Enhancing the magnetoelectric response of Metglas/polyvinylidene fluoride laminates by exploiting the flux concentration effect

Z. Fang,[1,2] S. G. Lu,[2] F. Li,[1] S. Datta,[1] Q. M. Zhang,[1,2,a)] and M. El Tahchi[3]
[1]*Department of Electrical Engineering, The Pennsylvania State University, University Park, Pennsylvania 16802, USA*
[2]*Materials Research Institute, The Pennsylvania State University, University Park, Pennsylvania 16802, USA*
[3]*LPA-GBMI, Faculty of Sciences II, Department of Physics, Lebanese University, P.O. Box 90656, Jdeidet, Lebanon*

(Received 30 June 2009; accepted 27 August 2009; published online 17 September 2009)

Magnetic flux concentration effect of Metglas as a function of its sheet aspect ratio was investigated for Metglas/polyvinylidene fluoride laminates. Taking advantage of this effect, the magnetoelectric voltage coefficient of 21.46 V/cm·Oe for a laminate with 1 mm wide and 30 mm long Metglas sheet (25 μm thick) is achieved, which is much higher than those reported earlier in similar laminates without making use of the flux concentration effect. The results demonstrate an effective means to significantly enhance the sensitivity of magnetostrictive/piezoelectric composite laminates as weak magnetic field sensors. © *2009 American Institute of Physics*. [doi:10.1063/1.3231614]

The magnetoelectric (ME) effect[1–5] is the appearance of an electrical signal upon applying a magnetic field $\boldsymbol{H}$ and/or the appearance of a magnetic signal upon applying an electric field $\boldsymbol{E}$. Although the ME effect was first observed in single phase materials[2] (e.g., Cr_2O_3), the composite laminates of the magnetostrictive layer (e.g., Terfenol-D and Metglas) and the piezoelectric layer [e.g., $Pb(ZrTi)O_3$ (PZT) and polyvinylidene fluoride (PVDF)] have attracted much attention due to the strong coupling effect between the magnetostrictive and the piezoelectric layers, which derive large ME coupling coefficients at room temperature and have potential to be used as high sensitivity magnetic sensors, electrical current sensors, and other devices.[4–10] In the ME laminates which exhibit large ME coupling coefficients, the ones with Metglas are attractive due to their low saturation magnetization field and, consequently, a relatively low dc bias magnetic field (<20 Oe), which is highly desirable for high sensitivity magnetic sensors. Indeed, a sensitivity of 2×10^{-11} T/Hz$^{1/2}$ at 1 Hz has been reported.[11]

For magnetic materials with very high permeability such as Metglas ($\mu_r > 45\,000$), the magnetic flux concentration effect can be quite significant.[12,13] Finite element simulation using COMSOL MULTIPHYSICS®, a three-dimensional finite element analysis and solver software package,[14] was carried out for Metglas sheets with dimensions similar to those used in the experiments in this letter. Illustrated in Fig. 1(a) is a two-dimensional simulation result of the flux concentration effect for a Metglas ($\mu_r = 45\,000$ is used in the simulation and the Metglas is infinitely extended in perpendicular to the *x*-*y* plane) sheet with a width (along the *y*-direction)/length (along the *x*-direction) ratio of 0.5 in free space. As can be seen, the flux density inside the Metglas is much higher than that in free space. As will be shown in this letter, this effect can be exploited to markedly enhance the ME coefficient of Metglas-based ME laminates and thus to improve the sensitivity of ME laminate-based magnetic sensors.

