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Enhanced magnetoelectric effect in three-phase $MnZnFe_2O_4/Tb_{1-x}Dy_xFe_{2-y}/Pb(Zr,Ti)O_3$ composites

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Three-phase magnetoelectric (ME) laminate composites made of high-permeability (μ) $MnZnFe_2O_4$, magnetostrictive $Tb_{1-x}Dy_xFe_{2-y}$, and piezoelectric $Pb(Zr,Ti)O_3$ layers and operated in longitudinal magnetization and transverse polarization (*L-T*) bending modes have been studied. High-μ $MnZnFe_2O_4$ layers act as flux concentrators. This results in increased apparent piezomagnetic coefficients in the magnetostrictive $Tb_{1-x}Dy_xFe_{2-y}$ layer and consequently larger induced voltages across the piezoelectric $Pb(Zr,Ti)O_3$ layers. We find the enhanced ME field coefficients of up to 8 times for longitudinal magnetization mode, and up to 28 times for transverse magnetization mode, under small dc magnetic field bias. © *2006 American Institute of Physics.*
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I. INTRODUCTION

The magnetoelectric (ME) effect is a polarization response to an applied magnetic field or conversely a magnetization response to an applied electric field. Two-phase magnetostrictive/piezoelectric laminated composites are known to have giant ME effects.[1–10] Giant ME field coefficients have been reported in *L-T* (longitudinally magnetized transversely poled) composites of piezoelectric $Pb(Zr,Ti)O_3$ (PZT) layers laminated with magnetostrictive $Tb_{1-x}Dy_xFe_{2-y}$ or Terfenol-D (Ref. 1) [$\alpha_{ME}^{(L\text{-}T)}=2$ V/cm Oe], Permendur (Ref. 2) [$\alpha_{ME}^{(L\text{-}T)}=0.8$ V/cm Oe], Fe–Ga (Ref. 3) [$\alpha_{ME}^{(L\text{-}T)}=0.4$ V/cm Oe], or $NiFe_2O_4$ (Ref. 4) [$\alpha_{ME}^{(L\text{-}T)}=0.45$ V/cm Oe] ones. In said laminates, the ME effect is a product tensor property—and not intrinsic to individual layers—combining the magnetoelastic and elastoelectric effects of individual layers, via an elastic coupling between layers: an applied *H* produces an elastic strain in the magnetostrictive phase that is stress coupled to that of the piezoelectric one, resulting in an induced voltage.[11–13]

However, these laminate composites exhibit significant ME effects only near an optimum dc magnetic bias $H_{dc,opti}$, where the effective piezomagnetic coefficient (i.e., $d\lambda/dH$) of the magnetostrictive layers is maximum.[14] Typical values of $H_{dc,opti}$ for ME laminates operated in a longitudinal magnetization *L-L* or *L-T* mode are about 500 Oe, whereas those operated in transverse magnetization *T-T* or *T-L* modes have a much higher $H_{dc,opti}\approx 4000$ Oe,[2,9] because of large demagnetization factor's effect in transverse magnetization modes. Here, we report that $H_{dc,opti}$ can be notably shifted over a wide range of values by flux concentration, via colamination with high-permeability $MnZnFe_2O_4$ layers into ME composites. Figure 1 illustrates the two constructions of three-phase ME laminates consisting of high-μ $MnZnFe_2O_4$, magnetostrictive $Tb_{1-x}Dy_xFe_{2-y}$, and piezoelectric $Pb(Zr,Ti)O_3$ layers. We find large enhancements in the ME field coefficient for both longitudinal and transverse magnetization modes under notably decreased magnetic biases, relative to laminates without colaminated $MnZnFe_2O_4$ layers.

