

## High magnetic field sensitivity in Pb ( Zr , Ti ) O 3 – Pb ( Mg 1 / 3 Nb 2 / 3 ) O 3 single crystal/Terfenol-D/Metglas magnetolectric laminate composites

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# High magnetic field sensitivity in $\text{Pb}(\text{Zr}, \text{Ti})\text{O}_3\text{-Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$ single crystal/Terfenol-D/Metglas magnetolectric laminate composites

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We report the magnetic field sensitivity results on five layer structure given as Metglas/Terfenol-D/PMN-PZT/Terfenol-D/Metglas, where PMN and PZT correspond to  $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3$  and  $\text{Pb}(\text{Zr}, \text{Ti})\text{O}_3$ , respectively. The piezoelectric constant ( $d_{33}$ ) of poled PMN-PZT was found to be 1600 pC/N with dielectric constant of 5380 at 1 kHz. The sensitivity measurements were conducted after attaching individual layers in the laminate clearly delineating the effect occurring in the response. The magnetolectric response for this five layer structure at 1 kHz was found to be 5 V/cm Oe at dc bias field of 1000 Oe under an ac drive of 1 Oe. At 1 kHz frequency, the sensor was able to deterministically measure step changes of 500 nT while at 10 Hz we can clearly identify the sensitivity of 1  $\mu\text{T}$ . These results are very promising for the cheap room-temperature magnetic field sensing technology. © 2010 American Institute of Physics. [doi:10.1063/1.3406142]

Dielectric polarization of a material under a magnetic field or induced magnetization under an electric field requires the simultaneous presence of long-range ordering of magnetic moments and electric dipoles. Investigations have revealed the presence of both ferroelectricity and magnetism in number of materials such as perovskite type  $\text{BiFeO}_3$ ,  $\text{BiMnO}_3$ , the boracite family,  $\text{BaMF}_4$  compounds (M, divalent transition metal ions), hexagonal  $\text{RMnO}_3$  (R, rare earths), and the rare earth molybdates but none seem to provide large coupling between them. Recently, rare earth manganates such as  $\text{TbMnO}_3$ ,  $\text{DyMnO}_3$ , and  $\text{TbMn}_2\text{O}_5$  have been reported to exhibit reproducible electric polarization under magnetic fields, however, the magnitude of the magnetolectric (ME) coefficient (unit of volt per centimeter per oersted) is quite small.<sup>1-3</sup> Single phase materials suffer from the drawback that the ME effect is considerably weak even at low temperatures. Better alternatives to single phase materials are ME composites. The composites exploit the product property of the materials (the ME effect is not present in individual phases).

In our early work, we reported the results on ME laminate composites (MLCs) made from the giant magnetostrictive material, Terfenol-D, and relaxor-based piezocrystals  $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-PbTiO}_3$  (PMN-PT).<sup>4</sup> ME behavior in laminate composites has now been reported for various material combinations including  $\text{Pb}(\text{Zr}, \text{Ti})\text{O}_3$  (PZT) or  $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3\text{-PbTiO}_3$  (PMN-PT) layers laminated with magnetostrictive  $\text{Tb}_{1-x}\text{Dy}_x\text{Fe}_{2-y}$ , Permendur,  $\text{Ni}_{1-x}\text{Co}_x\text{Fe}_2\text{O}_4$ , or  $\text{Co}_{1-x}\text{Zn}_x\text{Fe}_2\text{O}_4$ .<sup>5-7</sup> Both epoxy-glued and cofired laminates have been reported in literature. The epoxy-glued laminates are generally fabricated by sandwiching and bonding piezoelectric plate/disk/fibers between two layers of magnetostrictive plates/disks/foils. The best sensitivity data on epoxy-glued composites has been reported by

Dong *et al.*, where piezofiber was laminated between the high-permeability magnetostrictive FeBSiC alloy (Metglas) resulting in a large response of 22 V/cm Oe at 1 Hz.<sup>8</sup> The lower operating frequency is result of the foil-like structure which produces bending modes.

