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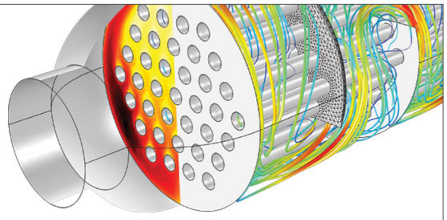

APPLIED PHYSICS LETTERS **87**, 222504 (2005)

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)λ, on the order of 30 ppm. However, the introduction of Ga into crystalline solution with Fe results in a significant enhancement of its magnetostriction, $(3/2)\lambda_{100} \approx 250$ 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 $Tb_{1-x}Dy_xFe_{2-y}$, Permendur, $Ni_{1-x}Co_xFe_2O_4$ (i.e., NFO), or $Co_{1-x}Zn_xFe_2O_4$ (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 12.7×6 ×1 mm, and were oriented along the $\langle 001\rangle_c$. 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 (dimensions of 14.0×6×0.5 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/

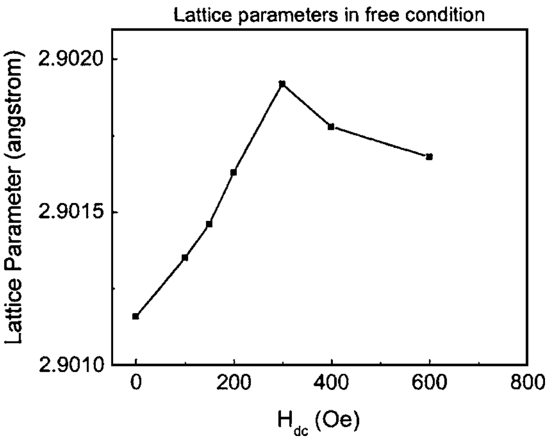

FIG. 1. Lattice parameter of a $\langle 001\rangle_c$ oriented Fe–20%Ga crystal as a function of dc magnetic bias. From the calculation of $(\Delta L/L)$ using the magnetostriction, the maximum strain of a Fe–20%Ga crystal at $H=300$ Oe should be 262 ppm.

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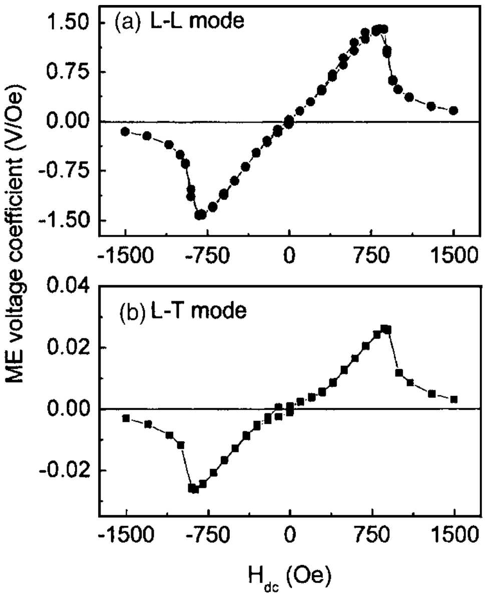

FIG. 2. ME voltage coefficient as a function of dc magnetic bias H_{dc} for (a) an *L-L* laminate and (b) *L-T* laminate. The static capacitance (after assembles) of the longitudinally poled PMN–PT layers was ~7.8 pF.

ferroelectric lead zirconate titanate (PZT) (Refs. 16 and 19) laminates.

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 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, $\partial V_{ME}^{L,L}/\partial H$, as a function of magnetic field bias, H_{dc}. These data were taken at frequency of $f=1$ kHz and a drive of $H_{ac}=1$ Oe. The value of $\partial V_{ME}^{L,L}/\partial H$ 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 $\partial V_{ME}^{L,L}/\partial H \sim 1.41$ V/Oe at $H_{dc} = 750$ Oe. (or, correspondingly, $\partial E_{ME}^{L,L}/\partial H = \sim 1$ V/cm Oe). For $H_{dc} > 750$ Oe, $\partial V_{ME}^{L,L}/\partial H$ 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 $\partial V_{ME}^{L,L}/\partial H$ that we report here for Fe–20%Ga/PMN–PT laminates is ~4× higher than that for *L-L* configurations of Fe–20%Ga/PZT.[10] It is also comparable to that of *L-L* or *C-C* configurations of Terfenol-D/PMN–PT,[20,22] and ~10× higher than that for *L-T* configurations of Terfenol-D/PZT or Terfenol-D/PMN-PT ones.[21] 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, $\partial V_{ME}^{L,T}/\partial H$, 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 $H_{ac}=1$ Oe. The measured value of $\partial V_{ME}^{L,T}/\partial H$ can be seen to be ~30 mV/Oe, which is a factor of ~40× smaller than that of the *L-L* mode. However, its ME field coefficient, $\partial E_{ME}^{L,T}/\partial H = 0.60$ V/cm Oe, is not much less than that of *L-L* mode.