II. FLUX CONCENTRATION AND ENHANCED APPARENT PIEZOMAGNETIC COEFFICIENTS

Under an applied magnetic field $\boldsymbol{H}$, the magnetic induction $\boldsymbol{B}$ inside a ferromagnetic material is strongly related to that material's relative permeability: $\boldsymbol{B}=\mu_0\mu_r\boldsymbol{H}$. Although magnetostrictive Terfenol-D has a large magnetostriction, its μ_r is low ($\leqslant 10$), thus requiring a relatively high bias ($H_{dc,opti}>300$ Oe) to achieve saturation of $\boldsymbol{B}$. However, by incorporating high-permeability ferromagnetic layers into two-phase Terfenol-D/PZT ME laminates, the effective permeability in ME laminates can be significantly increased, producing flux concentration in the composite. In turn, this will result in an apparent increase in the effective piezomagnetic coefficients, $d_{33,m}=\delta\varepsilon_{33}/\delta H$ and $d_{31,m}=\delta\varepsilon_{31}/\delta H$, in the

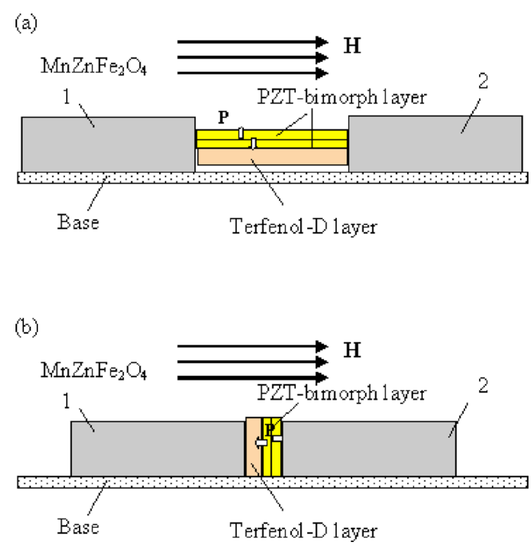

FIG. 1. (Color online) Three-phase $MnZnFe_2O_4$/Terfenol-D/PZT-bimorph magnetoelectric composite: (a) longitudinal magnetization construction and (b) transverse magnetization construction.

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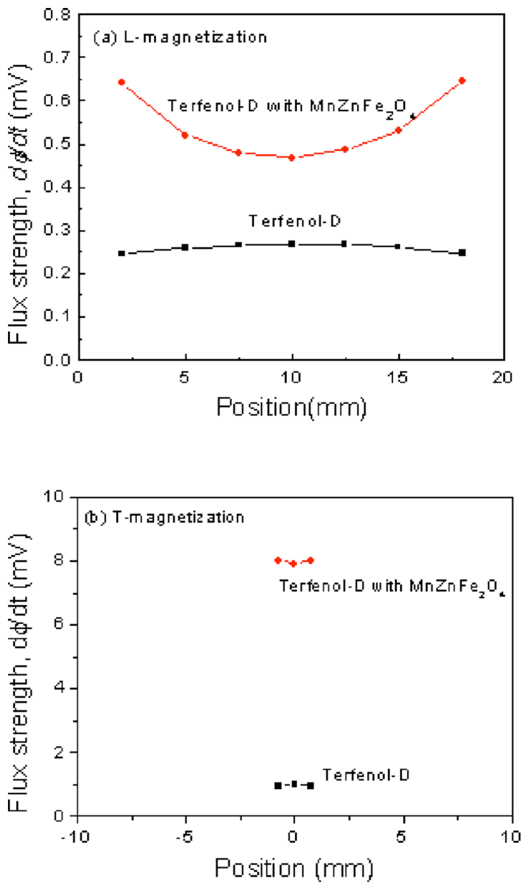

FIG. 2. (Color online) Magnetic fluxes and flux concentration effect in single Terfenol-D layer with/without $MnZnFe_2O_4$ layer for (a) longitudinal magnetization mode and (b) transverse magnetization mode.