The design of epoxy-glued laminate composites relies on maximizing the elastic coupling through the selection of high magnetostrictive constant material and high piezoelectric voltage constant ( $g$ ) ceramic. Srinivasan *et al.*<sup>9</sup> have derived the transverse ME coefficient for the layered composite as following:

$$\frac{\delta E_3}{\delta H_1} = \frac{-2d_{31}^p q_{11}^m v^m}{(s_{11}^m + s_{12}^m)\epsilon_{33}^{T,P} v^p + (s_{11}^p + s_{12}^p)\epsilon_{33}^{T,P} v^m - 2(d_{31}^p)^2 v^m}, \quad (1)$$

where  $d_{31}^p$  is the piezoelectric coefficient,  $v^m$  and  $v^p$  are the volume of magnetic and piezoelectric phase,  $s_{11}^p, s_{12}^p$  are the elastic compliances for piezoelectric phase,  $s_{11}^m, s_{12}^m$  are the elastic compliances for magnetostrictive phase,  $q_{11}^m$  is the piezomagnetic coefficient of the magnetic phase, and  $\epsilon_{33}^{T,P}$  is the permittivity of the piezoelectric phase. This equation indicates that high piezoelectric strain constant and piezomagnetic constant with matching elastic compliances will lead to higher magnitude of ME coefficient. However, results on piezofiber laminated with Metglas have shown that permeability and flexibility in the structure is important for achieving the giant ME coefficient and reducing the operating frequency.

Table I shows the magnitude of relevant magnetostrictive material parameters involved in design of magnetic field sensor based on ME effect. It can be seen from this table that Terfenol-D has giant magnitude of magnetostriction but low permeability while Metglas has giant magnitude of permeability but low magnetostriction. Thus, it is natural to conceive of a material which can provide both high magnetostriction and permeability. We adopted the path of 2-2 composite with alternate layer of Terfenol-D and Metglas to

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TABLE I. List of relevant magnetostrictive material parameters.

Material	Magnetostriction (ppm)	Relative permeability	Electrical resistivity ( $\mu\Omega$ cm)
Terfenol-D	800–1200	3–10	58
Metglas 2605SA1	27	45 000 (as cast) 600 000 (annealed)	130

combine high permeability and magnetostriction. In a symmetrical laminate, one would bond radially magnetized Terfenol-D on either side of piezoelectric layer to achieve radial mode deformation.

Piezoelectric single crystal with the composition  $\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})_y(\text{Zr}_x\text{Ti}_{1-x})_{1-y}\text{O}_3$  (PMN–PZT) (commercially available from CeraComp, Korea) was selected as the piezoelectric phase. The single crystal (X-tal) had the square shape with dimension of  $10 \times 10 \text{ mm}^2$  and thickness of 1 mm. Electroding was done by depositing gold electrodes on either side. The piezoelectric constant ( $d_{33}$ ) of poled PMN–PZT was found to be  $1600 \text{ pC/N}$  with dielectric constant of 5380 at 1 kHz. Thus, the piezoelectric voltage constant ( $g_{33}$ ) was

of the order of  $33.5 \times 10^{-3} \text{ Vm/N}$ . The laminate composite was fabricated in sequential steps as shown in Figs. 1(a)–1(c), where after each step we measured the ME response to quantify the effect of each layer.

