Fe–Ga/Pb(Mg$_{1/3}$Nb$_{2/3}$)O$_3$–PbTiO$_3$ magnetoelectric laminate composites

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We have found large magnetoelectric (ME) effects in long-type laminate composites of Fe–20%Ga magnetostrictive alloys and piezoelectric Pb(Mg$_{1/3}$Nb$_{2/3}$)O$_3$–PbTiO$_3$ single crystals. At lower frequencies, the ME voltage coefficient of a laminate with longitudinally magnetized and longitudinally polarized (i.e., $L$–$L$ mode) layers was 1.41 V/Oe (or 1.01 V/cm Oe). Near the natural resonant frequency (~91 kHz) of the laminate, the ME voltage coefficients were found to be dramatically increased to 50.7 V/Oe (36.2 V/cm Oe) for the $L$–$L$ mode. In addition, the laminate can detect a minute magnetic field as low as $\sim 2 \times 10^{-12}$ T at resonance frequency, and $\sim 1 \times 10^{-10}$ T at lower frequencies. © 2005 American Institute of Physics.

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Magnetostriction occurs in most ferromagnetic materials. Rare-earth systems, such as Tb$_{0.3}$Dy$_{0.7}$Fe$_2$, exhibit a giant Joule magnetostriction at relatively low magnetic biases. 1 However, these rare-earth materials are expensive. It is also commonly known that ordinary Fe has a small magnetostrictive strain $(3/2)\lambda$, on the order of 30 ppm. However, the introduction of Ga into crystalline solution with Fe results in a significant enhancement of its magnetostriction. $(3/2)\lambda_{100}$ = 250 ppm, as long as the distorted AO2 phase remains stable—even though Ga reduces the spin density of the solution, it enhances its magnetostriction. Recently, a number of investigations have focused on Fe–Ga alloys (or Galfenol) due to the combination of its high mechanical strength, good ductility, relatively large $(3/2)\lambda_{100}$ values, low saturation fields, high blocking stress, and low cost. 2–7 Galfenol has potential applications in acoustic projectors, acoustic sensors, and actuators. 8,9

The magnetoelectric (ME) effect is a coupling effect between the magnetic field and electric field. ME effects have been found in many two-phase composites of magnetostrictive materials, such as Tb$_{1-x}$Dy$_x$Fe$_2$, Permendur, Ni$_{1-x}$Co$_x$Fe$_2$O$_4$ (i.e., NFO), or Co$_{1-x}$Zn$_x$Fe$_2$O$_4$ (i.e., CFO), and piezoelectric materials, such as Pb(Zr,Ti)O$_3$ or PMN–PT layers. 10–21 In our prior article, we reported the magnetostrictive and magnetoelectric behavior of Fe–20 at. %Ga/Pb(Zr,Ti)O$_3$ laminates. 10 In this letter, we will show that Fe–Ga/Pb(Mg$_{1/3}$Nb$_{2/3}$)O$_3$–PbTiO$_3$ (PMN–PT) ME laminates have much better ME coupling effect and higher sensitivity to a small magnetic signal.

Crystals of Fe–20 at. % Ga were grown by a Bridgman method at the Ames Laboratory. The Fe–20 at. % Ga crystals were cut into rectangular plates of dimensions $12.7 \times 6 \times 1$ mm, and were oriented along the $\langle001\rangle_c$. The crystals were annealed at 1100 °C for 168 h, using heating and cooling rates of 10 deg/min, after which the crystals were considered to be in the “slow-cooled” state. We measured the lattice parameter of one of our Fe–20%Ga crystals (in the free condition) as a function of magnetic bias $H_{dc}$, as shown in Fig. 1. The lattice parameter increased with increasing $H$, reaching a maximum near $H_{dc}$ = 300 Oe. Using these data, we then estimated the value of the magnetostrictive strain to be 262 ppm, consistent with prior results. 7 Maximum strain was found when $H_{dc}$ was applied along the longitudinal axis, or $\langle 010 \rangle_c$, of the plates. Accordingly, this longitudinal axis is here designated as the principal magnetization direction of our ME laminate design. Piezoelectric single crystals were (001)-oriented PMN–PT layers with (i) longitudinal polarization and (ii) transverse polarization. Two long-type Fe–20%Ga/PMN–PT three-layer laminates were prototyped by (i) sandwiching one longitudinally poled piezoelectric single crystal PMN–PT layer (dimensions of $14.0 \times 6 \times 1$ mm$^3$) between two longitudinally magnetized Fe–20%Ga layers, i.e., a $(L-L)$ mode laminate; and, as a comparison, (ii) sandwiching one transversely poled piezoelectric single crystal PMN–PT layer (dimension of $14.0 \times 6 \times 0.5$ mm) between two longitudinally magnetized Fe–20%Ga layers, i.e., a $(L-T)$ mode laminate. These configurations are similar to prior ME modes in Terfenol-D/PZT (Ref. 20) and CFO–NFO/PMN–PT layers of the laminate.

FIG. 1. Lattice parameter of a $(001)_c$ oriented Fe–20%Ga crystal as a function of dc magnetic bias. From the calculation of $\Delta L/L$ using the magnetostriction, the maximum strain of a Fe–20%Ga crystal at $H$ = 300 Oe should be 262 ppm.
The voltages induced across the two electrodes of the PMN–PT layer in the Fe–20%Ga/PMN–PT laminate composite were measured for various $H_{dc}$ and ac magnetic drives ($H_{ac}$) over the frequency range of $f = 0.1–100$ kHz, using a charge amplifier combined with a phase-referencing (i.e., lock-in) method. An electromagnet was used to apply a dc magnetic bias $H_{dc}$ and a pair of Helmholtz coils was used to generate a small $H_{ac}$ via an input current $i_{coil}$. For a ME composite with very low static capacitance, it is necessary to use a charge amplifier to obtain correct induced ME voltages.