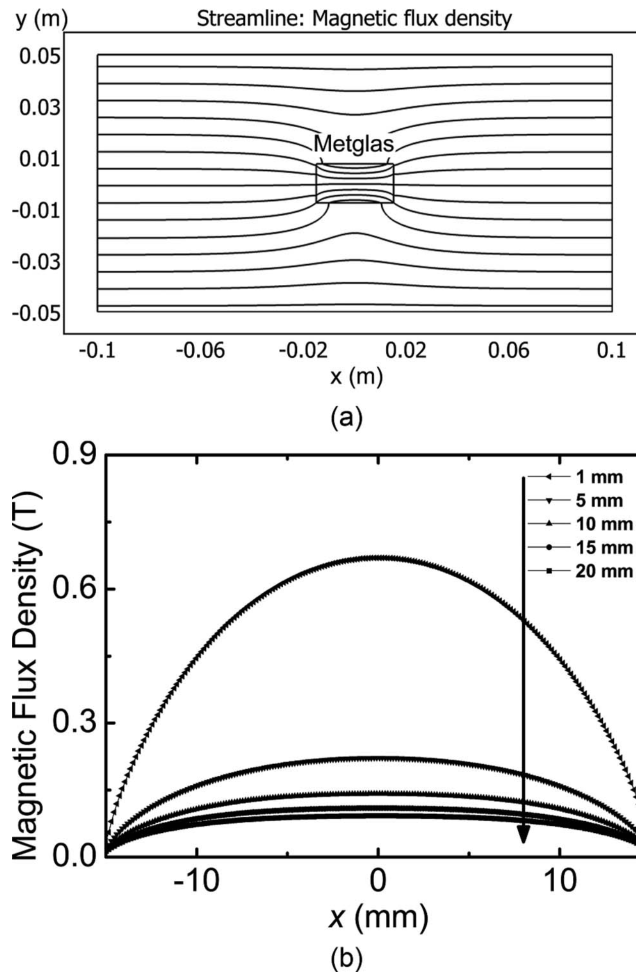

FIG. 1. (a) Magnetic flux density distribution in Metglas under a uniform external magnetic field in free space. (b) Magnetic flux density of the Metglas sheet along the *x*-axis (length direction) at $y=0$, $z=0$ (the origin is at the center of the Metglas sheet) with various aspect ratios of W_m/L_m. (The length and thickness are fixed at 30 mm and 25 μm respectively, and the width W_m is varied from 20 to 1 mm.) The external magnetic field is 1 Oe (10^{-4} T).

a)Electronic mail: qxz1@psu.edu.

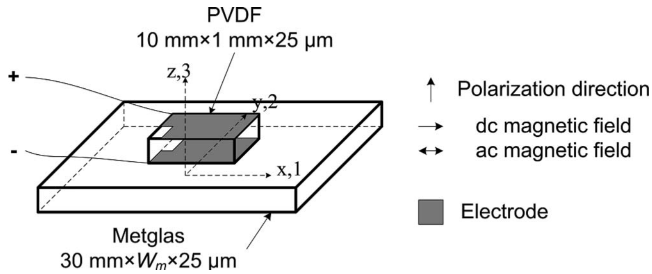

FIG. 2. Schematic diagram of Metglas/PVDF composite laminates.

TABLE I. Metglas/PVDF ME laminates dimensions for data in Fig. 3.

Device No.	MP0	MP1–4
Metglas size (mm)	30×15	30×15
PVDF size (mm)	30×15	10×5

Figure 2 presents a schematic diagram of the ME laminate, which consists of a piezoelectric layer with thickness t_p bonded to a magnetostrictive layer with thickness t_m. Iron-based magnetic alloy Metglas 2605SA1 (Metglas, Inc., SC, μ_r=45 000) of t_m=25 μm was used as the magnetostrictive layer and PVDF piezoelectric polymer (Ktech Corp., NM) of t_p=25 μm was used as the piezoelectric layer. Two layers were bonded together using nonconductive epoxy resin (5 min Epoxy, ITW Devcon, MA). To probe the magnetic flux concentration effect on the ME coupling coefficients of the laminates, the length L_m of the Metglas sheet is fixed at 30 mm, while the width W_m is varied from 20 down to 1 mm. As shown in Fig. 1, the magnetic flux density is the highest at the central region of the Metglas sheet, where the PVDF layer with width of W_p=1 mm and length of L_p=10 mm is bonded to realize the highest ME coupling.