In the first step, Terfenol-D with diameter of 13 mm and thickness of 2 mm was attached on one side of PMN–PZT

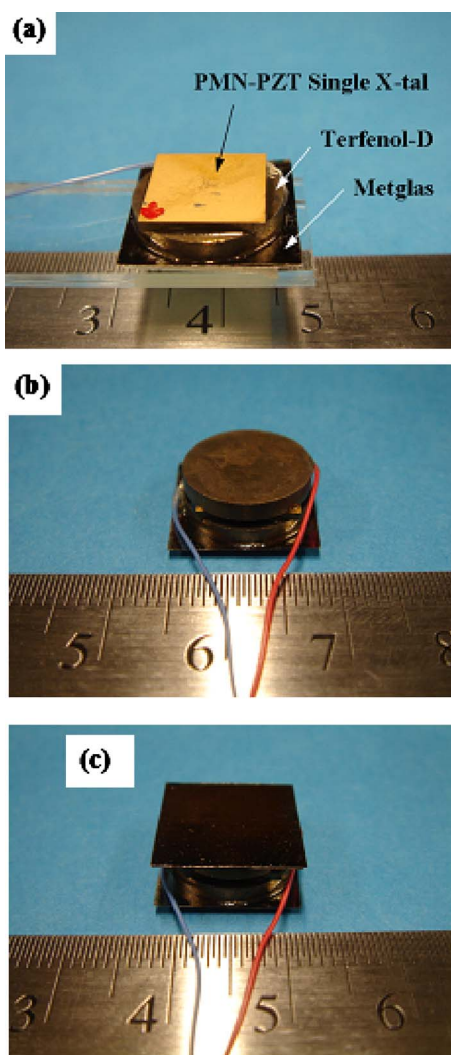


FIG. 1. (Color online) (a) PMN–PZT single crystal/Terfenol-D/Metglas laminate, (b) Terfenol-D/PMN–PZT single crystal/Terfenol-D/Metglas laminate, and (c) Metglas/Terfenol-D/PMN–PZT single crystal/Terfenol-D/Metglas laminate.

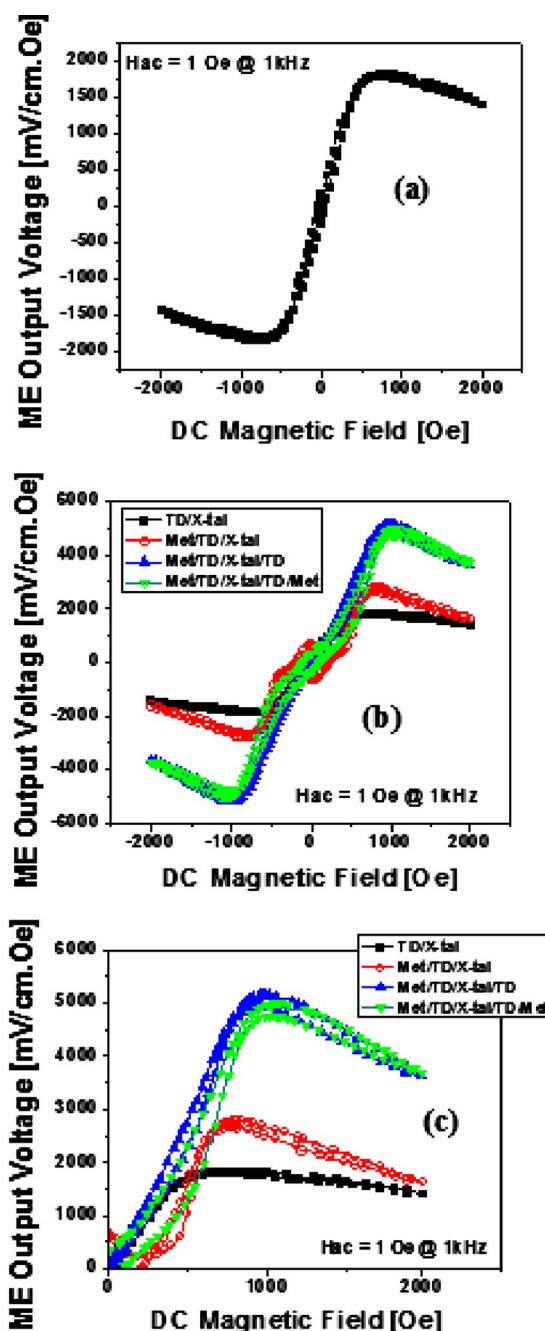


FIG. 2. (Color online) ME properties depending on attaching layers: (a) PMN–PZT single crystal/Terfenol-D, [(b) and (c)] PMN–PZT single crystal/Terfenol-D/Metglas laminate.

