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Magnetoelectric effect in Terfenol-D/Pb(Zr,TiO)$_3$/μ-metal laminate composites

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The authors have found that the required dc magnetic bias of magnetoelectric (ME) laminates can be significantly altered, by incorporating additional high-permeability μ-metal layers. Investigations have focused on $\mathrm{Tb}_{1-x}\mathrm{Dy}_x\mathrm{Fe}_{2-y}/\mathrm{Pb}(\mathrm{Zr}_{1-x}\mathrm{Ti}_x)\mathrm{O}_3/\mu$-metal laminates. The authors' results show that laminated μ-metal layers oriented perpendicular to an ac magnetic field concentrate flux in the composite, whereas those parallel to it attenuate flux. This results in (i) a significant decrease in the required dc magnetic bias H_{dc}, and (ii) an effective enhancement in the ME voltage coefficients at low H_{dc}, by a factor of up to 7.6. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2355459]

Composites consisting of magnetostrictive $\mathrm{Tb}_{1-x}\mathrm{Dy}_x\mathrm{Fe}_{2-y}$ (Terfenol-D), Permendur, $\mathrm{Ni}_{1-x}\mathrm{Co}_x\mathrm{Fe}_2\mathrm{O}_4$, or Galfenol layers laminated with piezoelectric Pb(Zr,Ti)O$_3$ (PZT) or $\mathrm{Pb}(\mathrm{Mg}_{1/3}\mathrm{Nb}_{2/3}\mathrm{O}_3-\mathrm{PbTiO}_3)$ ones are known to have large magnetoelectric (ME) effects,[1–14] offering potential applications as magnetic field sensors and transformers. However, these laminate composites exhibit significant ME effects only near an optimum dc magnetic bias $H_{\mathrm{dc,opti}}$, where the effective piezomagnetic coefficient (i.e., $d\lambda/dH$) of the magnetostrictive layers is maximum.[14] Typical values of $H_{\mathrm{dc,opti}}$ for ME laminates operated in a longitudinally magnetized longitudinally polarized (or LL) mode are about 500 Oe, whereas those operated in transversely magnetized transversely polarized and transversely polarized (TT) longitudinally polarized (TL) modes have an $H_{\mathrm{dc,opti}} \approx 4000$ Oe.[2,9] Here, we report that $H_{\mathrm{dc,opti}}$ can be notably shifted over a wide range of values by flux concentration, via colamination with high-permeability μ-metal layers into the ME composite. This, in turn, results in a significant enhancement in the ME voltage coefficient at low biases. We believe that our flux concentration method offers a unique approach to enhancing the sensitivity and directionality of ME magnetic sensors.

Under an applied magnetic field $\boldsymbol{H}$, the magnetic induction $\boldsymbol{B}$ $(=\mu_0\mu_r\boldsymbol{H})$ inside a ferromagnetic material is strongly related to that material's relative permeability μ_r. Although magnetostrictive Terfenol-D has a large magnetostriction, its μ_r is quite low ($\sim 3-10$), yielding a relatively high $H_{\mathrm{dc,opti}}$ (~ 500 Oe) for Terfenol-D/PZT (or T-D/PZT) laminates. However, by incorporating high-permeability ferromagnetic layers into ME T-D/PZT laminates, a higher effective permeability can be achieved, and thus, a larger effective piezomagnetic or magnetostrictive coefficient, $d\lambda/dH$, in Terfenol-D and a stronger ME coupling in the ME composite at lower $H_{\mathrm{dc,opti}}$. Magnetic flux concentrators (high-permeability amorphous alloys) have recently been used in Hall devices.[15] Figures 1(a) and 1(b) illustrate two configurations of T-D/PZT ME laminates, colaminated with μ metal using a thin soft silicone glue layer. The dimensions of the T-D/PZT/T-D three-layer laminate were 15 mm in length, 6 mm in width, and 4 mm in thickness, and those of the μ-metal layers were 12 mm in length, 7 mm in width, and 4 mm in thickness. The high-permeability *μ-metal* (AD-MU-80, AD-Vance Magnetics) layers were 80% nickel-iron alloys with a relative permeability of up to 100 000. Part (a) shows a configuration with two μ-metal layers fixed to the long ends of a rectangularly shaped Terfenol-D/PZT ME composite. The μ metal acts to concentrate external flux in the laminate that is parallel to its long axis, which as we will subsequently show shifts $H_{\mathrm{dc,opti}}$ to lower values. Part (b) shows a configuration with two μ-metal layers fixed to the top/bottom sides of a T-D/PZT/T-D thee-layer laminate. In this case, the μ-metal layers again act to concentrate flux in the laminate that is perpendicular to its length axis, but attenuate flux in laminates that are parallel to that axis, which as we will subsequently show shifts $H_{\mathrm{dc,opti}}$ to higher values.

