Magnetostrictive and magnetoelectric behavior of Fe–20 at. % Ga/Pb(Zr,Ti)O3 laminates

Shuxiang Dong  
Virginia Polytechnic Institute and State University

Junyi Zhai  
Virginia Polytechnic Institute and State University

Feiming Bai  
Virginia Polytechnic Institute and State University

JieFang Li  
Virginia Polytechnic Institute and State University

D. Viehland  
Virginia Polytechnic Institute and State University

See next page for additional authors
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Abstract
The magnetostrictive and magnetoelectric (ME) properties of laminate composites of Fe–20 at. % Ga and Pb(Zr,Ti)O₃ (PZT) have been studied for laminates of different geometries. The results show that (i) a long-type magnetostrictive Fe–20 at. % Ga crystal plate oriented along \( \langle 001 \rangle \) and magnetized in its longitudinal (or length) direction has higher magnetostriction than a disk-type one; and consequently (ii) a long-type Fe–20 at. % Ga/PZT laminate has a giant ME effect, and is sensitive to low-level magnetic fields.

Keywords
iron alloys, gallium alloys, lead compounds, ferromagnetic materials, piezoceramics, laminates, magnetostriction, magnetoelectric effects, magnetisation

Disciplines
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Comments
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Authors
Shuxiang Dong, Junyi Zhai, Feiming Bai, JieFang Li, D. Viehland, and Thomas A. Lograsso
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Shuxiang Dong, Junyi Zhai, Feiming Bai, JieFang Li, and D. Viehland
Materials Science & Engineering, Virginia Tech, 306 Holden Hall, Virginia 24061
T. A. Lograsso
Materials & Engineering Physics, Ames Laboratory, Ames, Iowa 50011

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I. INTRODUCTION

Magnetostriiction occurs in most ferromagnetic materials. Rare-earth systems, such as Tb₉₋ₓDyₓ₀₋₂Fe₂, exhibit a giant Joule magnetostriiction at relatively low magnetic biases.¹ However, these rare-earth materials are expensive. It is also commonly known that ordinary Fe has a small magnetostriiction 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 magnetostriiction, (3/2)λ=250 ppm, as long as the distorted A₂ (bcc α-Fe) phase remains stable—even though Ga reduces the spin density of the solution, it enhances its magnetostriiction. Recently, a number of investigations have focused on Fe–Ga (Galfenol) alloys due to the combination of its high mechanical strength, good ductility, relatively large (3/2)λ 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

Magnetoelectric materials have been very interesting since early Swiss¹⁰ and Russian¹¹ work, and have recently had a renaissance (now commonly called multiferroic) in Nature, Science, Phys. Rev. Lett.,¹²⁻¹⁶ and other high-impact journals. The magnetoelectric (ME) effect is a polarization P response to an applied magnetic field H, or conversely a magnetization M response to an applied electric field E. Previously, ME effects have been reported in composites of piezoelectric Pb(Zr,Ti)O₃ (PZT) or Pb(Mg₁/₃Nb₂/₃)₀₋₁PbTiO₃ (PMN–PT) layers laminated with magnetostrictive Tb₁₋ₓDyₓFe₂₋ₓ (Fermendur), Ni₁₋ₓCoₓFe₂O₄ (NFO), or Co₁₋ₓZnₓFe₂O₄ (CFO) ones.¹⁷⁻²⁹

In this article, we will show that laminate composites of magnetostriective Fe–20 at. % Ga crystals and piezoelectric PZT ceramics also have a large ME coupling. Neither material is itself “magnetoelectric,” however, a large ME product property results from the elastic interaction of magnetostrictive and piezoelectric layers. Furthermore, Fe–Ga alloys have the advantages of low saturation fields, relatively high magnetostriiction, and low costs. These features offer Fe–Ga/PZT ME laminates potential in magnetic field and electric current sensing applications.

