AIP | Applied Physics Letters

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Citation: Applied Physics Letters **85**, 5305 (2004); doi: 10.1063/1.1829159

View online: http://dx.doi.org/10.1063/1.1829159

View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/85/22?ver=pdfcov

Published by the AIP Publishing

APPLIED PHYSICS LETTERS VOLUME 85, NUMBER 22 29 NOVEMBER 2004

A longitudinal-longitudinal mode TERFENOL-D/$Pb(Mg_{1/3}Nb_{2/3})O_3$–$PbTiO_3$ laminate composite

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(Received 26 April 2004; accepted 5 October 2004)

We have found that laminates of longitudinally magnetized magnetostrictive TERFENOL-D layers and a longitudinally poled piezoelectric $Pb(Mg_{1/3}Nb_{2/3})O_3$-$PbTiO_3$ crystal have a giant magnetoelectric voltage coefficient of >430 mV/Oe under a low magnetic bias. Under a resonant drive operation at a frequency of 82.1 kHz, the magnetoelectric coefficient was dramatically increased to ~18.5 V/Oe. In addition, an ultrahigh magnetic field sensitivity of $<10^{-11}$ T has been observed. © *2004 American Institute of Physics*. [DOI: 10.1063/1.1829159]

Magnetoelectric (ME) effects have been reported in single phase materials,[1–3] and piezoelectric/magnetostrictive composite materials.[4–19] Most previous investigations have focused on laminate composites whose piezoelectric/magnetostrictive layers were polarized/magnetized in their thickness/transverse directions (or T-T mode), respectively. However, the ME coefficient of the T-T laminates is quite small, except under high magnetic bias. Recently, we have shown that a piezoelectric/magnetostrictive laminate operated in a longitudinally magnetized/ transversely poled (L-T) mode has a magnetoelectric voltage coefficient of 110 mV/Oe. Our L-T laminate exhibited an ultrahigh magnetic field sensitivity of $\sim 10^{-11}$ T.[17] In this letter, we demonstrate that a laminate composite consisting of longitudinally magnetized magnetostrictive TERFENOL-D layers and a longitudinally-poled piezoelectric $Pb(Mg_{1/3}Nb_{2/3})O_3$–$PbTiO_3$ crystal (or L-L mode) has even a higher ME voltage coefficient.

Two prototype L-L laminates were made by sandwiching one ⟨001⟩-oriented $0.7Pb(Mg_{1/3}Nb_{2/3})O_3$-$0.3PbTiO_3$ (PMN-PT) piezoelectric single crystal layer between two TERFENOL-D ($Tb_{1-x}Dy_xFe_{2-y}$) layers, as shown in Fig. 1. The length and width of each layer for the two prototypes was 12.7 and 6 mm, respectively. The thicknesses of laminates were 4.8 mm for prototype 1, and 3.0 mm for prototype 2. This configuration consists of longitudinally magnetized TERFENOL-D and longitudinally-poled piezoelectric layers, i.e., a (L-L) laminate composite. This composite has an optimum bi-effect, as PMN-PT has the strongest piezoelectric effect among various piezoelectric materials,[19] and TERFENOL-D has the strongest magnetostrictive effect among various magnetostrictive materials.[20] Furthermore, this long type longitudinally magnetized and longitudinally poled configuration favors the maximum combination of magnetostrictive and piezoelectric effects, as both the longitudinal strain and magnetoelastic coupling in TERFENOL-D, and the longitudinal piezoelectric voltage coefficient and electromechanical coupling in PMN-PT are almost 2× higher than the transverse ones.