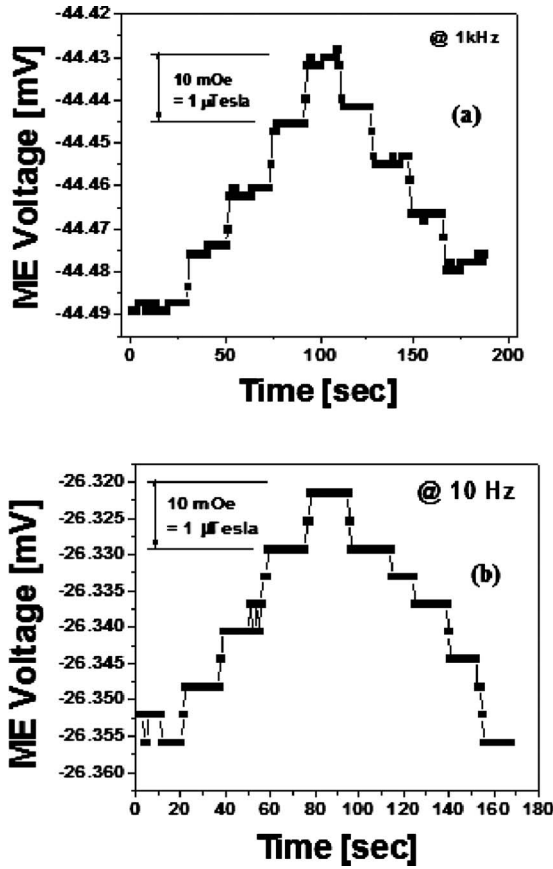


FIG. 3. Sensitivity limit of small dc magnetic field for the PMN-PZT single crystal/Terfenol-D/Metglas laminate under a constant  $H_{ac}=1$  Oe: (a) at  $f=1$  kHz and (b)  $f=10$  Hz.

single X-tal using epoxy (West System, USA) with curing temperature of 80 °C. Figure 2(a) shows the ME response of this two-layer laminate exhibiting high value of 1.8 V/cm Oe at 710 Oe dc bias magnetic field. The peak position is reflective of the maximum slope in  $\partial\lambda/\partial H$ , where  $\lambda$  is magnetostriction and  $H$  is static magnetic field. The shape of the curve in Fig. 2(a) follows the variation in  $\partial\lambda/\partial H$ , vanishing at low and high magnetic fields where  $\lambda$  disappears or goes to saturation, respectively. ME measurements were done in L-T mode configuration using an electromagnet. The samples were placed in the center of the Helmholtz coil under an ac magnetic field ( $H_{ac}$ ) which was located at the center of electromagnet. The voltage induced on the laminate was monitored using a lock-in amplifier.

In the second step, four layers of Metglas (2605SA1, Metglas Inc., USA) sheets with a total thickness of 100  $\mu\text{m}$  and area of  $15 \times 15 \text{ mm}^2$  were attached on top of the Terfenol-D/PMN-PZT laminate as shown in Fig. 1(a). The number of Metglas layers were selected based on previous studies conducted for PZT-Metglas laminates.<sup>10</sup> Figures 2(b) and 2(c) shows the effect of Metglas layers on the ME response which increased to 2.78 V/cm Oe. The peak position of ME coefficient also shifted higher to 812 Oe. At low magnitude of dc bias, this structure shows hysteretic behavior similar in nature to that observed for Fe-PZT-Fe structure by Laletin *et al.*<sup>11</sup> We expect that this hysteretic behavior is related to the interaction between Terfenol-D and Metglas