II. MAGNETOSTRICTION AND ME COUPLING MODES

A. Magnetostriective vibrational modes

Similar to magnetostriective Tb₁₋ₓDyₓFe₂₋ₓ (Terfenol-D) materials, the magnetostrijective strain of Fe–Ga crystals is anisotropic, depending significantly upon the direction along which a magnetic field H is applied. Consequently, Fe–Ga crystals can have large magnetostriective effects only when operated in particular modes. In addition, the magnetostrijective response of a Fe–Ga crystal to an applied H is also related to the crystal’s shape and size. However, for a given shape/size, there is a principal magnetostriective direction along the maximum dimension direction of the sample. Along this direction, the magnetostriective strain is a maximum. Correspondingly, the magnetostriective strain (or vibration) along this direction is defined as the principal strain (or vibration) mode. When H is applied parallel to the principal direction, the Fe–Ga crystal can be said to be operated in its longitudinal (or L) mode; whereas when H is applied perpendicular to this direction, it is designated as a transverse (or T) mode. As will be shown later in this article, L-mode long-type Fe–Ga crystal plates have larger magnetostriective strains under smaller applied magnetic fields, than the T-mode ones.

B. Magnetoelectric coupling modes

In laminate composites, such as the three-layer Fe–20 at. % Ga/PZT/Fe–20 at. % Ga one of this investigation, the layers of the bimaterial are stress coupled. When the magnetostriective layers are strained under H, the piezoelectric layers will undergo forced oscillation. Consequently, an electric field E (or voltage) is induced across the piezo-
Fe–20 at. % Ga crystals were oriented along the k plates of a diameter 12.7 mm and a thickness 1 mm. All and hard-type PZT with low piezoelectric constants but high soft-type piezoelectric PbO3 crystals were considered to be in the “slow-cooled” state. Both using heating and cooling rates of 10 °C/min, after which were prototyped by a Bridgman method at Ames Laboratory. The crystals were cut into rectangular plates of dimensions 12.7 × 6 × 1 mm³, or disk plates of a diameter 12.7 mm and a thickness 1 mm. All Fe–20 at. % Ga crystals were oriented along the (001) direction. The crystals were annealed at 1100 °C for 168 h, using heating and cooling rates of 10 °C/min, after which they were considered to be in the “slow-cooled” state. Both soft-type piezoelectric Pb(Zr,Ti)O₃ (PZT) with high d₅₃ piezoelectric constants but low mechanical quality factor Qₘ, and hard-type PZT with low piezoelectric constants but high Qₘ were used for ME laminates.

Three-layer long-type Fe–20 at. % Ga and PZT laminates were prototyped by (i) sandwiching one longitudinally poled rectangular PZT plate (soft or hard type, sizes: 14 × 6 × 1 mm³) between two longitudinally magnetized Fe–20 at. % Ga ones, i.e., (L–L) mode laminates (prototype No. 1 made of soft PZT, and prototype No. 2 made of hard); and (ii) sandwiching one transversely poled rectangular PZT plate (soft or hard, sizes: 14 × 6 × 0.5 mm³) between two longitudinally magnetized Fe–20Ga ones, i.e., (L–T) mode laminates (prototype No. 3 made of soft PZT, and prototype No. 4 of hard). The prototypes were laminated using epoxy resin, and were cured at 80 °C for 3–4 h under load. These configurations are similar to prior ME modes in Terfenol-D/PZT¹⁷ and CFO–NFO/PZT¹⁵,¹⁸ laminates. The static capacitance of the transversely poled PZT layers was 2.04 nF, whereas that of longitudinally poled PZT layers was only 0.036 nF. Figure 1 illustrates the laminate configurations of various operational modes.

The magnetostriction of the Fe–20 at. % Ga layers and Fe–20 at. % Ga/PZT laminated composites were measured, via a resistance strain-gauge method. The voltages induced across the two ends of the PZT layer in the Fe–20 at. % Ga/PZT laminate were measured for various dc magnetic biases (Hₐc) and ac magnetic drives (Hₐc) over the frequency range of 10⁻² < f < 10⁵ Hz, using a charge amplifier combined with a phase-locking (i.e., lock-in) method. An electromagnet was used to apply dc magnetic bias Hₐc and one pair of Helmholtz coils was used to generate a small Hₐc, via an input current Icoil, which was superimposed on Hₐc. Since the L–L ME composites have a very low static capacitance, we found it necessary to use a charge amplifier to obtain correct induced ME voltages, as the distributed capacitance of the connecting cables and electronic meters could notably affect the measured values.