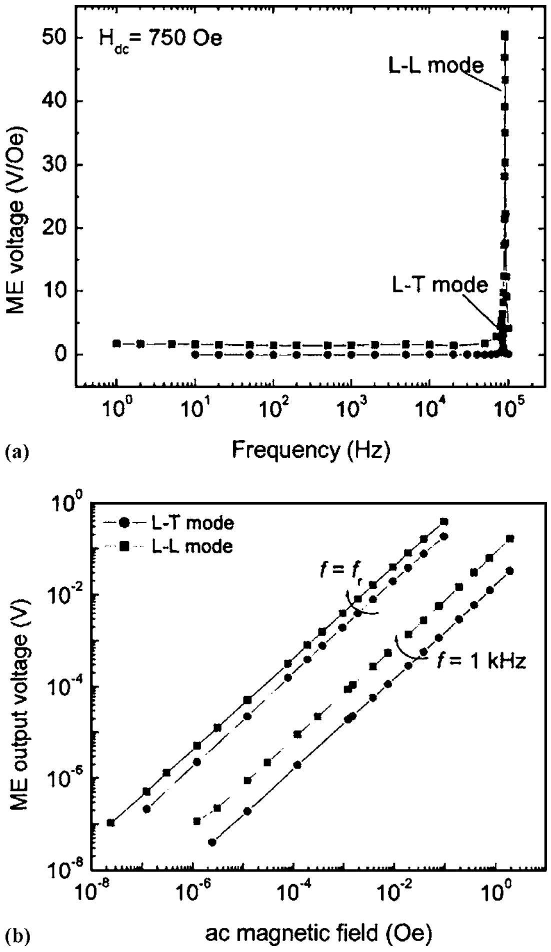

FIG. 3. ME responses of Fe–Ga/PMN–PT laminates, (a) induced ME voltage as a function of frequency of the ac magnetic field for both *L-L* and *L-T* modes. These data were taken under a constant dc magnetic bias of $H_{dc}=750$ Oe, and ac magnetic field of $H_{ac}=1$ Oe; and (b) induced ME voltages for *L-L* and *L-T* modes at low-frequency (1 kHz) and resonance frequency (f_r) under a $H_{dc}=750$ Oe as a function of ac magnetic field over the range of $10^{-8} < H_{ac} < 10$ Oe.

We noted that ME voltage coefficients for both the *L-L* and *L-T* mode are antisymmetric about H_{dc}. In our previous reports,[10] 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 $f_r = \sim 86$ kHz. The maximum ME voltage coefficient at resonance for the *L-L* mode was ~50.7 V/Oe (or 36.2 V/cm Oe for the field coefficient), which is ~36× 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 ~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_{ac}. In this figure, the induced ME voltage can be seen to have a good linear response to H_{ac} 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 ($f_r = 8.6 \times 10^4$ Hz) was as low as 2.3×10^{-8} (Oe), whereas its sensitivity at a low frequency of $f = 1 \times 10^3$ 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 1.2×10^{-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 $\partial V_{\mathrm{ME}}^{L,L}/\partial H > 1.4$ V/Oe, or $\partial E_{\mathrm{ME}}^{L,L}/\partial H = \sim 1.0$ V/cm Oe, under modest dc magnetic biases; (ii) a large *L-T* ME field coefficient of $\partial E_{\mathrm{ME}}^{L,T}/\partial H = 0.6$ V/cm Oe, or $\partial V_{\mathrm{ME}}^{L,T}/\partial H = 0.03$ 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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