AIP Applied Physics Letters

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Citation: Applied Physics Letters **87**, 062502 (2005); doi: 10.1063/1.2007868
View online: http://dx.doi.org/10.1063/1.2007868
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/87/6?ver=pdfcov
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Push-pull mode magnetostrictive/piezoelectric laminate composite with an enhanced magnetoelectric voltage coefficient

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(Received 21 March 2005; accepted 27 June 2005; published online 1 August 2005)

A magnetoelectric (ME) laminate composite consisting of a symmetric longitudinally poled piezoelectric $Pb(Mg_{1/3}Nb_{2/3})O_3-PbTiO_3$ crystal and two longitudinally magnetized magnetostrictive $Tb_{1-x}Dy_xFe_2$ layers has been developed that has a notably superior ME voltage coefficient, relative to previous laminate configurations. The symmetric nature of the longitudinally poled piezoelectric layer allows for operation in a *push-pull* mode that optimizes elastic coupling between layers. Our small laminate has a giant ME voltage coefficient of ~1.6 V/Oe at low frequencies, a significant enhancement of this coefficient to ~20 V/Oe under resonance drive, and an exceptional low-level magnetic field sensitivity of $\sim 10^{-12}$ T at $f=f_0$. © *2005 American Institute of Physics*. [DOI: 10.1063/1.2007868]

Magnetic-electric field conversion using variable reluctance coils based on Faraday's electromagnetic induction law is well known. Induced voltages using variable reluctance coils are proportional to the change rate of a magnetic field's flux, and the voltage strength therefore decreases with decreasing flux change rate.[1] Although other physical effects such as Hall,[2] magneto-optoelectronic,[3] and magnetoresistive ones[4] can be used for magnetic field detection, the applied magnetic field does not generate an electric field.

The magnetoelectric (ME) effect of a two-phase magnetostrictive/piezoelectric composite has significant potential for magnetic-electric field coupling.[5–16] In such laminates, the ME effect is a product tensor property combining the magneto-elastic and elasto-electric effects: an applied magnetic field (H) produces an elastic strain in the magnetic phase via magnetostrictive, which is then coupled to the piezoelectric phase inducing an electric field (E) across it via piezoelectricity. Theoretically, the elastically coupled magnetic and electrical energies can be entirely and reversibly transduced between corresponding forms—that is if there are no eddy-current losses in the magnetic phase, or any dielectric losses in the piezoelectric one. Large ME effects have been reported in 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 with magnetostrictive $Tb_{1-x}Dy_xFe_{2-y}$ (TERFENOL-D), Permendur, $Ni_{1-x}Co_xFe_2O_4$, or $Co_{1-x}Zn_xFe_2O_4$ ones.[6–16]

In this letter, we demonstrate that a laminate consisting of a *symmetric-longitudinally poled* piezoelectric PMN-PT crystal sandwiched between two longitudinally magnetized magnetostrictive TERFENOL-D layers—as shown in Fig. 1—has a significantly enhanced ME voltage coefficient relative to prior laminate composite configurations.

The magnetostrictive TERFENOL-D layers were of dimensions $12.7\times 6\times 1$ mm^3. They were (211) grain oriented along the length direction, which has the highest longitudinal magnetostrictive strain ($\lambda \sim 1000$ ppm). The piezoelectric PMN-PT crystal layers were of dimensions $15\times 6\times 1$ mm^3. These crystals were oriented along the (001), which has the highest longitudinal piezoelectric and electromechanical coupling coefficients. For TERFENOL-D layers magnetized along their longitudinal axis (i.e., L mode), magnetostrictive measurements of a laminate prototype (see Fig. 1) revealed a maximum longitudinal magnetostriction (expansion) of ~900 ppm under a dc magnetic bias of $H_{dc}=1500$ Oe. However, when these same layers were magnetized in their thickness directions (i.e., T mode), the maximum longitudinal magnetostriction (contraction) was only 330 ppm, even under a much higher dc bias of $H_{dc}=3000$ Oe. Clearly, the principle magnetization of the laminate configuration in Fig. 1 should be chosen to lie along the longitudinal axis of the laminate.

Under an applied H_{ac}, a symmetric expanding/contracting ac elastic strain will be excited about the center line at $\ell/2$ (ℓ is the laminate length) of the TERFENOL-D layers via magnetostriction. Correspondingly, the piezoelectric layer will also be driven into cyclical strain/motion, and an E field will be induced across it via piezoelectricity. This is the dual (magneto-elasto-electric) coupling that results in ME product tensor properties. As illustrated by color contours in Fig. 1, analysis by finite element methods showed that the center position of the piezoelectric and magnetostrictive layers is located on a stress concentration line, whereas, the two ends are located at stress-free points. The maximum stress-induced piezoelectric voltage will be found between