The magnetic flux in the ME laminates was measured using a search coil method, and the effect of μ-metal layers on the magnetic field distributions was observed by a conventional ferromagnetic powder method. The magnetostriction λ for both a Terfenol-D layer and a T-D/μ-metal laminate was measured as a function of dc magnetic bias (H_{dc}), via strain gauge method, and subsequently, the effective magnetostrictive coefficients ($d_{33,m}$ or $d_{31,m}$) were calculated, i.e., $d\lambda/dH_{\mathrm{dc}}$. The voltages induced across the two ends of the PZT layers were measured as a function of dc magnetic bias (H_{dc}) in response to a constant ac (1 kHz) magnetic drive of $H_{\mathrm{ac}}=1$ Oe, which was parallel to H_{dc}, via a lock-in amplifier. These measurements were performed at room temperature. An electromagnet was used to apply H_{dc}, and a pair of Helmholtz coils was used to generate a small H_{ac}. For LL mode composites with static capacitances close to or lower than the distributed capacitances of the connecting cables and electronics, we also used a charge (pre)amplifier.

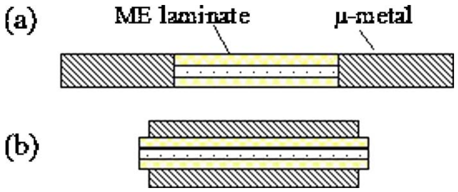

FIG. 1. Side view illustration of two configurations for Terfenol-D/PZT/Terfenol-D three-layer ME laminates, which incorporate μ-metal layers: (a) μ-metal layers attached at both ends of the ME laminate and (b) μ-metals layers attached at both sides of the ME laminate.

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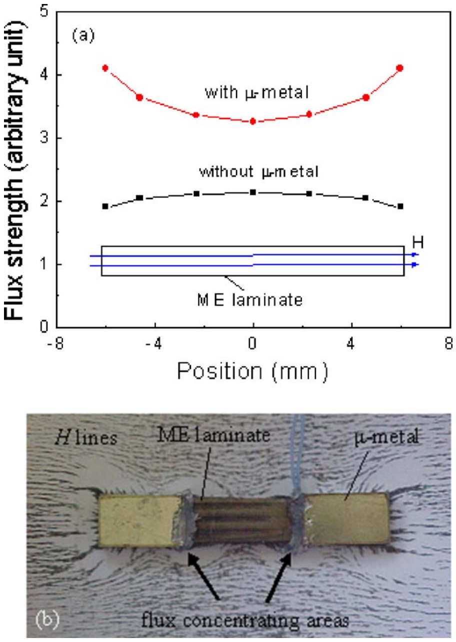

FIG. 2. Demonstration of flux concentration in ME laminates by μ-metal layers: (a) flux in laminate with/without μ-metal layers and (b) photograph of magnetic field distribution.

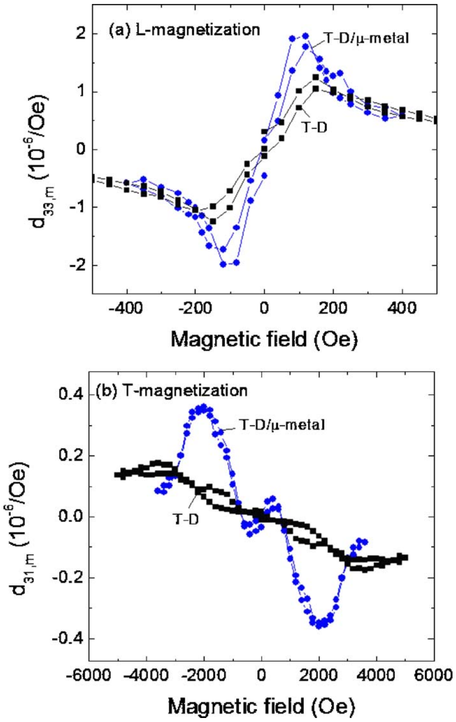

FIG. 3. Effective magnetostrictive coefficients of a single Terfenol-D layer with/without μ-metal layers that is (a) longitudinally magnetized with μ-metal layers at ends and (b) transversely magnetized with μ-metal layers on sides. These data were taken at room temperature.

First, we directly observed magnetic flux lines and flux concentration effect in ME laminates after incorporating μ-metal layers. Figure 2(a) shows how the flux lines are altered by μ-metal layers placed at the end of the laminate, and Fig. 2(b) provides a photo demonstrating how the field distribution is affected. In part (a), it can be seen that the flux is enhanced by a factor of ~2 by the incorporation of μ-metal layers. Clearly, the effect of high-permeability μ-metal layers placed at the two ends of the laminate is to concentrate flux.

