} l_p$, $k_{pe} = \omega/\upsilon_{pe}$, and $\upsilon_{pe} = 1/\sqrt{\rho_p s_{33}^E}$.

tive rod in a longitudinal-longitudinal configuration. The multilayer piezoelectric was symmetrically poled in its thickness direction, attached on one end of a magnetostrictive Fe–17 at. % Ga and/or Terfenol-D rod in a prestressed frame, and then operated in a LL mode. Note that this prestressed LL mode ME composite is notably different from previously reported ME laminated ones in which an epoxy resin was used to attach magnetostrictive and piezoelectric layers together without stress bias.[3,5–8] Especially, when working under high magnetic field drives, a prestressed configuration will notably decrease force-transfer losses between magnetostrictive and piezoelectric phases in ME composites. Under a magnetic field H applied along the longitudinal-axis direction of the ME composite of Fig. 1(a), the magnetostrictive rod is excited into a longitudinal motion, and the multilayer-piezoelectric stack, prestressed together with the magnetostrictive rod, is then forced to vibrate along that direction.

We can determine the magnetoelectric charge coupling (Q_{ME}) of the LL configuration in Fig. 1(a) using an equivalent circuit,[5] instead of the conventional Green's function method:[12,13] the former being much simpler in derivations than the latter. By coupling the constitutive equations of the magnetostrictive rod and multilayer-piezoelectric stack through an equation of motion, determining the forces (and mechanical/electrical currents) transmitted by each phase to the other at the interface, noting in the LL configuration that the magnetostrictive rod is mechanically connected in series to the multilayer piezoelectric and that mechanical displacement velocities at the interphase interfaces are continual, and supposing that the prestress frame is made from a nonmagnetic material with a high elastic stiffness (i.e., the mechanical velocities of the phases at the ends are zero), we can obtain the equivalent model given in Fig. 1(b). From which, we can estimate the Q_{ME} induced across the piezoelectric multilayer by a H_{ac} applied along the longitudinal axis of the magnetostrictive rod, given as

$$Q_{\mathrm{ME}} = \frac{d_{33,m} d_{33,p} N A_m A_p}{s_{33}^E A_m N l_p / l_m + s_{33}^H A_p / (1 - k_{33,p}^2)} H_{\mathrm{ac}}, \tag{1}$$

where A_m and A_p are cross-sectional areas of magnetostrictive rod and piezoelectric multilayer, respectively, N the number of layers in the piezostack, l_m and l_p the magnetostrictive rod length and the thickness of an individual layers in the piezoelectric stack, respectively, and s_{33}^H and s_{33}^E the elastic coefficients of magnetic and piezoelectric phases, respectively. This simple relationship provides important *insights into the physics of how Q_{ME} can be enhanced* in LL mode ME composites: through an increased number of dielectric capacitance layers that are thinner and by increasing the values of ($d_{33,m}$) and ($d_{33,p}$) via application of prestress.

EXPERIMENTS AND RESULT DISCUSSION

We made ME composites following the configuration of Fig. 1(a): magnetostrictive materials were grown by Etrema Products by a zone-melt method, which were cut into rods of 6.35 mm in diameter and 50 mm in length, each of which was then stacked together with a PZT piezoelectric (d_{33}) multilayer with dimensions of $5 \times 5 \times 18\ \mathrm{mm}^3$ (PZT-5H, APC, Mackeyville, PA), and finally assembled into a stiff nonmagnetic frame with a suitable prestress (3–5 MPa). In the piezoelectric multilayer, the polarization of each successive piezoelectric layer was reversed with respect to the prior, along the thickness direction. All N (N=140) piezoelectric layers were connected electrically in parallel with N+1 thin electrodes, yielding a large capacitance of $C_0 = NCp_0(\sim 1.6\mu\mathrm{F})$, where Cp_0 is the static capacitance of one piezoelectric layer. The voltages induced across the PZT multilayer were measured for various dc magnetic biases (H_{dc}) and ac magnetic drives (H_{ac}) over the frequency range of $1 < f < 10^5$ Hz using a lock-in amplifier. An electromagnet was used to apply a dc magnetic bias H_{dc}, and a pair of Helmholtz coils was used to generate a small H_{ac}, via an input current I_{coil}, which was superimposed on H_{dc}.

