Magnetoelectric effect in laminate composite of magnets/$0.7Pb(Mg_{1/3}Nb_{2/3})O_3-0.3PbTiO_3$ single crystal

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A magnetoelectric (ME) laminate composite was fabricated by sandwiching one $0.7Pb(Mg_{1/3}Nb_{2/3})O_3-0.3PbTiO_3$ (PMN-PT) piezoelectric single crystal layer between two NdFeB magnet layers along the thickness direction. The high ME effect was obtained by the product of the magnetic attractive-repellent effect in the magnet layers and the piezoelectric effect in the piezoelectric layer. The magnetoelectric voltage coefficient of the composite was measured to be ~12.5 mV/cm Oe with a flat frequency response in the range of 0.1–20 kHz. The induced ME voltage showed an excellent linear relationship to the applied ac magnetic field with field amplitude varying from 10^{-3} to 10 Oe. Other advantages included low heat generation, no bias magnetic field required, and high scale-down capability. These made the composite to be a promising ME material for realizing high-performance, small-size, and low-cost magnetic sensors. [DOI: 10.1063/1.2191948]

The magnetoelectric (ME) effect is an electric polarization response of a material to an applied magnetic field.[1] This effect has been a remarkable research topic in recent years due to its important application in magnetic sensors with high transduction efficiency between magnetic and electric energies. Hereinto, magnetostrictive/piezoelectric laminate composites, especially laminates of Terfenol-D alloy and $0.7Pb(Mg_{1/3}Nb_{2/3})O_3-0.3PbTiO_3$ (PMN-PT) single crystal, have shown enhanced ME effect compared to single-phase and other multiphase ME materials.[2–4] While extended studies concerning the ME effect in Terfenol-D/PMN-PT laminate composites have been published,[5,6] focuses were mainly put on obtaining good coupling between the two laminate phases. It is known that monolithic Terfenol-D suffers intrinsically from high cost, high eddy current-induced bandwidth limitation to a few kilohertz, and the need of an external dc bias magnetic field to maximize its magnetostrictive response.[7,8] Thus, configurations of magnetic sensors based on such laminate composites are generally complicated and not amenable to the miniaturization.

In this work, we fabricated a ME laminate composite by sandwiching one PMN-PT piezoelectric single crystal layer between two NdFeB magnet layers along the thickness direction. Figure 1 illustrates the geometry and working principle of the proposed composite. The north (N) and south (S) poles of the magnet layers are arranged in such a way that their faces are normal to the thickness direction. The polarization direction of the piezoelectric layer is also in the thickness direction. The NdFeB magnet layers were commercially supplied in the form of a plate with 10.6 mm long, 4 mm wide, and 1 mm thickness. They were galvanized using nickel, and their magnetic pole strength (q_m) is known to be 14.7 μWb. The PMN-PT piezoelectric layer was grown in-house by a modified Bridgman technique.[9] It has the same dimensions as the magnet layers, and its thickness is oriented in the ⟨001⟩ direction. After being electroded with silver and polarized along its thickness direction in a silicone oil bath, the relative dielectric constant ($\varepsilon_{33}^T/\varepsilon_0$) of the piezoelectric layer was evaluated to be 7990 at 1 kHz using an HP 4194A impedance analyzer. The piezoelectric charge coefficient (d_{33}) was measured to be 1700 pC/N using a quasistatic Berlincourt d_{33} meter. The piezoelectric voltage coefficient ($g_{33}=d_{33}/\varepsilon_{33}^T$) was determined to be 24 mV m/N. The PMN-PT piezoelectric layer was bonded between the two NdFeB magnet layers using a silver-loaded epoxy (Applied Products E-Solder 3021) to form the ME laminate composite.

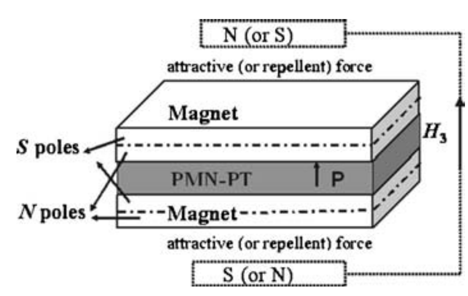

FIG. 1. Schematic diagram of NdFeB magnets/PMN-PT crystal laminate composite. The externally applied ac magnetic field (H_3) can exert the attractive or repellent forces on the two magnet layers simultaneously.