Next, we observed the effects of μ-metal layers on the effective magnetostrictive coefficients of a single Terfenol-D layer (i.e., without a PZT layer), as given in Fig. 3 for (a) a longitudinally magnetized layer with μ-metal end layers, i.e., $d_{33,m}$, and (b) a transversely magnetized one with μ-metal side layers, i.e, $d_{31,m}$. Both figure parts show the effective piezomagnetic coefficients as a function of H_{dc} for a Terfenol-D layer and a T-D/μ-metal laminate. Significant changes in the effective magnetostrictive coefficient can be seen for the T-D/μ-metal laminate, relative to the Terfenol-D single layer: (i) $H_{dc,opti}$ can be seen to be decreased for both modes by nearly 50% by the μ-metal layers, and (ii) the value of $d_{33,m}$ can be seen to be increased by nearly a factor of 2 a constant bias of 100 Oe, and that of $d_{31,m}$ by a factor of ~4 a constant bias of 1000 Oe. Clearly, the μ metal acts to concentrate external flux into the laminate, resulting in a higher effective piezomagnetic coefficient.

Figure 4(a) shows the ME voltage coefficient (V_{ME}) as a function of H_{dc}, both before and after colamination with μ-metal end layers operated in a LL mode. Inspection of this figure will reveal (i) that μ-metal end layers significantly decrease $H_{dc,opti}$ to 240 Oe, relative to ~500 Oe for the T-D/PZT laminate (comparisons with the T-D/μ-metal composite of Fig. 3 will reveal that PZT layers effectively act as a load increasing $H_{dc,opti}$ relative to the T-D/μ-metal composite), and (ii) that values of $V_{ME} \geqslant 1400$ mV/Oe can be achieved under $H_{dc}=240$ Oe for laminates with μ-metal end layers. At lower dc magnetic biases of $H_{dc} < 100$ Oe this enhancement in V_{ME} was increased by a factor of ~2 (please note that both the magnetic flux amplification and the enhancement of $d_{33,m}$ were also increased by a factor of ~2). These results demonstrate that μ-metal layers, fixed to the two ends of the ME laminate along the direction of the flux lines, concentrate the flux lines that pass through the laminate, apparently increasing V_{ME} at lower magnetic biases. This can be attributed to an increase in the effective permeability of the ME colaminate, resulting in stronger magnetic induction $\boldsymbol{B}$ [see Fig. 2(b)], in turn resulting in higher effective $d_{33,m}$ and V_{ME} coefficients.

Figure 4(b) shows V_{ME} as a function of H_{dc}, both before and after colamination with μ metal to both sides of the ME composite [see Fig. 1(b)] operated in the TL mode. A significant decrease in $H_{dc,opti}$ for the T-D/PZT μ-metal laminate can be seen. Without μ-metal layers, the TL mode had much higher values of $H_{dc,opti} \geqslant 4000$ Oe than the LL mode, and much lower values of V_{ME} than that previously reported at fields of $H_{dc} < 700$ Oe.[8,14] However, upon incorporation of μ-metal layers on the top/bottom sides of the laminate, $H_{dc,opti}$ was decreased to 700 Oe; in addition, the maximum value of V_{ME} was enhanced from ~670 mV/Oe under H_{dc} $=4000$ Oe to ~900 mV/Oe under $H_{dc}=700$ Oe. A more notable increase can be seen by comparing data taken at a constant bias of $H_{dc}=500$ Oe, where V_{ME} was increased by a factor of up to ~7.6 upon incorporation of the μ-metal layers. Finally, we show V_{ME} as a function of H_{dc}, both before and after colamination with μ metal to both sides of the ME composite [again see Fig. 1(b)] operated in the LL mode, as given in Fig. 4(c). The results show (i) a notably reduced V_{ME} by up to 85% over the lower biases range of $0 < H_{dc}$ < 300 Oe and (ii) an increase of $H_{dc,opti}$ from $H_{dc}=500$ Oe (without μ-metal layers) up to 1000 Oe (with μ-metal lay-