III. EXPERIMENTAL PROCEDURE

Crystals of Fe–20 at. % Ga were grown by a Bridgman method at Ames Laboratory. The crystals were cut into rectangular plates of dimensions 12.7 × 6 × 1 mm³, or disk plates of a diameter 12.7 mm and a thickness 1 mm. All Fe–20 at. % Ga crystals were annealed at 1100 °C for 168 h, using heating and cooling rates of 10 °C/min, after which they were considered to be in the “slow-cooled” state. Both soft-type piezoelectric Pb(Zr,Ti)O₃ (PZT) with high d₅₃ piezoelectric constants but low mechanical quality factor Qₘ, and hard-type PZT with low piezoelectric constants but high Qₘ were used for ME laminates.

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IV. RESULTS AND DISCUSSION

A. Magnetostriction of Fe–20 at. % Ga crystals

To determine an optimum magnetostriction mode, two types of (001), Fe–20 at. % Ga crystals plates—a long type and a disk type—were studied. These measurements clearly confirmed that an L-mode long-type Fe–20 at. % Ga crystal plate has larger magnetostriction at low magnetic fields. This is important in understanding the ME properties that will subsequently be presented in this article. Accordingly, in our ME studies, we have focused on this long-type configuration.

Figure 2 shows the εₘ–H response for a long-type rectangular Fe–20 at. % Ga crystal plate. Data are shown for both the longitudinal (expansion) strain where Hₐc is applied along the length of the rectangular Fe–Ga plate (i.e., L mode), and the transverse (contraction) strain where Hₐc is applied along the thickness of the rectangular plate (i.e., T mode). Higher magnetostrictive strains at lower biases (0–700 Oe) were found when H was applied along the longitudinal direction, relative to the transverse. The longitudinal magnetostrictive strain of a Fe–20 at. % Ga crystal plate was 330 ppm at Hₐc=700 Oe, whereas the transverse one was only 5 ppm. At lower Hₐc, the L-mode magnetostriction is a factor of ~60× higher, than that of the T mode. However, for H > 700 Oe, the magnetostriction of the L mode saturated; whereas that of the T mode did not saturate until
and transverse shape changes. For Fe–20 at. % Ga crystal plate and the induced longitudinal

The insets of Fig. 2 illustrate a photo of a rectangular shaped

higher demagnetization factor

$N$

tor angular type, the disk type has a higher demagnetization fac-
tive strain of the disk-type plate was only 100 ppm at

$H = 1000$ Oe; whereas, the maximum

tive piezomagnetic coefficient

$K_{dx}$

$\frac{\partial V_{ME}}{\partial H} \sim 345$ mV/Oe at a magnetic bias of $H_{dc}$

mode is required to reach saturation. However, their $T$ modes
have almost the same demagnetization factor $N$; Compari-
sions of the data in Figs. 2 and 3 will show that the magne-
tostrictive strain of the disk-type laminate is much lower than
that of the long-type ones for $0 < H_{dc} < 1000$ Oe. The insets
of Fig. 3 illustrate a photo of a disk-type Fe–20 at. % Ga
crystal plate, and the induced longitudinal (diameter) and
transverse (thickness) shape changes. For $H_{dc}$ applied longi-
dudinally, the disk-type crystals tend to become elliptical, and
the thickness is decreased; whereas for $H_{dc}$ applied in thick-
dness direction, the thickness expands and the diameter is
decreased.

B. Magnetostriction of Fe–20 at. % Ga/PZT laminates

The magnetostrictive strain for a long-type

Fe–20 at. % Ga/PZT composite was remeasured after lami-
nation, using a resistance strain gauge. These measurements
revealed a maximum magnetostrictive (expansion) strain
along the laminate’s length of $\sim 70$ ppm at a $H_{dc}=1000$ Oe,
as can be seen in Fig. 4. Comparisons of these data to those
of the free condition (see Fig. 2) will reveal that lamination
with PZT layers imposes a load to the Fe–20 at. % Ga lay-
ers which: (i) significantly decreases the magnetostrictive
strain relative to that of the free condition of the crystal; (ii)
lags the magnetostrictive response until 500 Oe; and (iii)
shifts the maximum strain to a higher $H_{dc}$, presumably due to
suppression of magnetic domain wall motion. Figure 4 also
illustrates the differential of the magnetostrictive strain to an
applied $H_{dc}$, which is a measure of the change in the effective
piezomagnetic coefficients with $H_{dc}$. To obtain a large effec-
tive piezomagnetic coefficient (i.e., $\delta \epsilon / \delta H$), these data show
that a dc bias of $H_{dc}=800$ Oe is required. Correspondingly, a
similar bias of $H_{dc}=800$ Oe will be needed to achieve opti-
mum ME effects.

