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Converse magnetoelectric effect in laminated composites of PMN–PT single crystal and Terfenol-D alloy

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We have found experimentally and theoretically that laminated composites comprising one layer of length-magnetized $Tb_{0.3}Dy_{0.7}Fe_{1.92}$ (Terfenol-D) magnetostrictive alloy sandwiched between two layers of thickness-polarized, electro-parallel-connected $0.7Pb(Mg_{1/3}Nb_{2/3})O_3-0.3PbTiO_3$ (PMN–PT) piezoelectric single crystal have a large converse magnetoelectric effect characterized by a large magnetic induction in response to an applied ac voltage. The reported converse magnetoelectric effect originates from the product of the converse piezoelectric effect in the PMN–PT layers and the converse magnetostrictive effect in the Terfenol-D layer. Large converse magnetoelectric coefficient in excess of 105 mG/V is obtained in the composites at a low magnetic bias field of 170 Oe. The measured magnetic induction has an excellent linear relationship to the applied ac voltage with amplitude varying from 50 to 160 V. These made the composites to be a promising material for direct realization of core-free magnetic flux control devices. © *2006 American Institute of Physics*. [DOI: 10.1063/1.2212054]

The magnetoelectric (ME) effect is an electric polarization response of a material to an applied magnetic field, while the converse magnetoelectric effect (CME) is a magnetization response of a material to an applied electric field.[1] Single-phase ME (and CME) materials, such as Cr_2O_3, are not recognized as practically viable materials for device applications due to their low Curie temperatures below the room temperature and weak coupling between polarization and magnetization.[2] In recent years, it has been found that laminated composites of piezoelectric and magnetostrictive materials possess superior ME performance owing to their giant product effect of the piezoelectric and magnetostrictive effects.[3] Among them, the ones formed by relaxor-based $(1-x)Pb(Mg_{1/3}Nb_{2/3})O_3-xPbTiO_3$ piezoelectric single crystals and rare-earth-based $Tb_xDy_{1-x}Fe_{2-y}$ magnetostrictive alloys have demonstrated remarkably enhanced ME effect for fabricating high-performance magnetic field devices.[4–6]

To date, seldom report has been made on the CME effect in the composites, especially when the effect is to be quantitatively measured by the CME coefficient (α_B) defined by an induced magnetic induction in response to an applied ac voltage (dB/dV). Besides, it is both physically interesting and technologically important to find a complementary material to electromagnets so that core-free magnetic flux control devices can be created to alleviate the Joule heating and bandwidth limitation problems intrinsic in traditional electromagnet-based control devices. In this work, we reported a large CME effect in laminated composites based on sandwiching one $Tb_{0.3}Dy_{0.7}Fe_{1.92}$ (Terfenol-D) magnetostrictive alloy plate between two $0.7Pb(Mg_{1/3}Nb_{2/3})O_3-0.3PbTiO_3$ (PMN–PT) piezoelectric single-crystal plates along the thickness direction.

Figure 1 illustrates the geometry and working principle of the proposed laminated composite. The PMN–PT plates, of dimensions 12 mm long, 6 mm wide, and 1 mm thick, and with their ⟨001⟩ and ⟨011⟩ crystallographic axes oriented in the length and thickness directions, respectively, were grown in-house using a modified Bridgman technique.[7] It was shown in our previous work that PMN–PT plates, when grown with these specific crystallographic orientations and polarized using the ⟨001⟩ axis in the thickness direction, possess ultrahigh transverse piezoelectric response with deformations along the length direction when excited electrically in the thickness direction.[7] The piezoelectric coefficient ($d_{31,p}$) and elastic compliance coefficient (s_{11}^E) of the PMN–PT plates were measured to be −1300 pC/N and 126 pm^2/N, respectively. The Terfenol-D plate was commercially supplied with the dimensions the same as the PMN–PT plates (Baotou Rare Earth Research Institute, China). It had

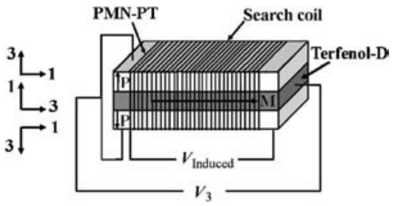

FIG. 1. Schematic diagram of our laminated composite. The arrows *P* and *M* denote the polarization and magnetization directions, respectively.