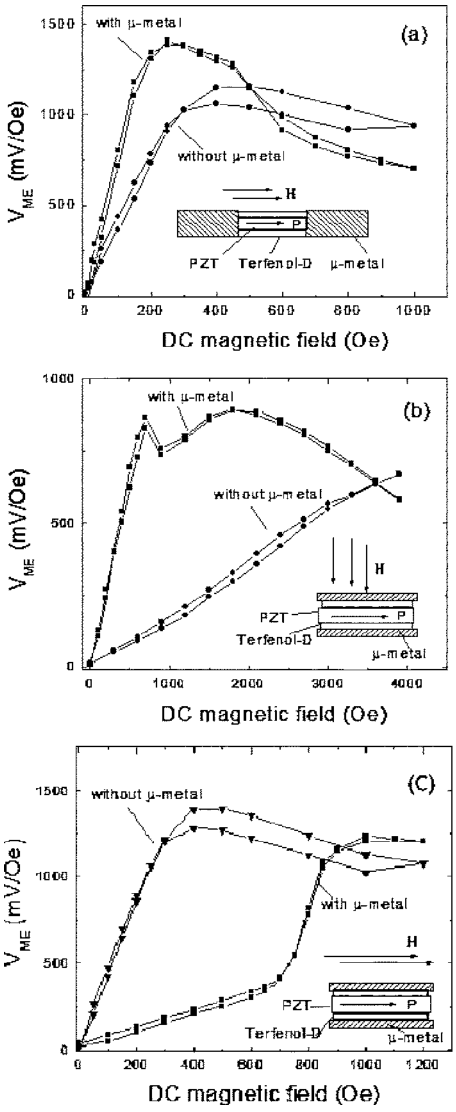

FIG. 4. Changes of required magnetic bias in Terfenol-D/PZT ME laminates colaminated with μ-metal (a) end caps, operated in a LL mode, (b) layers on both sides of the laminated, operated in a TL mode, and (c) layers on both sides of the laminated, operated in a LL mode. These data were taken at room temperature.

ers). In the case of Fig. 4(c), the flux is constrained within the μ-metal layers, causing a weaker magnetic induction $\boldsymbol{B}$ (or lower effective $d_{33,m}$); consequently, V_{ME} is decreased under a constant applied H_{dc}, and the maximum in V_{ME} shifted to higher values of $H_{\mathrm{dc,opti}}$.

We can predict how the μ-metal layers influence the ME voltage coefficient(s) V_{ME} both when the high-permeability layers are fixed (i) to the ends of the long axis of T-D/PZT laminates [see Fig. 1(a)], operated in a LL mode, and (ii) to both sides of T-D/PZT laminate [see Fig. 1(b)], operated in an TL mode. Following our prior equivalent scheme,[8] we know that V_{ME} for the LL (TL) laminates is a linear function of $d_{33,m}$ ($d_{31,m}$), given below as a simple proportionality for convinience:

$$\left|\frac{dV}{dH_3}\right| = V_{\mathrm{ME}} \sim d_{3i,m} \tag{1}$$

($i=1$ for TL mode and $i=3$ for the LL mode).

Clearly, higher values of $d_{33,m}$ and/or $d_{31,m}$, due to μ-metal layer flux concentration effects, will enhance V_{ME}. The changes in V_{ME} given in Fig. 4 can be understood using Eq. (1). For example, consider the LL mode, where the value of V_{ME} was nearly doubled at a constant bias of $H_{\mathrm{dc}}=100$ Oe, consistent with the doubling of $d_{33,m}$ [see Fig. 3(a)] and likewise of the flux [see Fig. 2(a)]. As another example, consider the TL mode, where V_{ME} was increased by a factor of between 3 and 4 at a constant bias of 1000 Oe, consistent with the proportional change in $d_{31,m}$ [see Fig. 3(b)].

Clearly, V_{ME} can be enhanced by the increase of the effective piezomagnetic coefficient due to flux concentration by colamination with μ-metal layers. Such concentration[15,16] may be optimized by using (i) a longer μ-metal layer (to increase the length ratio of μ-metal layers relative to that of the ME laminate) and (ii) T-shaped μ-metal layers. (to increase the area ratio of μ-metal layers relative to the ME laminate). In fact, our investigations revealed that $H_{\mathrm{dc,opti}}$ could be decreased to 60 Oe and V_{ME} increased by up to five times by use of longer μ-metal layers that further increase the average permeability of the composite (data not shown). Additionally, the anisotropy of the ME properties can be controlled, enabling the sensing of magnetic vector components along particularly defined axes, while rejecting that of other directions. Accordingly, we believe that this flux concentration method offers a unique approach to developing highly sensitive magnetic field sensors with strong directionality.

In summary, the effective permeability, magnetic induction strength, and magnetostrictive coefficient in ME laminates can be significantly altered by the incorporation of μ-metal layers into laminate composites of Terfenol-D and PZT; correspondingly, the $H_{\mathrm{dc,opti}}$ required to induce maximum ME properties can also be changed. Laminated μ-metal layers oriented perpendicular to the flux concentrate it, reducing $H_{\mathrm{dc,opti}}$, whereas those parallel to it attenuate flux.

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