First, we measured the magnetostrictions of a Terfenol-D rod under free and stress-bias conditions. Figure 2(a) shows the magnetostriction of a Terfenol-D rod as a function of dc magnetic field bias (H_{dc}). We found the maximum magnetostriction λ under low stress bias to be significantly higher than that in the free condition: this is because of a jump effect, as previously reported in Ref. 9 The action of an applied field along the longitudinal-axis direction seemingly induces the magnetization vector of the magnetic domains to change from its initial orientation that is perpendicular to the L axis (due to a preload) to being parallel to the direction along which H is applied. This jump effect in λ is also manifested in a higher effective piezomagnetic coefficient $d_{33,m}$ ($d\lambda/dH$) which subsequently favors a higher ME coefficient, as predicted in Eq. (1). Figure 2(b) shows the $d_{33,m}$ for the Terfenol-D rod as a function of H_{dc} under different stress conditions. These data were calculated from the slope of the magnetostriction λ versus H curve: in this figure, the effec-

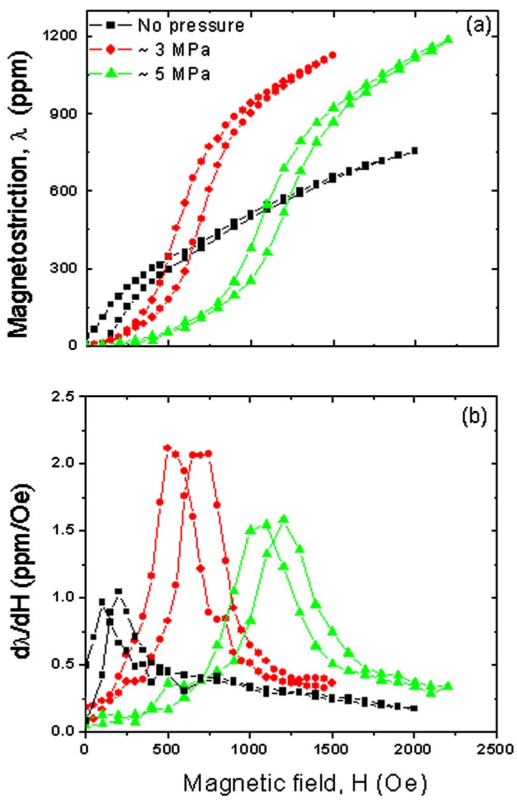

FIG. 2. (Color online) Magnetostrictive properties of a Terfenol-D rod under different stress-bias conditions: (a) magnetostriction λ and (b) effective piezomagnetic coefficient ($d_{33,m}=d\lambda/dH$).

tive $d_{33,m}$ in the stress-biased state of 3 MPa was approximately two times higher than that of the stress-free condition.

Next, we show the magnetoelectric charge coefficient as a function of H_{dc} for our LL configuration of a multilayer PZT stacked with rods of (a) Fe−17 at. % Ga and (b) Terfenol-D, as given in Fig. 3. The data in this figure were taken at a frequency of $f=1$ kHz and a drive of $H_{ac}=1$ Oe. The value of $\partial Q_{ME}^{L,L}/\partial H_{ac}$ can be seen to be strongly dependent on H_{dc}. The results show that the LL mode of Fe–Ga/PZT and Terfenol-D/PZT composites had maximum ME charge coefficients of $\partial Q_{ME}^{L,L}/\partial H_{ac}=6$ and 16 nC/Oe under

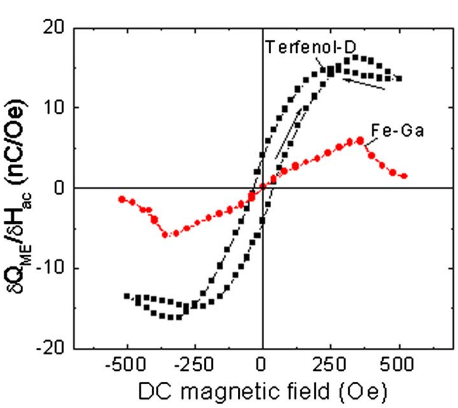

FIG. 3. (Color online) Magnetoelectric charge coefficient of Fe–Ga/PZT and Terfenol-D/PZT multilayer composites as a function of H_{dc} at $f=1$ kHz.

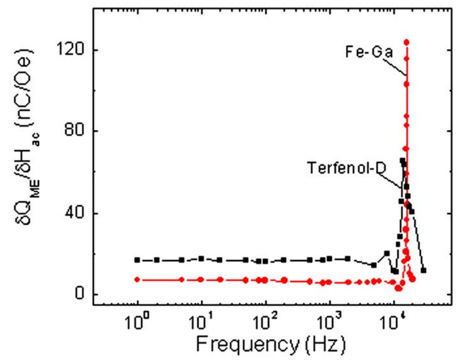

FIG. 4. (Color online) Magnetoelectric charge coefficient as a function of magnetic drive frequency for Fe–Ga/PZT and Terfenol-D/PZT multilayer composites under a constant magnetic bias of $H_{dc}=350$ Oe.

$H_{dc}=350$ Oe, respectively, or correspondingly, the maximum induced polarization changes of $\partial P_{ME}^{L,L}/\partial H_{ac}=1.7\times10^{-6}$ and 4.6×10^{-6}C/m^2 Oe (or s/m), respectively. In both cases, for $H_{dc}>400$ Oe, $\partial Q_{ME}^{L,L}/\partial H$ and $\partial P_{ME}^{L,L}/\partial H_{ac}$ decreased with increasing H_{dc}, as the magnetostrictive alloys approached saturation.