Figure 2(a) shows the ME voltage coefficients for the L-L mode, $\partial V_{ME}^{LL}/\partial H$, as a function of magnetic field bias, $H_{dc}$. These data were taken at frequency of $f = 1$ kHz and a drive of $H_{dc} = 1$ Oe. The value of $\partial V_{ME}^{LL}/\partial H$ can be seen to be strongly dependent on $H_{dc}$. In the dc magnetic bias range $0 < H_{dc} < 700$ Oe, the ME voltages of the L-L mode of Fe–20%Ga/PMN–PT composites increased with increasing $H_{dc}$, reaching a maximum ME effect of $\partial V_{ME}^{LL}/\partial H \approx 1.41$ V/Oe at $H_{dc} = 750$ Oe. (or, correspondingly, $\partial V_{ME}^{LT}/\partial H = \approx 1$ V/cm Oe). For $H_{dc} > 750$ Oe, $\partial V_{ME}^{LL}/\partial H$ decreased dramatically with increasing $H_{dc}$, as the Fe–Ga layers of the laminate approached saturation of its magnetostriction. It is relevant to note that the maximum value of $\partial V_{ME}^{LL}/\partial H$ that we report for Fe–20%Ga/PMN–PT laminates is $\approx 4$ times higher than that for L-L configurations of Fe–20%Ga/PZT. It is also comparable to that of L-L or C-C configurations of Terfenol-D/PMN–PT, and $\approx 10$ times higher than that for L-T configurations of Terfenol-D/PZT or Terfenol-D/PMN–PT ones. A long-type laminate favors the optimum combination of magnetostrictive and piezoelectric effects; in particular, the longitudinal magnetostrictive strain of Fe–20%Ga, and the longitudinal piezoelectric strain of PMN–PT are higher, than the corresponding transverse ones.

As a comparison, Fig. 2(b) shows the ME voltage coefficients, $\partial V_{ME}^{LT}/\partial H$, for the L-T mode of Fe–20%Ga/PMN–PT laminates as a function of $H_{dc}$. Again, these data were also taken at a frequency of $f = 1$ kHz and a drive of $H_{dc} = 1$ Oe. The measured value of $\partial V_{ME}^{LT}/\partial H$ can be seen to be $\approx 30$ mV/Oe, which is a factor of $\approx 40 \times$ smaller than that of the L-L mode. However, its ME field coefficient, $\partial V_{ME}^{LT}/\partial H = 0.6$ V/cm Oe, is not much less than that of L-L mode.

We noted that ME voltage coefficients for both the L-L and L-T mode are antisymmetric about $H_{dc}$. In our previous reports, we only illustrated the ME voltage amplitude as a function of $H_{dc}$.

Magnetic field responses of the Fe–Ga alloy/PMN–PT laminates were then measured over a wider frequency range of $10^2–10^5$ Hz, as given in Fig. 3(a). The results show that the Fe–Ga/PMN–PT laminates have a much enhanced ME response when operated near its resonance frequency of $f_r \approx 86$ kHz. The maximum ME voltage coefficient at resonance for the L-L mode was $\approx 50.7$ V/Oe (or 36.2 V/cm Oe for the field coefficient), which is $\approx 36 \times$ higher than that in the low-frequency range; whereas the maximum ME voltage coefficient at resonance for the L-T mode was only $3.6$ V/Oe (or $\approx 70$ V/cm Oe).

Low-level magnetic field responses of the Fe–Ga/PMN–PT laminates operated in the L-T and L-L modes are given in Fig. 3(b). It can be seen that the induced ME voltages are near linear functions of $H_{dc}$. In this figure, the in-
duced ME voltage can be seen to have a good linear response to $H_{m}$ over a wide field range from $10^{-12}$ T (or $10^{-8}$ Oe) to $\sim 10^{-3}$ T (or 10 Oe). The limit of magnetic field sensitivity for the $L$-$L$ mode operated at resonance ($f_{r} = 8.6 \times 10^{6}$ Hz) was as low as $2.3 \times 10^{-5}$ (Oe), whereas its sensitivity at a low frequency of $f = 1 \times 10^{4}$ Hz was $\sim 1.2 \times 10^{-6}$ (Oe). As a comparison, Fig. 3(b) also illustrates the limit of magnetic field sensitivities for the $L$-$T$ mode operated both at resonance and low frequencies. It can be seen that the limit of magnetic field sensitivities for $L$-$T$ mode are $1.2 \times 10^{-7}$ (Oe) at resonance, and $\sim 2.5 \times 10^{-6}$ (Oe) at low frequencies. These results demonstrate that our Fe–Ga/PMN–PT laminates were exceptional sensitive to minute magnetic field variations.

In summary, laminate composites of Fe–20%Ga/PMN–PT have been found to have (i) a large $L$-$L$ ME voltage coefficient of $\partial V_{ME}^{LL}/\partial H > 1.4$ V/Oe, or $\partial E_{ME}^{LL}/\partial H = \sim 1.0$ V/cm Oe, under modest dc magnetic biases; (ii) a large $L$-$T$ ME field coefficient of $\partial E_{ME}^{LT}/\partial H = 0.6$ V/cm Oe, or $\partial V_{ME}^{LT}/\partial H = 0.03$ V/Oe, again under modest bias; (iii) a dramatic enhancement in the ME response near the resonance frequency; and (iv) a high sensitivity to minute magnetic field variations of $< 10^{-12}$ T (10$^{-8}$ Oe) at resonance.

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