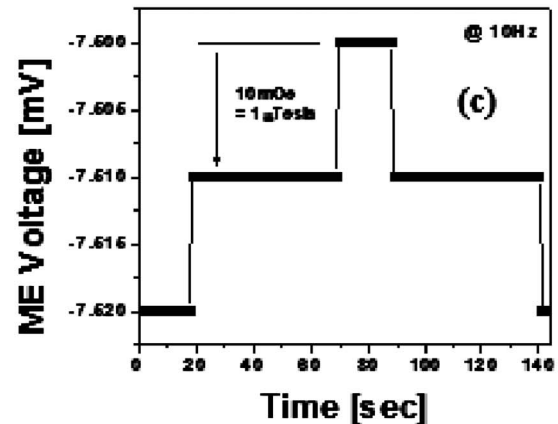
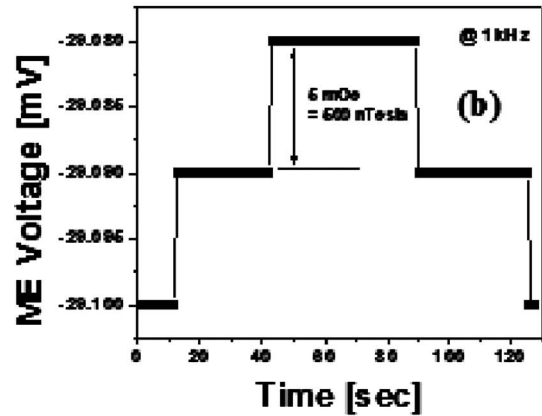
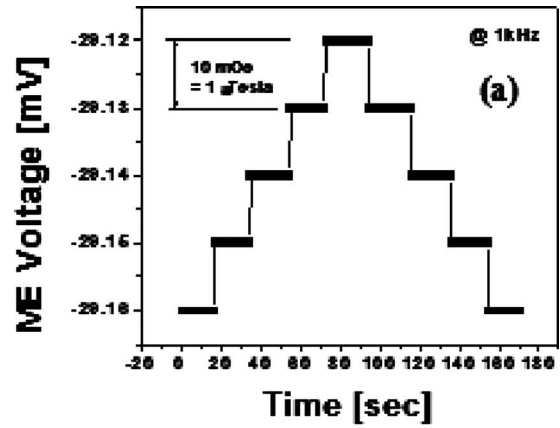


FIG. 4. Sensitivity limit of small dc magnetic field for the Metglas/Terfenol-D/PMN-PZT single crystal/Terfenol-D/Metglas laminate under a constant  $H_{ac}=1$  Oe: (a) and (b) at  $f=1$  kHz, and (c) at  $f=10$  Hz.

mainly due to the presence of ferromagnetic metal Fe. The shift in the ME peak position is indicative of this interaction resulting from the change in the slope of magnetostriction and increase in the amplitude of the ME coefficient indicates change in the magnitude of effective piezomagnetic coefficient. If we neglect the contributions from the exchange field and interface effects, the biaxial stress due to the Metglas layer exerted onto the Terfenol-D can be calculated to be:

$$\Delta\sigma = \frac{-2E_{\text{Metglas}}E_{\text{Terfenol-D}}t_{\text{Metglas}}\Delta\epsilon}{(1-\nu)(2E_{\text{Metglas}}t_{\text{Metglas}} + E_{\text{Terfenol-D}}t_{\text{Terfenol-D}})}$$

where  $E$  is the elastic modulus,  $\Delta\epsilon$  is the strain,  $t$  is the thickness, and  $\nu=0.3$  is the Poisson's ratio. Taking the values

