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Shuxiang Dong, Jie-Fang Li, and D. Viehland

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Characterization of magnetoelectric laminate composites operated in longitudinal-transverse and transverse–transverse modes

Shuxiang Dong, a) Jie-Fang Li, and D. Viehland

Materials Science and Engineering, Virginia Tech, Blacksburg, Virginia 24061

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Magnetostrictive and piezoelectric laminate composites of terfenol-D and Pb(Zr1−xTi3)O3 have been studied. The magnetoelastic (ME) coefficients have been characterized for the different operational modes: (i) a longitudinally magnetized and transversely polarized longitudinal transverse (LT) mode, and (ii) a transversely magnetized and transversely polarized transverse–transverse (TT) mode. The results demonstrate that the (LT) magnetoelastic mode has dramatically higher ME coefficients than the TT one. The LT magnetoelastic coefficient is up to 5–7 times larger than the TT one, when operated in low magnetic bias ranges. © 2004 American Institute of Physics. [DOI: 10.1063/1.1644027]

I. INTRODUCTION

The magnetoelectric effect is a polarization \( \vec{P} \) response to an applied magnetic field \( \vec{H} \), or conversely a magnetization \( \vec{M} \) response to an applied electric field \( \vec{E} \). Magnetoelectric (ME) single-phase materials have been studied,\(^2\)–4 most are ferroelectromagnetic. To date, a single phase material with a high inherent coupling between magnetization and polarization has not been found.

Magnetostrictive behavior also exists as a composite effect in multiphase systems of piezoelectric and magnetostrictive materials.\(^5\)–14 Piezoelectric/magnetostrictive composites have been investigated experimentally and analytically. Ceramic–ceramic particle composites of two phases\(^5\) and of three phases\(^6\) (i.e., Terfenol-D/piezoelectric ceramic particles/polymer composites) have been studied. Investigations have shown that the ME coupling of particulate composites is low.

Magnetostrictive–piezoelectric laminate composites have much higher ME coefficients than that of single-phase materials or particulate composites.\(^7\)–14 This has been shown both by experiment and by phenomenological analysis. Previous investigations of ME laminates have focused on piezoelectric and magnetostrictive layers that were, respectively, poled/magnetized along their thickness directions.\(^7\)–8,11,14 For previous laminate designs, experimental and analytical investigations have shown relatively large ME coefficients, but only under high dc magnetic bias, and where the transverse coefficients (perpendicular to thickness) are significantly larger than the longitudinal (parallel to thickness) ones.\(^9\)–10,12,13

In this article, the working modes and induced magnetoelectric voltage behavior of a long-type laminate, respectively, magnetized in the longitudinal (length) direction and poled in the transverse (thickness) direction, are investigated. We will show that our laminate design has much larger longitudinal ME voltage coefficients, relative to transverse ones, when operated in low magnetic bias ranges. We believe that the approach is important for magnetic field sensor applications and future laminate designs.

II. OPERATIONAL MODES AND ME COUPLING OF LAMINATE COMPOSITES

A. Magnetostrictive (L-mode) and (T-mode) of terfenol-D

Terfenol-D materials have a superior magnetostrictive strain \( \varepsilon_m \) and magnetoelastic coupling factor \( k_m \). However, the magnetostrictive strain is anisotropic, depending significantly upon the direction along which the magnetic field \( \vec{H} \) is applied. Consequently, terfenol-D can have large magnetostrictive effects only when operated in particular modes.

Figure 1 shows the \( \varepsilon_m - \vec{H} \) response for a long-type terfenol-D plate that is grain-oriented in the thickness direction (i.e., (211)). Data are shown for both a longitudinal strain where \( \vec{H} \) is applied along the length of the Terfenol-D plate, and a transverse strain where \( \vec{H} \) is applied along the thickness of the Terfenol-D plate. Much larger induced strains were found when the magnetic field is applied along the longitudinal direction, relative to the transverse direction (though the grain orientation is in the transverse or thickness direction). The longitudinal magnetostrictive strain of a terfenol-D plate was \( \sim 8 \) times greater than that of the transverse strain. The inserts of Fig. 1 illustrate the induced shape changes of rectangular shaped plates by an \( \vec{H} \) applied along the longitudinal and transverse directions. For \( \vec{H} \) applied longitudinally, the length of the rectangular plate expands and its thickness contracts; whereas for \( \vec{H} \) applied transversely, the length of the plate contracts and its thickness expands. Because the principle strain (expanding or contracting) of the Terfenol-D plate is along the longitudinal direction, we define the magnetostrictive modes relative to the principal strain direction in terms of the piezoelectric modes.\(^15\) (i) when the applied magnetic field \( \vec{H} \) is parallel to the principle...
strain (length) direction, the Terfenol-D plate is designated to be operated in its longitudinal magnetostrictive mode (L-mode); and (ii) when the applied magnetic field \( \vec{H} \) is perpendicular to the principle strain (thickness) direction, the Terfenol-D plate is designated to be operated in its transverse magnetostrictive mode (T-mode).