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The working principle of the proposed ME laminate composite is as follows (Fig. 1). Applying an external ac magnetic field (H_3) along the thickness direction of the composite exerts the attractive or repellent forces on the two magnet layers simultaneously. These magnetic forces thus act on the sandwiched piezoelectric layer, causing it to produce piezoelectric voltages (or charges). It is important to note that this magnetic force-induced ME effect is essentially different from the magnetostrictive strain-induced ME effect used in operating traditional magnetostrictive/piezoelectric laminate composites.[3–6]

According to the Coulomb law of magnetostatics, the magnitude of attractive or repellant force applied on each magnet layer F_3 is

$$F_3 = H_3 \cdot q_m, \tag{1}$$

where H_3 is the externally applied ac magnetic field and q_m is the magnetic pole strength of magnet layer. For a certain magnet, q_m is a constant. In our design, q_m is known to be 14.7 μWb. As our piezoelectric layer has been thickness polarized, the piezoelectric constitutive equations for the layer can be expressed as[10]

$$S_3 = s_{33}^D T_3 + g_{33} D_3, \tag{2a}$$

$$E_3 = - g_{33} T_3 + \beta_{33}^T D_3, \tag{2b}$$

where E_3 and D_3 are the electric field and electric displacement along the thickness direction, respectively; T_3 and S_3 are the stress and strain along the thickness direction, respectively; s_{33}^D is the elastic compliance coefficient at constant electric displacement; g_{33} is the piezoelectric voltage constant; and β_{33}^T is the dielectric impermittivity at constant stress. The stress in the piezoelectric layer T_3 as a result of transferring F_3 from the top and bottom magnet layers can be expressed as

$$T_3 = \frac{2F_3}{A}, \tag{3}$$

where A is the interfacial area of the magnet layers and the piezoelectric layer. Putting Eqs. (1) and (3) into Eq. (2), the magnetoelectric voltage coefficient (α_E) of the composite is obtained as follows:

$$\alpha_E = \frac{dE_3}{dH_3} = -2\frac{q_m g_{33}}{A}. \tag{4}$$

From Eq. (4), it is clear that α_E depends on q_m of the magnet layers, g_{33} of the piezoelectric layer, and the interfacial area A. Substituting the corresponding material parameters of NdFeB magnet layers and PMN-PT piezoelectric layer into Eq. (4), α_E of the proposed composite is predicted to be 13.2 mV/cm Oe.

Figure 2 shows the ME voltage (V_3) induced by an applied ac magnetic field (H_3) of 1 Oe peak as a function of time at the frequency of 1 kHz. Agreed with Eq. (4), V_3 and H_3 are of opposite phase. Besides, V_3 follows steadily H_3 and has the maximum amplitude of ~1.25 mV when H_3 peaks at 1 Oe.

Figure 3 plots the induced ME voltage (V_3) as a function of applied ac magnetic field (H_3) in the field range of 10^{-3}–10 Oe under a drive frequency of 1 kHz. It is seen that V_3 has a good linear relationship with H_3 over this magnetic field range. The result also demonstrates that the composite is very sensitive to H_3 variations even at a small H_3 of 10^{-3} Oe. A higher sensitivity of 10^{-5}–10^{-6} Oe could be obtained if shielding of magnetic noises could be adopted and composite fabrication techniques could be improved.[11] From the slope of the plot, α_E is determined to be ~12.5 mV/cm Oe at 1 kHz. This measured α_E coincides reasonably well with the predicted value of 13.2 mV/cm Oe. Nevertheless, α_E (~12.5 mV/cm Oe) of this composite is small in comparison with that (~320 mV/cm Oe at an optimal bias magnetic field of 500 Oe) of Terfenol-D/PMN-PT laminate composites with transverse-transverse configuration at the same frequency.[3]

Figure 4 shows the frequency dependence of the ME voltage coefficient (α_E) at an applied ac magnetic field (H_3) of 1 Oe peak. It is obvious that α_E has a flat response in measured frequency range from 0.1 to 20 kHz. Owing to the insulation of the magnet layers, not like the low-resistive Terfenol-D alloy (resistivity ~0.6 $\mu\Omega$ m),[7,8] the eddy current-induced thermal losses in the magnet layers are neglectable even the composite is exposed to a high drive frequency of 20 kHz. Consequently, while possessing a reduced α_E, the current composite can work stably for frequencies up to and, great probably, beyond 20 kHz without subject to redundant thermal-induced depolarization of its piezoelectric layer. This permits promising applications of the composite in solid-state magnetic sensors. Another important advantage of using the composite is that no external dc bias magnetic field is needed as compared with the traditional magnetostrictive/piezoelectric laminate composites.[3–6] This

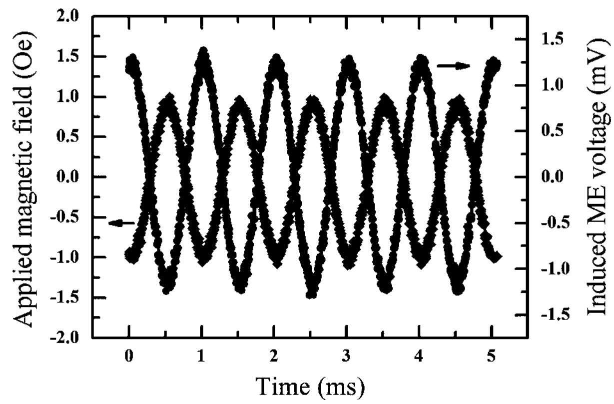

FIG. 2. ME voltage induced by an applied ac magnetic field of 1 Oe peak as a function of time at the frequency of 1 kHz.