In Fig. 4, we show the frequency dependence of $\partial Q_{ME}^{L,L}/\partial H$ over a wide frequency range of $1<f<10^5$ Hz for a multilayer PZT stacked together with a rod of (i) Fe−17at.%Ga and (ii) Terfenol-D. The measured results show (i) a frequency independent value of $\partial Q_{ME}^{L,L}/\partial H$ ~6 nC/Oe (or $\partial P_{ME}^{L,L}/\partial H_{ac}=1.7\times10^{-6}$ C/m^2 Oe or s/m) and ~16 nC/Oe (or $\partial P_{ME}^{L,L}/\partial H_{ac}=4.6\times10^{-6}$ C/m^2 Oe or s/m) over the quasistatic frequency range of $1<f<2\times10^3$ Hz and (ii) a strong resonance enhancement of $\partial Q_{ME}^{L,L}/\partial H$ at f ~12 kHz, with values as high as 123 nC/Oe ($\partial P_{ME}^{L,L}/\partial H_{ac}=3.5\times10^{-5}$ C/m^2 Oe or s/m) for Fe–Ga/PZT and (60 nC/Oe ($\partial P_{ME}^{L,L}/\partial H_{ac}=2\times10^{-5}$ C/m^2 Oe or s/m) for Terfenol-D/PZT. Clearly, the ME charge coefficients at resonance are 4–20 times higher than those at sub-resonant conditions. Furthermore, in Fig. 4, it can be seen that the Fe–Ga/PZT composites exhibit higher values of $\partial Q_{ME}^{L,L}/\partial H$ than Terfenol-D/PZT ones at resonance, even though the corresponding subresonant values are notably lower: we attribute this to a high mechanical quality factor Q_m in Fe–Ga.

The values of $\partial Q_{ME}^{L,L}/\partial H$ that we report here for PZT-stack and Terfenol-D or Fe–Ga rod composites are ~100–1000 times higher than those of prior ME laminates, as can be seen by comparisons with previously reported data given in Table I and subsequently about 10×10^6 times higher than that of the best single phase ME material

TABLE I. ME charge coefficients for previously reported and current values at $f=1$ kHz.

ME composite	TT (Ref. 5)	LT (Ref. 1)	LL (Ref. 5)	Current sample
ME charge coefficient (pC/Oe)	~130	130	9.3	16 000
Static capacitance C_0 (nF)	~2	1.3	0.093	1600
Required H_{dc}	>4000 Oe	400	400	350

Cr_2O_3.[14] Even calculating the ME charge coefficient per unit volume [i.e., $\partial Q_{\rm ME}^{L,L}/\partial H_{\rm ac}(A_m l_m + NA_p l_p)$], our current ME composite is still 10–100 times than that of the prior ME laminate configuration. It is important to note that the resultant polarization changes in our piezostack/magnetostrictive rod composites are quite high, for example, with $H_{\rm ac} = 100$ Oe, it was over 1% —3.5×10^{-3} C/m^2 Oe—of the total polarization of PZT-5H ($P_s \approx 0.3$ C/m^2). This represents a significant advancement in coupling between polarization and magnetic field, accounting for about 10% of the polarization that can be switched reversibly without introducing significant hysteretic losses about a minor hysteresis loop.

Such large ME charge effects require not only a good force transfer between magnetostrictive and piezoelectric layers but in addition that the piezolayer has a high capacitance. This is achieved by combining the high force advantage of the LL configuration with the high capacitance of a multilayer one. A long-type multilayer LL mode ME composite favors the optimum combination of magnetostrictive and piezoelectric effects, and an applied prestress enhances the magnetoelastic couplings. In addition, the total static capacitance C_0 of the piezoelectric multilayer configuration is much higher than that of its corresponding LL one. Note that the ME voltage coefficient in current ME configuration was not enhanced. The possible cause is due to the weak stiffness of the frame (made of brass), which results in a significant elastoelectric coupling loss in piezoelectric stack. Further improvement is necessary for obtaining both high ME charge and voltage couplings in this prestressed ME configuration.

CONCLUSION

In summary, composites of magnetostrictive polycrystalline Fe-17 at. %Ga or Terfenol-D rods and a piezoelectric PZT multilayer stack were constructed. Our experimental results confirm (i) a two times enhancement in effective piezomagnetic coefficient $d_{33,m}$ under stress bias, (ii) a large LL ME charge coefficient of $\partial Q_{\rm ME}^{L,L}/\partial H \sim 16$nC/Oe under moderate dc magnetic biases, which is 100–1000 times higher than that previously reported for any other laminate configuration, and (iii) a dramatic enhancement in the ME charge coupling coefficient to ~120 nC/Oe near the resonance frequency. Our finding has potential applications as magnetoelectric converters or energy harvesters.

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