B. Magnetolectric (L-T) and (T-T) modes

Figure 2 illustrates our three-layer [terfenol-D//PZT//terfenol-D] laminate composite. The magnetostrictive plates were magnetized along the longitudinal or length direction, which is the direction of largest \( e_m \). The piezoelectric plates were polarized in their thickness direction. This long-type configuration intensifies the principle strain/vibration along the longitudinal axis. Correspondingly, this favors the tensor property matrix for Terfenol-D, as its longitudinal strain is much higher than its transverse. Our configuration is significantly different than previous reports, where both the magnetostrictive and piezoelectric layers were magnetized/polarized in their thickness directions. Our design was chosen so that the laminate could be efficiently operated in the L-mode. Because of the significantly larger \( e_m \) in the L-mode, the ME effect will be pronouncedly larger relative to the conventional T-mode one.

When \( \vec{H} \) is applied along the longitudinal axis of the laminate [see Fig. 2(b)], both longitudinal (33) and transverse (31) piezomagnetic or magnetostrictive modes will be excited. However, because the lamimates length is significantly larger than its other physical dimensions, the longitudinal (33) vibration mode will be intensified as the principle vibrational mode, and the transverse (31) mode can be neglected. When \( \vec{H} \) is applied along the thickness direction of the laminate [see Fig. 2(c)], again both longitudinal and transverse magnetostrictive modes will be excited. However, because of the geometrical design of the laminate, the (31) vibration mode (longitudinal direction) will be intensified as the principle vibrational mode, and the (33) mode can be neglected.

Because the layers of the bimaterial are stress coupled, when the magnetostrictive plates are strained under \( \vec{H} \), the piezoelectric plates (which are poled in their thickness direction) will undergo forced oscillation. Consequently, an electric field \( \vec{E} \) (or voltage) is induced across the thickness of the piezoelectric plate due to piezoelectric effect. This mechanically coupled response from applied magnetic field to induced electric field is called as the magnetolectric (ME) effect of the laminate.

Relative to the principle vibrational mode, when the applied magnetic field \( \vec{H} \) is parallel to and the induced electric field \( \vec{E} \) from piezoelectric plate is perpendicular to principle vibration mode, the magnetoelastic laminate is designated to be operated in its longitudinal-transverse (LT) mode. When both the applied \( \vec{H} \) and induced \( \vec{E} \) are perpendicular to the principle vibration mode, the magneto-electric laminated is designated to be operated in its transverse–transverse (TT) mode.

C. Magnetoelastoelectric coupling

Based on the magnetoelastoelectric equivalent circuit method, the longitudinal \( \left| \frac{dV}{dH_3} \right|_{(LT)} \) and transverse \( \left| \frac{dV}{dH_3} \right|_{(TT)} \) ME voltage coefficients can be derived as

\[
\frac{dV}{dH_3} \biggr|_{LT} = -\frac{n(1-n)A_d d_{31,m}^2}{\varepsilon_3^2 \varepsilon_m^2 \varepsilon_1^0} \left[ (1 + n^2 d_{31,m}^2) \right] + (1 - n) \left( s_{13}^m s_{33}^m \right) \right]
\]

(1a)

\[
\frac{dV}{dH_3} \biggr|_{TT} = -\frac{n(1-n)A_d d_{31,m}^2}{\varepsilon_3^2 \varepsilon_m^2 \varepsilon_1^0} \left[ (1 + n^2 d_{31,m}^2) \right] + (1 - n) \left( s_{11}^m s_{11}^m \right) \right]
\]

(1b)

where \( V \) is the induced ME voltage, \( H_3 \) (subscript “3” indicates the magnetization direction in piezomagnetic constitutive equation) is the exciting ac magnetic field, \( H_\infty \), \( \beta \) is a factor related to applied magnetic field bias \( H_b \) and at the optimum \( H_b=0 \), its value equals 1; \( s_{11}^m \) and \( s_{33}^m \) are the elastic compliances.
of the piezoelectric and magnetostrictive layers, \( k_{31,p} \) and \( \varepsilon_{33}^T \) are the longitudinal piezomagnetic and transverse piezoelectric coefficients, \( n \) is a geometric thickness ratio of the Terfenol-D layer to the total thickness of the laminate, and \( A \) is the cross area of the laminate layers. Table I lists the relevant materials parameters needed to predict the values of the ME voltage coefficients for terfenol-D and PZT.