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the highly magnetostrictive [112] crystallographic axis along the length direction and thus magnetization was relatively easy in this direction. The piezomagnetic coefficient ($d_{33,m}$) and elastic compliance coefficient (s_{33}^H) of the Terfenol-D plate were known to be 5.2 nm/A and 87 pm^2/N, respectively. To construct such a laminated composite (Fig. 1), a Terfenol-D plate was bonded between two PMN–PT plates arranged in reverse polarization directions using a silver-loaded epoxy (Applied Products E-Solder 3021), and the two PMN–PT plates were connected electrically in parallel to form the input terminals.

The working principle of the proposed composite is as follows (Fig. 1). First, applying an ac voltage (V_3) to the input terminals of the composite produces two equimagnitude, opposite-sign ac electric fields (E_3) across the thickness of the PMN–PT plates, leading to transverse piezoelectric strains in the PMN–PT plates due to the converse piezoelectric effect; second, these piezoelectrically induced strains result in mechanical stresses in the sandwiched Terfenol-D plate, causing it to produce magnetic induction (B_3) [to be measured by time integral of the induced voltage (V_{Induced}) in a search coil] along the length of the composite as a result of the converse magnetostrictive effect. It is essential to note that this converse piezoelectric/converse magnetostrictive induced CME effect is different from the direct magnetostrictive/direct piezoelectric induced direct ME effect in traditional ME laminated composites.[3–6]

To enable quasistatic modeling of the composite in Fig. 1, the composite is simplified as an in-plane, two-dimensional object with the length much larger than its width. Accordingly, only the geometric information involved in the length and thickness directions are considered. Both the polarization (P) and magnetization (M) directions are arranged to coincide with the 3-direction of the coordinate system. As our PMN–PT plates have been thickness-polarized and deformations are essentially along their length, the piezoelectric constitutive equations are[7]

$$S_{1,p} = s_{11}^E T_{1,p} + d_{31,p} E_3, \tag{1a}$$

$$D_3 = d_{31,p} T_{1,p} + \varepsilon_{33}^T E_3, \tag{1b}$$

where E_3 and D_3 are the electric field and electric displacement along the thickness direction, respectively, $T_{1,p}$ and $S_{1,p}$ are the stress and strain along the length direction, respectively, s_{11}^E is the elastic compliance coefficient at constant electric field strength, $d_{31,p}$ is the piezoelectric coefficient, and ε_{33}^T is the dielectric permittivity at constant stress. For the Terfenol-D plate, since it has the highly magnetostrictive axis (or the easy magnetization axis) oriented along its length and deformations are essentially in the length direction, the magnetostrictive constitutive equations are[8]

$$S_{3,m} = s_{33}^H T_{3,m} + d_{33,m} H_3, \tag{2a}$$

$$B_3 = d_{33,m} T_{3,m} + \mu_{33}^T H_3, \tag{2b}$$

where B_3 and H_3 are the magnetic induction and magnetic field strength along the length direction, respectively, $T_{3,m}$ and $S_{3,m}$ are the stress and strain along the length direction, respectively, s_{33}^H is the elastic compliance coefficient at constant magnetic field strength, $d_{33,m}$ is the piezomagnetic coefficient, and μ_{33}^T is the magnetic permeability at constant stress. The boundary conditions for the composite are

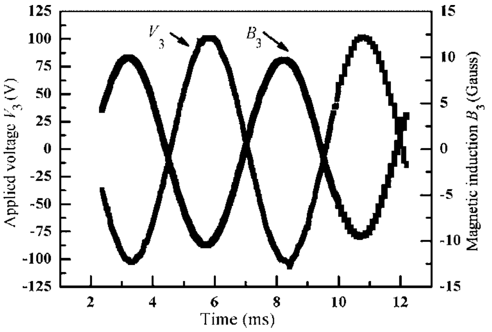

FIG. 2. Waveforms of the measured magnetic induction (B_3) due to an applied ac voltage (V_3) of 100 V peak at the frequency of 200 Hz and with the magnetic bias field (H_{Bias}) of 170 Oe.