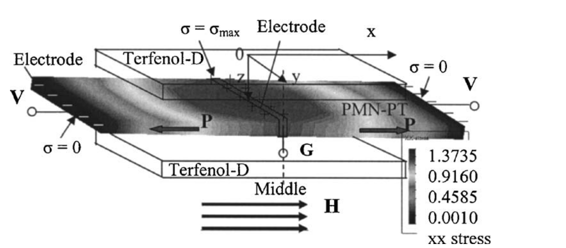

FIG. 1. Push-pull ME laminate having two longitudinal magnetization magnetostrictive TERFENOL-D layers sandwiching one symmetric longitudinally polarized piezoelectric PMN-PT layer. The induced ME voltage was measured between center and end electrodes of the piezoelectric layer, while it is being stressed by two magnetostrictive layers under an applied magnetic field H. The center of the piezoelectric layer has the maximum stress concentration (as shown in the figure), whereas the ends are stress free. The static capacitance of the PMN-PT layer between the center and end electrodes was 66 pF.

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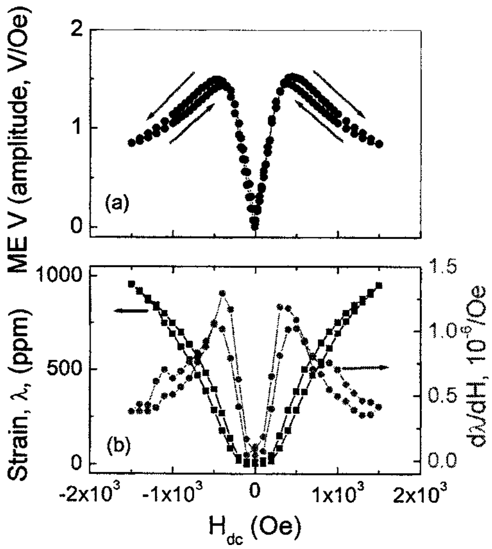

FIG. 2. (a) Dependence of the ME voltage coupling coefficient on an applied dc magnetic bias, for $H_{ac}=1$ Oe and $f=10^3$ Hz; and (b) magnetostriction strain and its differential (i.e., the effective piezomagnetic coefficient) as a function of dc magnetic bias.

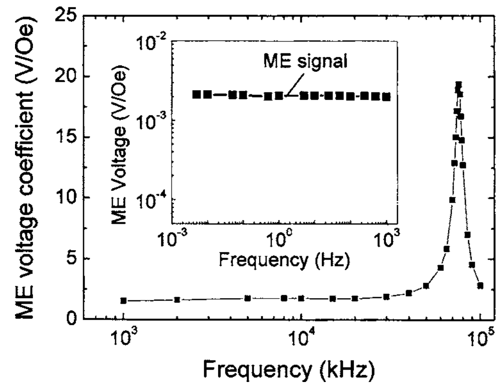

FIG. 3. Dependence of ME voltage coefficient on the frequency of the ac magnetic field over the range of $5\times10^{-3}<f<10^5$ Hz. The figure shows the resonance enhancement for $f>10^3$ Hz; whereas the inset shows the flat frequency response for $f<10^3$ Hz. These data were taken using a $H_{dc}=500$ Oe, and a $H_{ac}=1$ Oe.

the center and end electrodes, as between these points the stress difference is largest. To take advantage of this fact, the piezoelectric layer in the laminate configuration of Fig. 1 was symmetrically poled along its longitudinal direction about the centerline; and thus, the induced ME voltage difference from the center electrode to either end electrode is the same. In *L-L* mode laminates,[13] the induced ME voltage is measured between two end electrodes of one longitudinal-polarized piezoelectric layer. These two end electrodes are in reverse polarity, but are located at stress-free areas; thus, the induced ME electric voltage of *L-L* mode laminates is small, although it is bigger than either *L-T* or *T-T* ones. Because of the symmetric nature of the expanding/contracting motion of our new laminate in Fig. 1 about the centerline, we designate the ME coupling mode as *push-pull*.

The induced ME voltage for the push-pull laminate was measured at various H_{dc} and H_{ac} in the frequency range of $5\times10^{-3}<f<10^5$ Hz using a charge amplifier method combined with a lock-in amplifier technique. A pair of Helmholtz coils was used to generate a small H_{ac}, via an input current I_{coil}. The ME voltage coefficient $\partial V_{ME}/\partial H$ used in this letter designates that measured between the centerline and either (or both) ends of the PZT layer in the laminate configuration of Fig. 1, under a given H_{ac}.