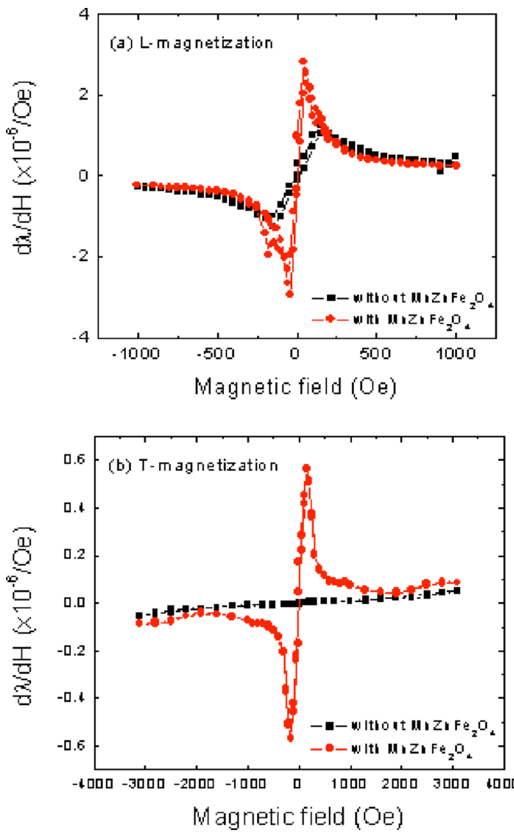

FIG. 3. (Color online) Effective piezomagnetic coefficients of a single Terfenol-D layer with/without $MnZnFe_2O_4$ layer as a function of H_{dc} for (a) $d_{33,m}$ for longitudinal magnetization mode and (b) $d_{31,m}$ for transverse magnetization mode.

magnetostrictive Terfenol-D layer(s) under dc bias and subsequently enhanced ME effects in the composite at lower H_{dc}.

Figure 2(a) shows the flux as a function of position (along Terfenol-D's length) in a single Terfenol-D layer for longitudinal magnetization mode, and Fig. 2(b) shows the flux of a single Terfenol-D layer for transverse magnetization mode, both with and without attached $MnZnFe_2O_4$. The results show by attaching high-μ $MnZnFe_2O_4$ layers that magnetic flux in a Terfenol-D layer is increased by a factor of 2–3 times for longitudinally magnetized mode and 6–8 times for transverse magnetization mode.

Enhancements in magnetic flux will further result in much higher effective piezomagnetic coefficients of a single Terfenol-D layer. Figure 3(a) and 3(b) show the effective piezomagnetic coefficients ($d_{33,m}=\delta\varepsilon_{33}/\delta H$ and $d_{31,m}=\delta\varepsilon_{31}/\delta H$) of a single Terfenol-D layer with/without soft-ferrite $MnZnFe_2O_4$ as a function of H for longitudinal and transverse magnetization modes. Correspondingly, the effective longitudinal piezomagnetic coefficients, $d_{33,m}$, were enhanced by a factor of $\sim$3 times. Note that in small dc magnetic bias range of $H_{dc}<100$ Oe, this enhancement is about eight times. In the transverse magnetization mode, the strong magnetic flux concentration will cause a rapid magnetic saturation in thickness direction of Terfenol-D layer, resulting in a dramatic enhancement in transverse piezomagnetic coefficient, $d_{31,m}$, by a factor of $\sim$10 times. Furthermore, this enhancement is as high as 30 times under $H_{dc}<150$ Oe. We will see that the enhancements in effective piezomagnetic coefficients will further result in the enhanced ME effects.