$$T_{1,p} 2t_p = - T_{3,m} t_m, \tag{3}$$

$$S_{1,p} = S_{3,m}, \tag{4}$$

where t_p and t_m are the thicknesses of the PMN–PT and Terfenol-D plates, respectively. Combining Eqs. (1a), (2a), (2b), (3), and (4), and setting $H_3=0$ in Eqs. (2a) and (2b), the CME coefficient (α_B) of the composite is expressed as

$$\alpha_B = \left| \frac{dB_3}{dV_3} \right| = \left| \frac{2d_{31,p} d_{33,m}}{s_{11}^E t_m + 2 s_{33}^H t_p} \right| . \tag{5}$$

From Eq. (5), it is clear that α_B of the composite depends on $d_{31,p}$, s_{11}^E, and t_p of the PMN–PT plates as well as $d_{33,m}$, s_{33}^H, and t_m of the Terfenol-D plate. Substituting the corresponding material and geometric parameters into Eq. (5), α_B of the proposed composite is predicted to be 450 mG/V.

The CME effect in the fabricated composites was characterized using an in-house automated measurement system.[9] To acquire the induced magnetic induction (B_3), a search coil was wrapped around a composite sample, and the whole composite-search coil assembly was placed between the pole gap of an electromagnet (Myltem PEM-8005K). By energizing the electromagnet with a dc current supply (Sorensen DHP200-15), a magnetic bias field (H_{Bias}) was generated in the pole gap and was measured with a Hall-effect probe connected to a Gaussmeter (F. W. Bell 7030). A dynamic signal analyzer (Ono Sokki CF5220) connected to a voltage amplifier (NF-HAS 4025) was employed to provide an applied ac voltage (V_3) for the PMN–PT plates of the sample. The change of magnetic flux due to the applied V_3 led to an induced voltage (V_{Induced}) in the search coil (Fig. 1). By integrating this V_{Induced} with respect to time using an integrating fluxmeter (Walker LDJ MF-10D), the corresponding B_3 induced in the sample was measured by the dynamic signal analyzer. From the slope of B_3-V_3 plot, α_B was determined. By plotting α_B as a function of H_{Bias}, the H_{Bias} required to optimally operate the sample was obtained.

Figure 2 shows the waveforms of the measured magnetic induction (B_3) due to an applied ac voltage (V_3) of 100 V peak at the frequency of 200 Hz and with the magnetic bias field (H_{Bias}) of 170 Oe. Agreed with Eq. (5), B_3 and V_3 are of opposite phase, since the piezoelectric coefficient $d_{31,p}$ carries a negative sign. Moreover, B_3 follows steadily V_3 and has the maximum amplitude of 10.5 G when V_3 peaks at 100 V. This demonstrates the ability of stable signal conversion from V_3 to B_3 in our composite sample.

Figure 3 plots the measured magnetic induction (B_3) as a function of applied ac voltage (V_3) for various magnetic bias

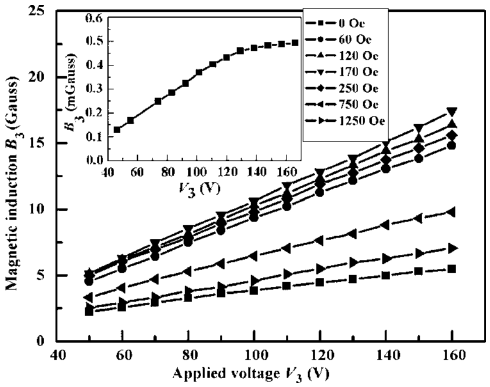

FIG. 3. Measured magnetic induction (B_3) as a function of applied ac voltage (V_3) for various magnetic bias fields (H_{Bias}) at the frequency of 200 Hz. The inset shows the result of a similar composite sample based on unpolarized PMN–PT single crystal plates.

fields (H_{Bias}) at the frequency of 200 Hz. It is seen that B_3 has good linear responses to V_3 in the entire V_3 range of 50–160 V for all H_{Bias}. Specifically, the greatest B_3-V_3 response occurs at $H_{\mathrm{Bias}}=170$ Oe. From the slope of the B_3-V_3 plot at $H_{\mathrm{Bias}}=170$ Oe, the converse magnetoelectric coefficient (α_B) is determined to be 105 mG/V. A higher α_B could be obtained if shielding of magnetic noises could be adopted and composite fabrication techniques could be improved. Nevertheless, this measured maximum α_B agrees with the predicted value of 450 mG/V based on Eq. (5).