The ME voltage coefficient $\partial V_{ME}/\partial H$ of push-pull laminates was found to be strongly dependent on H_{dc} [see Fig. 2(a)]. This reflects the fact that $\partial V_{ME}/\partial H$ has a differential dependence of the magnetostriction λ on magnetic bias H (i.e., $\delta\lambda/\delta H$, or effective piezomagnetic coefficient) [see Fig. 2(b)]. These data were taken at a frequency of $f=1$ kHz and a drive of $H_{ac}=1$ Oe. The results show that push-pull trilayer laminates of TERFENOL-D/PMN-PT/TERFENOL-D have a giant ME effect: the maximum ME voltage coefficient at low frequency was ~1550 mV/Oe at $H_{dc}=450$ Oe, which is close to the dc bias at which $\delta\lambda/\delta H$ was also maximum ($\sim1.2\times10^{-6}$ Oe^{-1}). This value of $\partial V_{ME}/\partial H$ for our push-pull laminate is ~4x higher than that of prior *L-L* modes[13] and ~14× higher than that of *L-T* ones.[14] For $H_{dc}>450$ Oe, the value of $\partial V_{ME}/\partial H$ decreased significantly with increasing H_{dc}, as the effective piezomagnetic coefficient decreased.

The ME voltage coefficient of our push-pull laminate was then measured over a frequency range of $5\times10^{-3}<f<10^5$ Hz, using a $H_{ac}=1$ Oe. A flat frequency response was found for $f<10^3$ Hz, as can be seen in the inset of Fig. 3. A giant induced ME voltage of ~1.6 V/Oe was nearly unchanged with decreasing *f*, even at an extremely low frequency (ELF) of 5 mHz. For such ELF H-field variations, variable reluctance coils fail to provide a detectable magnetic-electric field coupling signal, as their inductance is negligible due to an extremely low change rate of the magnetic field's flux. At higher frequencies of $f>10^3$ Hz, the results in Fig. 3 for our push-pull laminate showed a significant enhancement in the ME voltage coefficient with increasing *f*, as the resonance condition was approached. At a resonance frequency of ~78 kHz, the value of $\partial V_{ME}/\partial H$ was dramatically increased to ~20 V/Oe (or a corresponding ME field coefficient of ~30.8 V/cm Oe). This is ~12.5× higher than that of the lower frequency data.

Next, measurements of low-level magnetic field variations were performed for our push-pull laminate at a relatively low frequency of $f=1$ Hz. Measurements were performed in a μ-metal magnetically shielded environment at room temperature. The value of $\partial V_{ME}/\partial H$ can be seen in Fig. 4 to be a near linear function of H_{ac} over the range of approximately $3\times10^{-11}<H_{ac}<10^{-3}$ T (or $3\times10^{-7}<H_{ac}<10$ Oe). We then confirmed that even significantly higher sensitivities to minute H-field variations might be achieved using our push-pull laminate over a narrow bandwidth around the resonance frequency of $f=77.5$ kHz. Under resonant operation, a near linear dependence of $\partial V_{ME}/\partial H$ on H_{ac} can be seen in Fig. 4. Finally, the inset of Fig. 4 shows that a minute H-field variation of $H_{ac}=1.2\times10^{-12}$ T at $f=77.5$ kHz is still detectable under resonant operation. Recently, a magnetoresistive sensor has been reported to detect minute H-field variations of $\sim3\times10^{-12}$ T at $f=1$ Hz;[4] however, this was achieved only at 77 K using a current of 5 mA. Our new push-pull mode laminate operates at room temperature and consumes much less power. Furthermore, improvements in magenetostrictive and piezoelectric materials, laminate configurations, and operational modes have not yet even been considered. Clearly, our push-pull ME laminate may be

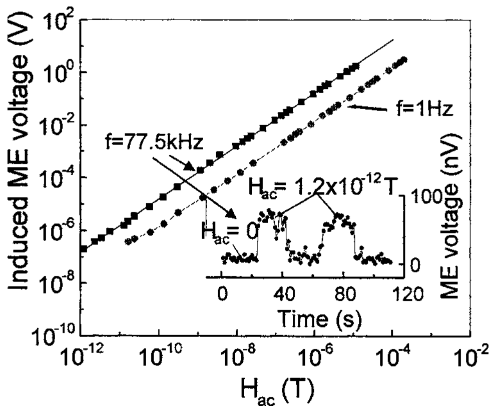

FIG. 4. Magnetic field sensitivity measurements taken under a dc magnetic bias of 450 Oe. The figure shows the induced ME voltage as a function of magnetic field over the range of $10^{-12} < H_{ac} < 10^{-3}$ T at drive frequencies of $f=1$ Hz and $f=77.5$ kHz (resonance condition); whereas the inset of the figure shows the change in the ME voltage as a function of time, in response to a minute magnetic field variation of 1.2×10^{-12} T.

quite important for applications requiring detection of minute magnetic fields variation at ambient conditions.

In summary, we have developed a push-pull mode trilayer laminate. Our push-pull laminate configuration optimizes the elastic interactions between magnetostrictive and piezoelectric layers by use of a symmetric poling of the piezoelectric layer about its centerline, where the stresses are maximum. This then results in a significantly increased ME voltage coefficient—approximately 4x and 14x higher than prior *L-L* and *L-T* mode laminates, respectively. In addition, we have demonstrated a sensitivity to minute H-field variations down to $\sim 10^{-12}$ T at room temperature and under resonant conditions.

This research was supported by the Office of Naval Research under Grant Nos. N000140210340, N000140210126, and MURI N000140110761.

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