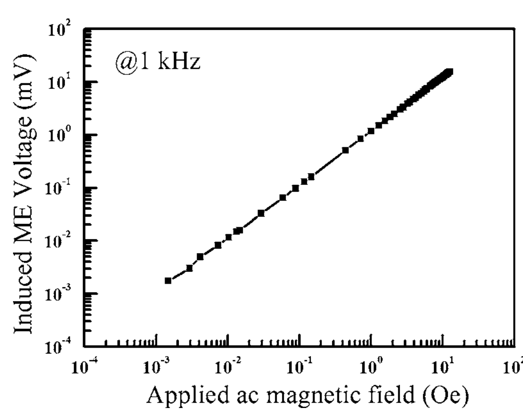

FIG. 3. Induced ME voltage as a function of applied ac magnetic field in the field range of 10^{-3}–10 Oe under a drive frequency of 1 kHz.

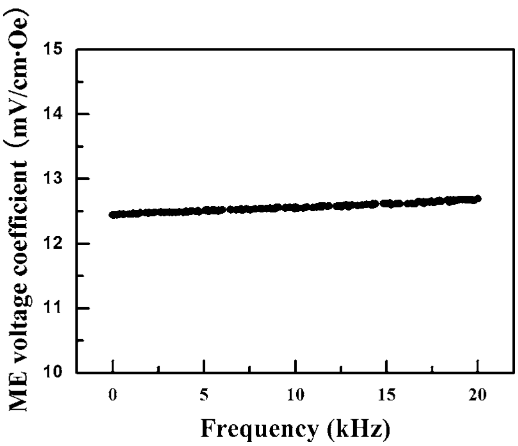

FIG. 4. Frequency dependence of ME voltage coefficient at an applied ac magnetic field of 1 Oe peak.

is favorable for the miniaturization and cost saving of practical devices.[10] In addition, magnetic sensors based on the ME effect are field sensors; they are different from search coils or superconducting quantum interface devices (SQUIDs), which are flux sensors. This suggests that a constant sensitivity may be provided as the sensor size is scaled down, giving great potential of using this composite in microelectromechanical system (MEMS) scale system.

In summary, we have fabricated a ME laminate composite by sandwiching a PMN-PT piezoelectric single crystal layer between two NdFeB magnet layers in the thickness direction. The experimental results have demonstrated that this composite possess a high α_E of ~ 12.5 mV/cm Oe, a flat frequency response from 0.1 to 20 kHz, an excellent linear relationship between V_3 and H_3 in the field range of $10^{-3}-10$ Oe, and a high sensitivity to small H_3 of $<10^{-3}$ Oe. Comparing with the conventional magnetostrictive/piezoelectric laminate composites, the current composite does not need any bias magnetic field, can be made much smaller in size, and is more effective in cost. The excellent ME performance together with the inherent benefits make this composite to be a promising and practicable ME material for magnetic sensors.

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[1] L. D. Landau and E. Lifshitz, *Electrodynamics of Continuous Media* (Pergamon, Oxford, 1960).
[2] V. J. Folen, G. T. Rado, and E. W. Stalder, Phys. Rev. Lett. **6**, 607 (1961).
[3] S. Dong, F. Bai, J. F. Li, and D. Viehland, IEEE Trans. Ultrason. Ferroelectr. Freq. Control **51**, 794 (2004).
[4] J. Ryu, A. V. Carazo, K. Uchino, and H. Kim, J. Electroceram. **7**, 24 (2001).
[5] S. Dong, J. F. Li, and D. Viehland, Appl. Phys. Lett. **85**, 5305 (2004).
[6] S. Dong, J. Y. Zhai, Z. P. Xing, J. F. Li, and D. Viehland, Appl. Phys. Lett. **86**, 102901 (2005).
[7] S. W. Or, T. Li, and H. L. W. Chan, J. Appl. Phys. **97**, 10M308 (2005).
[8] G. Engdahl, *Magnetostrictive Materials Handbook* (Academic, New York, 2000).
[9] H. S. Luo, G. S. Shen, P. C. Wang, X.-H. Le, and Z.-W. Yin, Int. J. Inorg. Mater. **12**, 768 (1997).
[10] T. Ikeda, *Fundamentals of Piezoelectricity* (Oxford University Press, Oxford, 1990).
[11] S. Dong, J. F. Li, and D. Viehland, Appl. Phys. Lett. **85**, 2307 (2004).