The voltages induced across the two ends of the PMN-PT layer in the L-L laminates were measured for various dc magnetic biases (H_{dc}) and ac magnetic drives (H_{ac}) at a drive frequency of $f = 1$ kHz, using a lock-in amplifier method. A pair of Helmholtz coils was used to generate a small H_{ac}, via an input current I_{coil}. The ME voltage coefficients $\partial V_{ME}^{L,L}/\partial H$ of the L-L laminates were found to be strongly dependent on direct current magnetic field bias H_{dc}. This reflects the bias dependence of the pseudo-piezomagnetic coefficient of TERFENOL-D.[20] Figure 2 shows ME voltage coefficients for L-L mode as a function of H_{dc}. As a comparison, the ME voltage coefficients for L-T mode is also illustrated. The maximum ME voltage coefficient using a drive of $H_{ac} = 1$ Oe (amplitude) was found to be ~240 mV(peak)/Oe(amplitude) for prototype 1, and ~430 mV(peak)/Oe(amplitude) for prototype 2, for $550 < H_{dc} < 800$ Oe. This is 2–4× higher than our recent results for (L-T) laminates[17] and 10–20× higher than previous two-phase laminate composites (see Ref. 21). This showed that L-L mode has an optimum operation mode, and also an optimum configuration (No. 2) relative to other laminates. Please note that we have used the ME voltage coefficient (in units of mV/Oe), rather than the ME field coefficient (in units of mV/cm Oe). The representation $\partial V_{ME}^{L,L}/\partial H$ was chosen because in use as a magnetic sensor it is a direct measure of the physical quantity being detected. However, because the length of the piezoelectric plate is much bigger than its thickness, the ME field coefficient of this laminate is small (<1 V/cmOe).

The induced ME voltage of the L-L laminate was found to be a linear function of H_{ac}, as shown in Fig. 3. 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^{-11} T (or 10^{-7} Oe) to $\sim 10^{-3}$ T (or 10 Oe). These measurements were performed at ambient conditions, without magnetic shielding. If the equivalent environmental noise can be lowered to less than 1 nV, then the L-L laminate would have the potential to detect a low-level ac magnetic field of 10^{-12} T (or correspondingly 10^{-8} Oe). These results demonstrate that

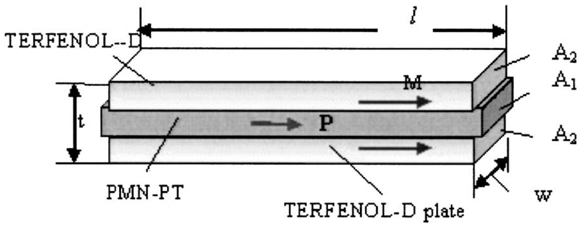

FIG. 1. L-L mode piezomagnetic/piezoelectric laminate composite.

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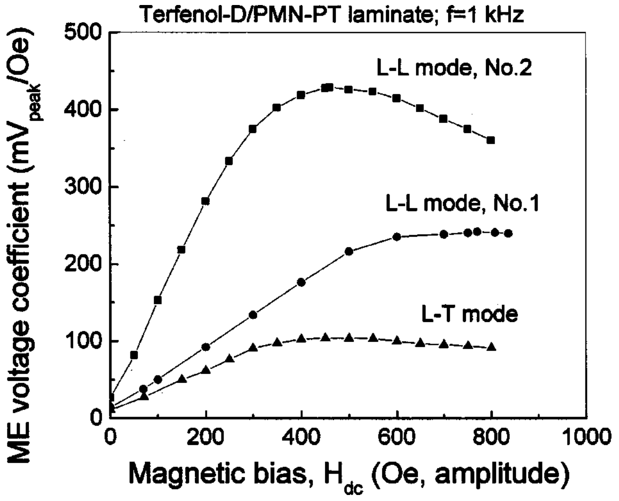

FIG. 2. ME voltage coefficient as a function of dc magnetic bias H_{dc}.