In Eq. (1), it can be seen that high piezomagnetic and piezoelectric coefficients result in a large ME voltage coefficient. In particular, the ME voltage coefficient is very sensitive to the piezoelectric constant, as it is proportional to \( d_{31,p}^2 \). Detrimental to the ME coefficient is a high dielectric constant and a high elastic compliance. Using the materials parameters in Table I and the laminate geometry shown in Fig. 2, the (L-T) mode ME voltage coefficient \( |dV/dH_3|_{LT} \) can be predicted to be \( \sim 5 \) times greater than that of the (TT) one. This is due to the larger magnetically induced strain \( \varepsilon_{mL} \) for the longitudinal direction, as shown in Fig. 1.

Equation (1) offers an important theoretical tool for the design and optimization of the laminate composite of terfenol-D and PZT. According to Eq. (1), the maximum ME voltage coefficients of the ME laminate operated in (LT) or (TT) mode can be estimated as (i) the maximum value for \( |dV/dH_3|_{TT} \) at \( n \sim 0.76 \) is \( \sim 11 \) (mV/Oe), and the maximum value for \( |dV/dH_3|_{LT} \) at \( n \sim 0.64 \) is \( \sim 54 \) (mV/Oe). We will see that this estimation is close to our measured values.

### III. EXPERIMENTAL PROCEDURE

A long rectangular-shaped laminate of terfenol-D and PZT (samples No. 4) was fabricated. The terfenol-D plates were grain oriented in their thickness direction, and the PZT plates were poled in their thickness direction. The dimensions of the terfenol-D plates were \( 12.0 \times 6.0 \times 1.0 \) mm\(^3\), and the dimensions of PZT plate was \( 12.0 \times 6.0 \times 0.5 \) mm\(^3\). The PZT plate was laminated between terfenol-D plates using a conductive epoxy resin (E-solder), and cured at 80 °C for 3–4 h under load. Figure 3 shows a photo of the prototype laminates. The prototypes were operated in both the (LT) and (TT) modes. The mode of operation is determined by the direction of the applied dc magnetic bias \( H_{dc} \) and ac magnetic field \( H_{ac} \), as shown in Figs. 2(b) and 2(c).

For comparisons, a conventional disc type three-layer laminate of terfenol-D and PZT (sample No. 3) was assembled, in which the magnetization and polarization were both oriented in their thickness directions, i.e., the (TT) mode. The terfenol-D discs were 12.7 mm in diameter and 1.0 mm in thickness; and the PZT discs (APC840) were 12.7 mm in diameter and 0.5 mm in thickness. Again, the three layers were laminated together using a conductive epoxy (E-solder).

Figure 4 illustrates the measurement system used for the characterization of the ME effect. An electromagnet was used to apply a dc magnetic bias \( H_{dc} \) of 70 to 4000 Oe. Small Helmholtz coils were used to excite an ac magnetic field \( H_{ac} \) of 0.4 to 1.2 Oe, which was superimposed on \( H_{dc} \). The ME voltage induced on the laminate by \( H_{dc} \) was detected using a lock-in amplifier. The laminates were placed in the center of small Helmholtz coils. If the laminates longitudinal axis is parallel to \( H_{ac} \), it is operated in the (LT) mode. Whereas, if the sample’s longitudinal axis and main face are perpendicular to \( H_{ac} \), it is operated in the (TT) mode.

<table>
<thead>
<tr>
<th>Density (kg/m(^3))</th>
<th>Elastic constants ((\times 10^{-12} \text{ m}^2/\text{N}))</th>
<th>Piezoelectric/magnetic Constants</th>
<th>Coupling factor ( k_{11} )</th>
<th>( \varepsilon_{33}^T/e_0 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>PZT-5(^a)</td>
<td>7600</td>
<td>((s_{11}^p))</td>
<td>((d_{33,p}))</td>
<td>580 (pC/N)</td>
</tr>
<tr>
<td>T-D(^c)</td>
<td>9230</td>
<td>((s_{11}^p))</td>
<td>((d_{33,m}))</td>
<td>40</td>
</tr>
</tbody>
</table>

\(^a\)Cited from Sunnytec Company, Suzhou, China.
\(^b\)Measured value after assembly.
\(^c\)Cited from Reference 16.
IV. RESULTS AND DISCUSSION

A. (TT) Magnetoelectric mode

The two laminate prototypes (No. 3 and No. 4) were first operated in a (TT) mode. Characterization was performed using a small ac magnetic signal $H_{ac}$ and a measurement frequency of $f = 1$ kHz under different magnetic bias.