In order to examine the effect of ac coupling that may directly run through Terfenol-D itself during the measurement of B_3, a similar composite sample based on unpolarized PMN–PT single crystal plates instead of the polarized ones was also fabricated and characterized for comparison with the current composite sample. As shown in the inset of Fig. 3, while the ac coupling effect (characterized by B_3) exhibits an increasing trend with increasing V_3, B_3 remains at extremely low values of <0.5 mG even at an increased V_3 of 160 V. Comparing to the measured B_3 of 5.4–17.4 G in the proposed composite sample at the same V_3 (the main plot of Fig. 3), this ac coupling effect is indeed negligible ($<0.1\%$). It is also found that the measured B_3 due to the effect of ac coupling is independent of H_{Bias}. Details can be found from the inset of Fig. 4.

Figure 4 shows the dependence of CME coefficient (α_B) on magnetic bias field (H_{Bias}) at the frequency of 200 Hz. The values of α_B are obtained from the slopes of the B_3-V_3 plots in Fig. 3 at various H_{Bias}. It is obvious that α_B increases initially up to 105 mG/V at $H_{\mathrm{Bias}}=170$ Oe and then decreases with increasing H_{Bias}. Physically, the change in α_B with increasing H_{Bias} can be explained by the H_{Bias}-induced motion of the available non-180° domain walls in the Terfenol-D plate.[10] That is, as H_{Bias} is increased near 170 Oe, the compliance associated with increased deformation contribution from this non-180° domain-wall motion is maximized, resulting in a maximum in strain and hence a maximum in both $d_{33,m}$ and α_B [Eq. (5)]. Beyond this optimal H_{Bias}, constraining of non-180° domain-wall motion due to interaction with H_{Bias} gives rise to a decrease in strain and hence a decrease in both $d_{33,m}$ and α_B. It is noted that the deformation contribution from the motion of 180° domain walls is insignificant in Terfenol-D as it produces changes in magnetization without accompanying strain.[10] Therefore, in order to obtain the largest B_3, the optimal H_{Bias} of 170 Oe should be applied to operate the composite. Importantly, this H_{Bias} is relatively low and easily to be obtained in practical applications by using permanent magnets.

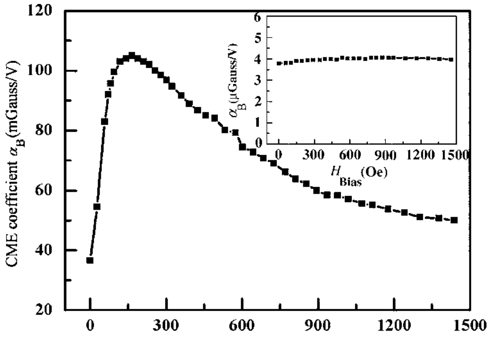

FIG. 4. Dependence of CME coefficient (α_B) on magnetic bias field (H_{Bias}) at the frequency of 200 Hz. The inset shows the result of a similar composite sample based on unpolarized PMN–PT single crystal plates.

The inset of Fig. 4 indicates the effect of ac coupling on the α_B measurement as a function of H_{Bias}. It is evident that the measured α_B stays at a very low average value of 4 μG/V in the whole H_{Bias} range, and the ac coupling effect can thus be neglected ($<0.1\%$).

In summary, a large CME effect has been found experimentally and theoretically in laminated composites consisting of one Terfenol-D alloy plate bonded between two electro-parallel-connected PMN–PT single-crystal plates in the thickness direction. The results have demonstrated that these composites possess a high α_B of 105 mG/V at a very low H_{Bias} of 170 Oe, an excellent linear relationship between B_3 and V_3 in the V_3 range of 50–160 V and for H_{Bias} varying from 0 to 1400 Oe, and a negligible ac coupling effect ran directly through Terfenol-D. Comparing to the conventional electromagnet-based magnetic flux control devices, the current composites do not need any magnetic coil, and thus have reduced Joule heating losses, improved operational bandwidths, and better scale-down capability.

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