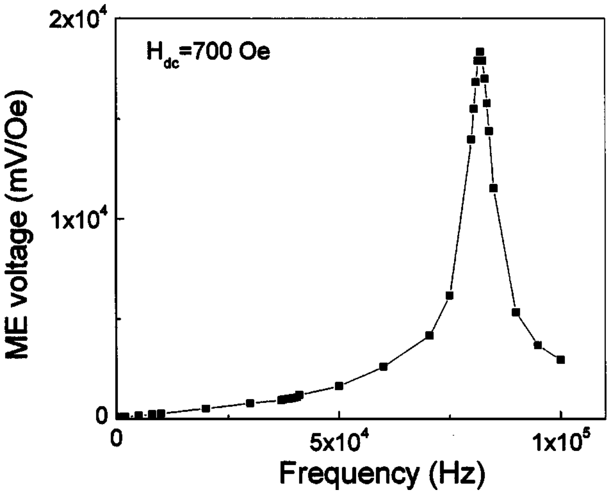

FIG. 4. Resonance ME response.

the L-L laminate has an ultrahigh sensitivity to small magnetic field variations.

Our experimental results show that L-L laminates have a near flat low-frequency ME response over the bandwidth of millihertz to kilohertz, using a $H_{ac}=1$ Oe. However, at higher frequencies, the induced ME voltage is dramatically increased with increasing frequency. Near its resonance frequency of ~82.1 kHz, the ME response of the L-L laminate reached a maximum value of 18.5 V/Oe, which is almost 43 times higher than that in the low-frequency range (see Fig. 4).

We also note that our L-L laminate has an almost linear dc magnetic field bias response in the range of $0 < H_{dc} < 300$ Oe, as can be seen in Fig. 2. Accordingly, this offers the possibility to use the L-L laminate for low-level dc magnetic field detection. Recently, we have found it feasible to detect a small dc magnetic field signal of 5×10^{-6} T, and we are currently investigating this in more detail.

Clearly, the L-L laminate has a very high ME voltage coefficient and an extremely high magnetic field sensitivity at low H_{dc} in low-frequency range. This is a significant achievement for a totally passive (~0 μW power consumed) small, inexpensive, simple, and compact material operated at room temperature, which in addition has a voltage directly proportional to the field. It offers tremendous opportunities in sensitive low-level magnetic field and/or electric current sensors.

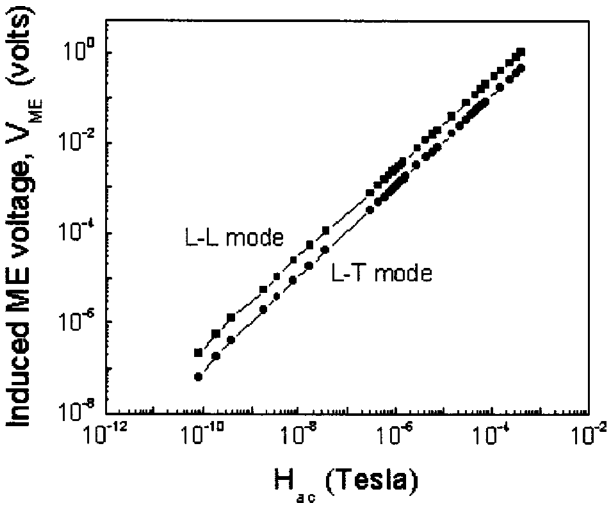

FIG. 3. Induced ME voltage at $H_{dc}=700$ Oe and $f=1$ kHz as a function of magnetic field over the range of $10^{-11} < H_{ac} < 10^{-3}$ T.

In summary, laminates of TERFENOL-D and ⟨001⟩-oriented PMN-PT crystals operated in a L-L mode have been found to have (i) a giant ME voltage coefficient of >430 mVpeak/Oe in low-frequency range at low H_{dc}, (ii) a dramatic increase (18.46 V/Oe) in ME response at its resonance frequency, and (iii) an ultrahigh magnetic field sensitivity.

This research was supported by the Office of Naval Research under Grant Nos. N000140210340, N000140210126, and MURI N000140110761.

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