III. THEORETICAL ANALYSIS

Two three-phase ME laminate configurations were constructed and tested: (i) a longitudinal one shown in Fig. 1(a), consisting of high-permeability $MnZnFe_2O_4$ layers attached at both ends of a Terfenol-D/PZT three-layer laminate by silicon rubber, and (ii) a transverse one shown in Fig. 1(b), consisting of two $MnZnFe_2O_4$ layers attached on both sides. The Terfenol-D/PZT three-layer laminate is a bimorph type,[15] where the two piezoelectric layers are poled in reverse thickness directions. In the longitudinal magnetization mode, the high-μ $MnZnFe_2O_4$ layers concentrate external flux along the long axis of the laminate, which, as we will subsequently show, decreases the required H_{dc}. In the transverse mode, the high-μ $MnZnFe_2O_4$ layers attached at the sides of the laminate concentrate flux that is incident along the thickness direction decreasing $H_{dc,opti}$ but attenuates flux parallel to the long axis.

Under an applied ac magnetic field H_{ac}, the magnetostrictive layer in Terfenol-D/PZT laminates strain along the longitudinal axis, exciting a bending motion mode due to an unsymmetric strain about the longitudinal neutral face (line). Since the $MnZnFe_2O_4$ layers are softly attached to the laminate, we assume that they do not affect the bending motion.

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Using piezoelectric and piezomagnetic constitutive equations within a small bending elastic theory, the ME field coefficient under a low-frequency (quasistatic) drive can be given as

$$\frac{dE}{dH} \approx \frac{d_{3i,m} g_{31,p} t_m (t_m - 2t_n)(t_p + 2t_m - 2t_n)}{4 s_{33}^H (1 - \nu_p) D_{\text{comp}} / Y_p} \quad (i = 1 \text{ or } 3), \tag{1}$$

where $d_{33,m}$ (or $d_{31,m}$) is the longitudinal (or transverse) piezomagnetic coefficient for the longitudinal (or transverse) magnetization mode(s), $g_{31,p}$ the transverse piezoelectric-voltage coefficients, t_m the thickness of the magnetostrictive layer, t_p the thickness of the piezoelectric layers, t_n the neutral-face position of the Terfenol-D/PZT composite, s_{33}^H the elastic compliances of the magnetostrictive layer, Y_p and ν_p Young's modulus and Poisson's ratio of the piezoelectric layers, and D_{comp} the composite stiffness of the laminate. Composite's stiffness is a resultant of that of the constituent layers given as

$$D_{\text{comp}} = D_m + D_p - t_m (t_m - 2t_n)^2 k_{33,m}^2 / 4 s_{33}^B, \tag{2}$$

where $k_{33,m}$ is the magnetoelastic coupling coefficient, and D_m and D_p the bending stiffness of the magnetostrictive and piezoelectric layers, respectively. From (1), we can see that dE/dH is proportional to (i) $d_{33,m}$ or $d_{31,m}$ ($\delta\varepsilon_{33}/\delta H$ or $\delta\varepsilon_{31}/\delta H$) of the magnetostrictive layer(s) and (ii) $g_{31,p}$ of the piezoelectric layer(s). The effect of flux concentration by incorporation of high-μ $MnZnFe_2O_4$ in ME composite is to enhance the effective piezomagnetic coefficient $d_{33,m}$ or $d_{31,m}$ and consequently, also enhancing dE/dH.

IV. RESULTS AND DISCUSSION

Rectangular-shaped (30 mm length, 15.0 mm width, and 6.0 mm thickness) $MnZnFe_2O_4$ layers with a $\mu_r > 5000$ were colaminated with a Terfenol-D/PZT-bimorph three-layer laminate (20 mm length, 6 mm width, and 3 mm thickness): in both longitudinal *L*- and transverse *T*-magnetization modes, as shown in Figs. 1(a) and 1(b), respectively. The voltages induced across the PZT layers were measured as a function of H_{dc} in response to a constant H_{ac} of 1 Oe, via a lock-in amplifier. An electromagnet was used to apply H_{dc}, and a pair of Helmholtz coils was used to generate a small H_{ac}.