Figures 5(a)–5(c) show the transverse induced ME voltage, $V_{TT}$, of the laminate as a function of $H_{ac}$ at various $H_{dc}$. For a given $H_{dc}$, the value of $V_{TT}$ can be seen to vary linearly with $H_{ac}$, increasing in magnitude with increasing $H_{dc}$. For $H_{dc} = 70$ Oe, the value of $V_{TT}$ was $\approx 1.0–1.2$ mV under an ac magnetic field of 1.0 Oe (peak) [see Fig. 5(a)]; however for $H_{dc} = 500$ Oe, $V_{TT}$ was increased to $\approx 9–11$ mV [see Fig. 5(b)]. An even higher $V_{TT}$ of $17–22$ mV was obtained for $H_{dc} = 1000$ Oe [see Fig. 5(c)]. These results clearly demonstrate a linear coupling between the measured voltage and $H_{ac}$, at various magnetic biases between 0 and 1000 Oe.

Figure 6 shows the magnetoelectric voltage coefficient $|dV/dH_{ac}|_{TT}$ as a function of $H_{dc}$ for $f = 1$ kHz and $H_{ac} = 1.0$ Oe (peak). The value of $|dV/dH_{ac}|_{TT}$ can be seen to increase in a near linear manner with increasing $H_{dc}$ over the range of $0 < H_{dc} < 1000$ Oe. With increasing $H_{dc}$ above this range, a maximum in $|dV/dH_{ac}|_{TT}$ was gradually approached. The maximum observed value was $|dV/dH_{ac}|_{TT} = 55–65$ V/Oe, which was obtained for $H_{dc} = 3000$ Oe. In the range of $H_{dc} > 3000$ Oe, $|dV/dH_{ac}|_{TT}$ tended to its saturation value.

1. (LT) Magnetoelectric mode

The two laminate prototypes (No. 3 and No. 4) were next operated in a (LT) mode. Figures 7(a)–7(c) show the longitudinal induced magnetoelectric voltage $V_{LT}$ as a function of $H_{ac}$ at various $H_{dc}$. For a constant $H_{dc}$, the value of $V_{LT}$ was linearly proportional to $H_{ac}$. Under constant drive and bias conditions, $V_{LT}$ was dramatically higher for the (LT) mode, relative to the (TT) one. For $H_{dc} = 70$ Oe, the value of $V_{LT}$ for sample No. 4 was $\approx 9$ mV ($H_{ac} = 1.0$ Oe, and $f = 1$ kHz), as shown in Fig. 7(a). This is $\approx 7$ times higher than that of the (TT) mode. Although, under these operational conditions, the conventional disc-type sample (No. 3), operated in its LT mode, had a very low induced ME voltage. When the bias was increased to 500 Oe, the value of $V_{LT}$ for sample No. 4 was increased to $\approx 55$ mV [see Fig. 7(b)].
which is 5 times higher than that of the (TT) mode at the same bias, and which is also close to our calculated value of 54 mV. With increasing bias to 1000 Oe, \( V_{LT} \) was slightly decreased [see Fig. 7(c)]; however it remained significantly higher than that of the (TT) mode operated under the same conditions.

Figure 8 shows the magnetoelectric voltage coefficient \( |dV/dH_3|_{LT} \) as a function of \( H_{dc} \), for \( H_{dc} = 1.0 \text{ Oe} \) and \( f = 1 \text{ kHz} \). In the range of 0 \(< H_{dc} < 400 \) Oe, \( |dV/dH_3|_{LT} \) can be seen to increase in a near-linear manner with \( H_{dc} \). For sample No. 4, a maximum value of \( |dV/dH_3|_{LT} = 56 \text{ mV/Oe} \) was reached for \( 500 < H_{dc} < 700 \) Oe. For \( H_{dc} > 10^3 \) Oe, \( |dV/dH_3|_{LT} \) decreased gradually with increase of \( H_{dc} \). The maximum value of \( |dV/dH_3|_{LT} \) corresponds to the maximum of the slope in the \( \varepsilon_m - H \) response, shown in Fig. 1. Above this point, saturation of \( \varepsilon_m \) is approached.

V. SUMMARY

Laminates of magnetostrictive terfenol-D and piezoelectric PZT have been characterized. The ME laminates were operated in both a longitudinal magnetized/transverse polarized (LT) mode, and a transverse magnetized/transverse polarized (TT) mode. Clearly, the (LT) magnetoelectric mode has significantly higher ME effects than the conventional (TT) one. In particular, under moderate magnetic biases, the (LT) ME effect is dramatically higher. The results have shown a maximum value of \( |dV/dH_3|_{LT} = 56 \text{ mV/Oe} \) at a dc magnetic bias of 600 Oe, which was 5 times higher than that of the (TT) mode, when operated in low magnetic bias ranges. These results coincide with our analytical predictions. In addition, a linear coupling between the measured voltage and \( H_{ac} \) for (LT) laminate was observed for various magnetic biases between 0 and 400 Oe. Accordingly, magnetoelectric (LT) laminates have much promise for magnetic field and electric current sensors.