C. ME responses

Figure 5(a) shows the ME voltage coefficient for a long-
type three-layer Fe–Ga/PZT/Fe–Ga laminate (prototype No.
1) as a function of $H_{dc}$ for both $L$–$L$ and $T$–$L$ modes. The
data in this figure were taken at a frequency of $f=1$ kHz and
a drive of $H_{dc}=1$ Oe. The value of $\partial V_{ME}^{L-L} / \partial H$ can be seen to
be strongly dependent on $H_{dc}$. The results show that the $L$–$L$
mode of Fe–20%Ga/PZT laminates has a maximum ME ef-
effect of $\partial V_{ME}^{L-L} / \partial H \sim 345$ mV/Oe at a magnetic bias of $H_{dc}$

FIG. 2. Magnetostrictive strains of free (001), Fe–20 at. % Ga crystals of various geometries: (a) a rectangle of dimensions $12 \times 6 \times 1$ mm$^3$ and (b) a disk of diameter $12.7$ mm and a thickness of $1$ mm.

FIG. 3. Magnetostrictive strains of free rectangular type Fe–20 at. % Ga crystal plates (of dimensions $12.7 \times 6.0 \times 1.0$ mm$^3$) as a function of dc magnetic bias $H_{dc}$.

FIG. 4. Magnetostriction of Fe–Ga/PZT/Fe–Ga laminate as a function of $H_{dc}$, and its differential dependence on $H_{dc}$. 

FIG. 5(a) shows the ME voltage coefficient for a long-type three-layer Fe–Ga/PZT/Fe–Ga laminate (prototype No. 1) as a function of $H_{dc}$ for both $L$–$L$ and $T$–$L$ modes. The data in this figure were taken at a frequency of $f=1$ kHz and a drive of $H_{dc}=1$ Oe. The value of $\partial V_{ME}^{L-L} / \partial H$ can be seen to be strongly dependent on $H_{dc}$. The results show that the $L$–$L$ mode of Fe–20%Ga/PZT laminates has a maximum ME effect of $\partial V_{ME}^{L-L} / \partial H \sim 345$ mV/Oe at a magnetic bias of $H_{dc}$.
=800 Oe, where the slope of the magnetostriction of the laminate shown in Fig. 4 is highest. For \( H_{dc} > 800 \) Oe, \( \partial V_{ME}^{L-L} / \partial H \) decreased dramatically with increasing \( H_{dc} \), as the Fe–20 at. % 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/PZT laminates is close to that of \( L-L \) configurations of Terfenol-D/PZT and Terfenol-D/PMN–PT ones,\(^{26,28} \) even though the saturated magnetostriction (\( \lambda_s \)) of Fe–Ga is smaller than that of Terfenol-D. This demonstrates that Fe–20 at. % Ga has comparably magnetoelastic coupling as that of Terfenol-D, under moderate magnetic fields. A long-type laminate favors the optimum combination of magnetostrictive and piezoelectric effects; in particular, the longitudinal magnetostrictive strain of Fe–20 at. % Ga, and the longitudinal piezoelectric strain of PZT are higher than the corresponding transverse ones. For the \( T-L \) mode, we also observed a relatively large ME voltage coefficient of ~220 mV/Oe at a notably higher dc magnetic bias of \( H_{dc} = 1500 \) Oe, as shown in Fig. 5(b).

In addition, it is relevant to note that the value of the transverse magnetostriction for a free Fe–20 at. % Ga crystal was quite low for \( H_{dc} < 1000 \) Oe. However, when this same crystal was laminated to form a ME composite and operated in a \( T \) mode, a relatively large ME voltage coefficient of ~220 mV/Oe was still observed. This indicates that there is another factor influencing the ME coupling, presumably the elasto–electric coupling factor. This possibility is supported by reports\(^{30} \) of increases in effective piezoelectric constants under uniaxial stress, which could enhance elastic–electric coupling, consequently increasing the ME output voltage.