Figure 4 shows dE/dH as a function of H_{dc}. Data are shown for laminates with and without attached $MnZnFe_2O_4$ layers. The results clearly show a much higher dE/dH for Terfenol-D/PZT laminates with attached soft-ferrite layers, in particular, at lower biases of $H_{dc} \leqslant 50$ Oe. In the case of $MnZnFe_2O_4$ layers attached in the *L*-magnetization mode, part (a) of this figure shows a maximum value of $|dE/dH| = \sim 3$ V/cm Oe under $H_{dc} = 56$ Oe, whereas without attached $MnZnFe_2O_4$ layers, the maximum value was $|dE/dH| = \sim 1$ V/cm Oe under a much larger bias of $H_{dc} = 270$ Oe. The former maximum is approximately three times higher than the latter, because the effective piezomagnetic coefficient $d_{33,m}$ in the former is again approximately three times higher than that in the latter. Note, however, that under the same low magnetic biases of $H_{dc} < 50$ Oe, $|dE/dH|$ was about eight times higher with $MnZnFe_2O_4$ attached, than without, as predicted in Fig. 3(a). In the *T*-magnetization mode, a more dramatic enhancement of $|dE/dH|$ was observed upon attachment of $MnZnFe_2O_4$ layers, as shown in part (b) of the figure. The value of $|dE/dH|$ was about 28 times higher with $MnZnFe_2O_4$ layers attached than without, again due to corresponding increases in $d_{33,m}$ by a factor of ~30 times under $H_{dc} < 150$ Oe, as shown in Fig. 3(b).

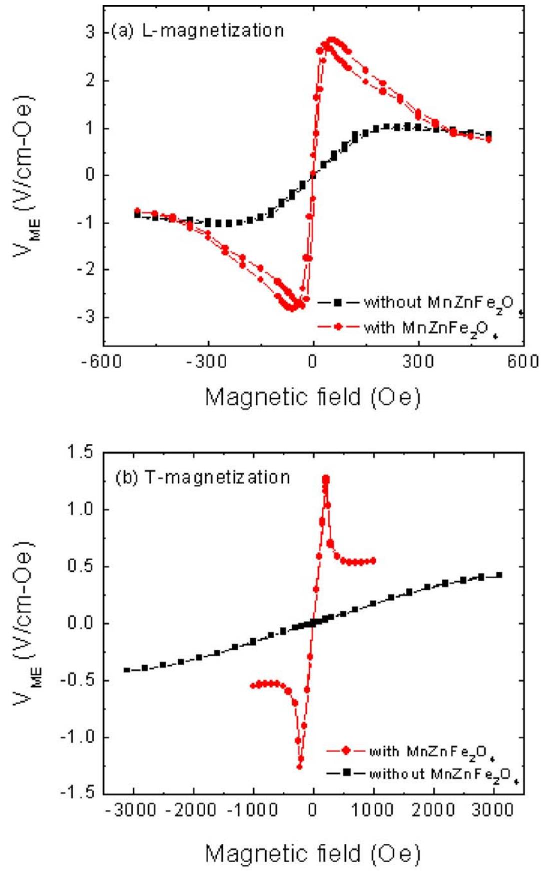

FIG. 4. (Color online) Measured ME voltages as a function of dc magnetic bias H_{dc} at $H_{ac} = 1$ Oe for $MnZnFe_2O_4$/Terfenol-D/PZT composite and a normal Terfenol-D/PZT bimorph: (a) Longitudinal magnetization mode and (b) transverse magnetization mode.

V. SUMMARY

In summary, high-μ $MnZnFe_2O_4$ layers have been incorporated into Terfenol-D/PZT laminates, producing a strong magnetic flux concentration. This, in turn, results in a larger effective piezomagnetic coefficient in the Terfenol-D layer and consequently a proportionally enhanced value of the ME field coefficient. Upon colaminating Terfenol-D/PZT with $MnZnFe_2O_4$ layers, we find enhancement in $|dE/dH|$ of between 8 and 28 times under moderate magnetic biases.

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