Presented in Fig. 1(b) are the simulation results of the flux density for Metglas with 25 μm thick, 30 mm long, and with width W_m varying from 20 to 1 mm (Fig. 2) under an external magnetic field of 1 Oe (10^{-4} T) in free space. The data are presented for the flux density along the length direction (x-direction) at y=0, z=0 [see Figs. 1(a) and 2 for the coordination system]. Figure 1(b) shows that as the aspect ratio (W_m/L_m) decreases, the flux density at the central region of the Metglas sheet (x=0) increases markedly. $B(x=0)$ for W_m=1 mm is about seven times higher than that for W_m=20 mm. The simulated results shown here are consistent with the measurement on a steel bar magnet and an alnico bar magnet.[15] The variation in magnetic flux density at the center of Metglas with the aspect ratio is also concordant with the calculated results.[16]

When the piezoelectric layer is bonded to different sites of a Metglas sheet, the variation in B in Metglas sheet can cause the variation in the ME voltage coefficient $\alpha_{ME}=\Delta E/\Delta H$, where E is the electrical field generated in the piezoelectric layer and H is the externally applied magnetic field. As shown in Fig. 3, the measured α_{ME} is the largest for MP4 which is placed at the center of a Metglas sheet (see the inset in Fig. 3). When the PVDF layer is placed at other regions of the Metglas (MP1–3), α_{ME} is reduced. For comparison, α_{ME} for a Metglas/PVDF laminate in which the PVDF layer covers the whole Metglas sheet (MP0) is also characterized. Table I summarizes the dimensions of the Metglas sheets and PVDF layers. The results show that α_{ME} is the highest for a laminate with the PVDF layer bonded at the center region of the Metglas sheet.

By bonding a PVDF layer with L_p=10 mm and W_p=1 mm at the center region of the Metglas sheet with L_m=30 mm and varying W_m, we investigate the enhancement of the flux concentration effect on the ME response. Figure 4 presents the experimental results of α_{ME} for the Metglas/PVDF laminates with varying W_m from 20 to 1 mm. As can be seen, α_{ME} increases with decreasing W_m, while peaking at a lower dc bias field. Both the phenomena are a result of an increased magnetic flux concentration in Metglas and, hence, the ratio of α_{ME} (W_m=1 mm)/α_{ME}(W_m=20 mm) $\sim$2 is the same as the ratio of H_{peak} (W_m=20 mm)/H_{peak} (W_m=1 mm).

Due to the elastic coupling between the Metglas and PVDF which causes the ME effect, α_{ME} of the Metglas/PVDF laminates investigated in this letter can be expressed as

$$\alpha_{ME} = \frac{\Delta E}{\Delta H_a} = \frac{\Delta E}{\Delta S}\frac{\Delta S}{\Delta H_a}, \tag{1}$$

where E is the induced electric field across the PVDF layer, S is the magnetostrictive strain caused by the application of magnetic field, and H_a is the external ac magnetic field. In Eq. (1), $\Delta E/\Delta S$ is a constant of the PVDF (piezoelectric coefficient). While

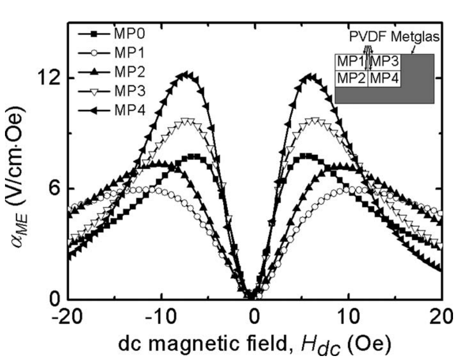

FIG. 3. α_{ME} as a function of dc bias magnetic field for Metglas/PVDF laminates with PVDF bonded on different regions of the Metglas sheet. The data were measured at 20 Hz and H_{ac}=0.38 Oe.