For comparisons, Fig. 6 shows the ME voltage coefficients (prototype No. 3) for both the \( L-T \) and \( T-T \) modes as a function of \( H_{dc} \). These data were also taken at a frequency of \( f = 1 \) kHz and a drive of \( H_{ac} = 1 \) Oe. The measured value of the ME voltage coefficients can be seen to be ~33 and ~4 mV/Oe for the \( L-T \) and \( T-T \) modes, respectively. This is a factor of \( \sim 10^3 \) smaller than that of the \( L-L \) mode. However, the corresponding ME field coefficient was larger for the \( L-T \) mode \( [(\partial E_{ME}^{L-T} / \partial H) = 640 \) mV/cm Oe], relative to that of the \( L-L \) mode \( [(\partial E_{ME}^{L-L} / \partial H) = 272 \) mV/cm Oe].

D. ME sensitivity

Low-level magnetic field responses of the Fe–Ga/PZT laminate (prototype No. 1) operated in the \( L-L \) mode are shown in Fig. 7. It can be seen that the induced ME voltage is a near linear function 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 \( 10^{-9} \) T (or \( 10^{-5} \) Oe) to \( \sim 10^{-3} \) T (or 10 Oe). These results demonstrate that our Fe–Ga/PZT laminate is quite sensitive to minute magnetic field variations. Further sensitivity improvements should be possible by replacing the PZT layers in the laminate with PMN–PT single crystal ones, which have significantly higher piezoelectric coefficients.\(^{26} \)

![Figure 5](image1.png)  
**FIG. 5.** ME voltage coefficients of prototype No. 1 at \( f = 1 \) kHz for various modes: (a) \( L-L \) and (b) \( T-L \).

![Figure 6](image2.png)  
**FIG. 6.** ME voltage coefficients of prototype No. 2 at \( f = 1 \) kHz for various modes: (a) \( L-T \) and (b) \( T-T \).

![Figure 7](image3.png)  
**FIG. 7.** Illustration of the magnetic field sensitivity. The induced ME voltage for prototype No. 1 under a \( H_{dc} = 750 \) Oe and a measurement frequency of \( f = 1 \) kHz as a function of ac magnetic field over the range of \( 10^{-9} < H_{ac} < 10^{-3} \) T.
E. Frequency dependence of ME response

The frequency dependence of the induced ME voltage for the Fe–Ga/PZT laminates was then measured over a wider frequency range of $10^{-2} < f < 10^3$ Hz. The results show that the Fe–Ga/PZT laminate (prototype No. 1) has a very flat frequency response in the low-frequency range of $10^{-2} < f < 10^3$ Hz, as can be seen in Fig. 8.

Upon approaching the natural resonance frequency, the induced ME voltage for both $L-L$ and $L-T$ modes was significantly enhanced, as shown in Fig. 9. The maximum ME voltage coefficient at resonance ($f_0 = 92.5$ kHz) for the $L-L$ mode (prototype No. 2) was approximately $5.7$ V/cm Oe (or correspondingly $5.7$ V/cm Oe for the field coefficient); whereas, for the $L-T$ mode (prototype No. 4), it was $3.3$ V/cm Oe (correspondingly $66$ V/cm Oe) at a resonance frequency of $f_0 = 96$ kHz. Clearly, the ME voltage coefficients at resonance are $20-100$ higher than those at subresonant conditions. (Note that the resonance frequencies for the $L-L$ and $L-T$ modes are different because their magnetostrictive layer thickness ratios are different, resulting from a difference in their mean acoustic velocities.)

Although at low frequencies ME laminates of hard PZT layers have lower induced ME voltages than those with soft ones, the opposite is true under resonant conditions—i.e., laminates of hard PZT have higher ME coefficients near $f_0$. This is because hard PZT has a higher mechanical quality factor $Q_m$ than soft types. These values of ME voltage coefficients achieved from Fe–Ga/PZT laminates are comparable to, or slightly higher than, previous reports for Terfenol-D/PZT and CFO–NFO/PZT laminates.

V. SUMMARY

In summary, a long-type magnetostrictive Fe–20 at. % Ga crystal plate has been found to have a higher magnetostrictive strain at lower fields, than a disk-type one. Furthermore, this long-type laminate of Fe–20% Ga and PZT has been found to have: (i) a large $L-L$ ME voltage coefficient of $\delta V_{L-L}^{\text{ME}}/\delta H > 345$ mV/Oe under modest dc magnetic biases; (ii) a dramatic enhancement in the ME response near the resonance frequency; and (iii) a high sensitivity to minute magnetic field variations. These results demonstrate the feasibility of fabricating low-cost, highly-sensitive magnetic field and/or electric current sensors using Fe–20 at. % Ga/PZT laminates.

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