for tensile modulus as  $E_{\text{Metglas}}=100$  GPa,  $E_{\text{Terfenol}}=25$  GPa,  $t_{\text{Terfenol}}=2$  mm,  $t_{\text{Metglas}}=100$   $\mu\text{m}$ , and  $\Delta\varepsilon=27$  ppm, the magnitude of biaxial stress can be found to be  $5.51 \times 10^6$  N/m<sup>2</sup>. This additional stress can induce a mechanical strain of about 220.4 ppm in Terfenol-D, resulting in increase in effective piezomagnetic coefficient of Terfenol-D and contributing to the ME coefficient as:

$$\frac{\delta E_3}{\delta H_1} = \left( \frac{q_{11}^{m,\text{eff}}}{q_{11}^m} \right) \left( \frac{v^{m,\text{eff}}}{v^m} \right). \quad (2)$$

In the third step, we attached Terfenol-D on other side of PMN–PZT layer as shown in Fig. 1(b). The effect of this layer on ME response is shown in Figs. 2(b) and 2(c) indicating that ME coefficient increases to 5.15 V/cm Oe at dc bias field of 1011 Oe. This is approximately 78% increase in the ME coefficient compared to the second step. By summing the magnitude of ME coefficient in the first and second step, one can arrive at the value of 4.58 mV/cm Oe which is smaller than that obtained in the third step. Thus, this asymmetric structure results in effective stress transfer on the piezoelectric layer. The low field hysteretic behavior observed in earlier step was found to disappear which could be explained based on cancellation of any fringing fields due to ferromagnetic Fe. In the last step, four layers of Metglas were attached on the Terfenol-D to achieve the symmetrical structure Metglas/Terfenol-D/PMN–PZT/Terfenol-D/Metglas as shown in Fig. 1(c). The ME response decreased slightly to 4.97 V/cm Oe at dc bias field of 1000 Oe as shown in Fig. 2(b). Further, the hysteresis between the forward and backward drive can be again seen for the symmetrical structure as shown in Fig. 2(c). We expect that this slight decrease in ME coefficient is related to the out of phase stress contribution from the two opposing Metglas layers. Overall, these results are very exciting showing a giant ME response at low frequency of 1 kHz.

Next, we quantified the magnetic field sensitivity of these laminate structures. Figures 3(a) and 3(b) shows the variation in output voltage for a step-change in dc magnetic field at specific frequency. The three layer structure was found to have sensitivity of 1  $\mu\text{T}$  at 1 kHz. At 10 Hz, we can identify the step changes in 1  $\mu\text{T}$  but there is drift in the signal indicating the limit of this topology. Figures 4(a)–4(c) shows the magnetic field sensitivity of five layer structure at various frequencies. At 1 kHz frequency, the sensor was able to deterministically measure step changes of 500 nT as

shown in Fig. 4(b) while at 10 Hz we can clearly identify the sensitivity of 1  $\mu\text{T}$ . This improvement is correlated with the increase in ME coefficient but we think that the effective permeability of the system also plays an important role. Hashin and Shtrikman<sup>12</sup> have shown that for a multiphase material in an embedded configuration the effective permeability is given as:

$$\mu_{\text{eff}} = \mu_{\text{Terfenol}} + \frac{v^{\text{Metglas}}}{\frac{1}{\mu_{\text{Metglas}} - \mu_{\text{Terfenol}}} + \frac{v^{\text{Terfenol}}}{3\mu_{\text{Terfenol}}}}. \quad (3)$$

Using the  $\mu_{\text{Terfenol}}=5$ , and  $\mu_{\text{Metglas}}=45000$ , it can be shown that  $\mu_{\text{eff}}=6.268$  for the two layer laminate of Metglas and Terfenol-D as shown in Fig. 1(a). This approximate calculation shows that there is an increase in about 25% in the magnitude of permeability. These results are highly encouraging and show that this simple technology can provide effective room temperature magnetic field sensors.

In summary, we describe the fabrication of MLC using the highest piezoelectric constant, highest magnetostriction, and highest permeability materials. The results show that this sensor can distinguish small dc magnetic fields of 500 nT at 1 kHz under applied ac field of 1 Oe.

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