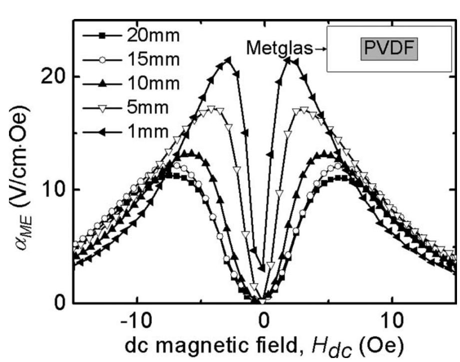

FIG. 4. α_{ME} as a function of dc bias magnetic field for Metglas/PVDF laminates with varying W_m. The data were measured at 20 Hz and H_{ac}=0.38 Oe.

$$\frac{\Delta S}{\Delta H_a}=\frac{\Delta S}{\Delta B_M}\frac{\Delta B_M}{\Delta H_a}, \tag{2}$$

where B_M is the magnetic flux density. For a magnetostrictive alloy such as Metglas, $\Delta S/\Delta B_M$ does not change much with the sample dimensions studied here. Moreover the Young's modulus of Metglas (100–110 GPa) is much higher than that of PVDF (3 GPa), so $\Delta S/\Delta B_M$ will not change as the aspect ratio is reduced. On the other hand, $\Delta B_M/\Delta H_a$ will increase as the aspect ratio is decreased due to the flux concentration effect (Fig. 1),[17,18] which will result in a marked increase in α_{ME}.

For a magnetic material under an external magnetic field H_a in free space, the magnetic flux density B_M is[19]

$$B_M=\mu_r\mu_0(H_a-H_d), \tag{3}$$

where H_d is the demagnetization field.

Since $\mu_r=45\,000$, approximately we have[15]

$$\frac{B_M}{\mu_0 H_a}=\frac{1}{N_d}, \tag{4}$$

where N_d is the demagnetizing factor. The value of N_d depends mainly on the aspect ratio of the magnetic material. For Metglas with $W_m \gg L_m$, H_d is nearly equal to H_a. Consequently, B_M inside the Metglas is nearly the same as that in the free space ($B_M=\mu_0 H_a$). On the other hand, as the aspect ratio approaches zero, H_d at the central region of the Metglas sheet will approach zero. As a result, $B_M=\mu_r\mu_0 H_a$ is 4.5 T at $x=0$ in Fig. 2. Figure 1(b) shows that for the Metglas sheet with $W_m=1$ mm, B_M at $x=0$ is about 0.7 T, almost seven times larger than that for $W_m=20$ mm. The results here indicate the potential of improving α_{ME} further in Metglas/PVDF laminates by further reducing the aspect ratio.

Comparing the experimental results in Fig. 4 with the simulation results of Fig. 1(b) reveals that although there is an increase in α_{ME} with a decreased aspect ratio of the Metglas sheet, the experimental increase is much smaller than that of the simulation. This can be partly caused by the occurrence of other piezoelectric vibration modes, such as the bending mode, which becomes severe for laminates with very narrow Metglas sheet and will greatly reduce the ME coupling coefficient. An improved design such as using double sided PVDF sandwiched Metglas may reduce this effect.[11] Another possible reason is that in the experiment, the Metglas/PVDF laminates are not in the ideal free space, but instead are situated in two magnets with a gap of 50 mm. This reduces the magnetic flux concentration effect, compared with that in free space.

It should be noted that although in the experiments investigated here, L_m is fixed at 30 mm for the convenience of the experiment, the aspect ratio can be reduced by reducing both L_m and W_m. These small sized Metglas/PVDF laminates as magnetic sensors can be used in miniature magnetic sensors and by integrating them directly with microelectronic circuit, as has been done with the integrated pyroelectric sensors, and various spurious noises and stray capacitance due to the electrical leads can be also reduced markedly.[20] Furthermore by using other piezoelectric materials such as PZT fiber to increase the piezoelectric voltage constant, the sensitivity can be further improved.[21] These considerations suggest the potential of the Metglas (or magnetostrictive alloy with high μ_r)/piezoelectric material laminates in achieving very high sensitivity for weak magnetic field sensing.

In summary, we show that very high permeability of the Metglas can be exploited to significantly increase the magnetic flux density in the Metglas sheet with very small aspect ratio. Consequently, the magnetostriction and α_{ME} of the Metglas/PVDF ME composite laminate with very small aspect ratio of Metglas sheet can be enhanced remarkably. The results here show the potential of the ME composite laminates as very high sensitivity magnetic sensors by exploiting the magnetic flux concentration effect.

This work was financially supported by NSF under Grant No. ECCS-0824202.

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