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Converse magnetoelectric effect in laminated composites of PMN-PT single crystal and Terfenol-D alloy
Giant magnetoelectric effect in mechanically clamped heterostructures of magnetostrictive alloy and piezoelectric crystal-alloy cymbal

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We experimentally and theoretically report a giant magnetoelectric (ME) effect in a heterostructure made by clamping a bar-shaped magnetostrictive Tb0.3Dy0.7Fe1.92 alloy actuator and a cymbal-type piezoelectric 0.7Pb(Mg1/3Nb2/3)O3–0.3PbTiO3 (PMN–PT) single crystal-brass alloy transducer in a brass alloy frame. The reported ME effect originates from the stress-mediated product of the magnetostrictive effect in the Terfenol-D bar actuator and the mechanically transformed/amplified piezoelectric effect in the PMN PT-brass cymbal transducer under mechanically clamped conditions. The heterostructure exhibits a giant ME voltage coefficient ($\alpha_V$) of 440 mV/Oe at an optimal magnetic bias field of 400 Oe besides showing good linearity between the induced ME voltage and the applied ac magnetic field in the field range of 10−3–10 Oe. This $\alpha_V$ is larger than that of conventional Terfenol-D/PMN-PT laminated composites.


The magnetoelectric (ME) coupling between magnetization and polarization in multiferroic materials has attracted much attention in recent years, not only because of the undiscovered science underpinning this effect, but also due to the tremendous application potential for the associated materials and the resulting devices.1,2 While the ME effect was proposed by Pierre Curie as early as in 1894,3 it was not experimentally demonstrated until the first observation in an antiferromagnetic Cr2O3 single crystal in 1961.4 The effect in single-phase materials is usually very weak and can only be measured at temperatures far below room temperature. Instead of the intrinsic effect, a much stronger extrinsic effect can be obtained in two-phase or three-phase laminated composites using magnetostrictive and piezoelectric materials as the active constituent phases to impart the stress-mediated product of the magnetostrictive and piezoelectric effects to the composites.5–12 To date, it is known that laminated composites consisting of magnetostrictive Tb03Dy2,Fe1,92 (Terfenol-D) alloy and piezoelectric 0.7Pb(Mg1/3Nb2/3)O3–0.3PbTiO3 (PMN–PT) single crystal with either longitudinal-transverse (L-T) or longitudinal-longitudinal (L-L) configuration possess the highest ME voltage coefficient ($\alpha_V$) and detection sensitivity.8,10,12

In fact, the reported laminated composites are generally designed based on the direct combination of magnetostrictive and piezoelectric materials to form some classical (e.g., L-T, and L-L) configurations and are fabricated using an adhesive lamination technique, resulting in limited performance and property tailorability. By introducing mechanical transformation/amplification and clamping mechanisms into the laminated composites, it is practically viable to realize a class of composites showing enhanced ME effect with improved property tailorability. In this letter, we report a giant ME effect in a heterostructure composed of a bar-shaped magnetostrictive Terfenol-D alloy actuator and a cymbal-type piezoelectric PMN-PT single crystal-brass alloy transducer assembled in a brass alloy frame. The proposed heterostructure, which features an enhanced piezoelectric effect and an improved mechanical stress coupling between the Terfenol-D bar actuator and the PMN-PT-brass cymbal transducer, is crucial to the future development of ME composites and devices.

Figure 1 shows the schematic and geometry of the proposed ME heterostructure. A high-quality PMN-PT single crystal ingot was grown directly from the melt by a modified Bridgman technique13 and then diced to give (001)-oriented plates with dimensions of 12.8 mm length ($l_p$), 6 mm width ($w_P$), and 1 mm thickness ($t_P$). The as-prepared plates were polarized along their thickness (or z-) direction, and the polarized plates were sandwiched between two protruded rectangular brass alloy caps along the thickness direction using silver-loaded epoxy to form the cymbal-type piezoelectric PMN-PT single crystal-brass alloy transducer. The two protruded rectangular brass alloy caps, each of 3 mm top-flange length ($l_C$), 9 mm bottom-flange length ($l_{Cb}$), and 0.6 mm flange height ($t_{Cb}$), were made by die-punching a brass alloy sheet with thickness ($t_C$) of 0.3 mm and the same width ($w_C=w_P=6$ mm) as the PMN-PT plates. The bar-shaped magnetostrictive Terfenol-D alloy actuator was commercially supplied (Baotou Research Institute of Rare Earth, China) with dimensions of $12 \times 3 \times 3$ mm$^3$ and the highly magneto-
strictive [112] crystallographic axis oriented along the longitudinal (or 3-) direction. A Terfenol-D bar actuator and a PMN-PT-brass cymbal transducer were assembled and clamped in a brass alloy frame to form a ME heterostructure.

The working principle of our heterostructure (Fig. 1) is essentially based on the stress-mediated product of the magnetostrictive effect in the Terfenol-D bar actuator and the mechanically transformed/amplified piezoelectric effect in the PMN-PT-brass cymbal transducer under mechanically clamped conditions. In operation, applying an ac magnetic field (\(H_{\text{ac}}\)) along the 3-direction of the heterostructure excites the Terfenol-D bar actuator into a longitudinal motion as a result of the magnetostrictive effect in the Terfenol-D bar actuator. The longitudinal stress (\(T_3\)) produced by the Terfenol-D bar actuator, due to the improved mechanical clamping effect of the brass alloy frame, is effectively transformed and amplified by the protruded rectangular brass alloy caps of the PMN-PT-brass cymbal transducer, leading to an enhanced transverse stress (\(T_1\)) of opposite sign (to be further described in the next paragraph). The combined \(T_3\) and \(T_1\), upon acting on the central PMN-PT plate, cause it to produce increased ME voltage (\(V_{\text{ME}}\)) owing to the mechanically transformed/amplified piezoelectric effect in the PMN-PT-brass cymbal transducer.

Theoretically, the ME activity in our heterostructure can be modeled using an equivalent-circuit approach.\(^8\) The induced ME voltage (\(V_{\text{ME}}\)) across the PMN-PT plate by an applied ac magnetic field (\(H_{\text{ac}}\)) can be written as

\[
V_{\text{ME}} = \frac{d_{33}^{\text{eff}} d_{33,\text{m}} A_{33,p}}{\varepsilon_{33}^{T} [\varepsilon_{33} A_{33,p}/(1 - k_{33,p}^2)]} H_{\text{ac}},
\]

where \(A_{\text{m}}\) and \(l_{\text{m}}\) are the cross-section area and length of the Terfenol-D bar actuator, respectively; \(A_{33,p}\) and \(t_p\) are the electroded area and thickness of the PMN-PT plate, respectively; \(s_{33}\) and \(d_{33,\text{m}}\) are the elastic compliance coefficient and piezomagnetic coefficient of Terfenol-D, respectively; \(\varepsilon_{33}^{T}\), \(s_{33}\), and \(k_{33,p}\) are the dielectric permittivity, elastic compliance coefficient, and electromechanical coupling coefficient of PMN-PT, respectively; and \(d_{33}^{\text{eff}}\) is the effective piezoelectric strain coefficient of the PMN-PT-brass cymbal transducer. For the cymbal-type assembly,\(^4\) the piezoelectric \(d_{33,p}\) and \(d_{31,p}\) contributions from the central PMN-PT plate are combined and mediated by the mechanical transformation/amplification effect in the protruded rectangular brass alloy caps to provide \(d_{33}^{\text{eff}}\), giving\(^3\)

\[
d_{33}^{\text{eff}} = d_{33} - Ad_{31}, \quad A = \frac{l_{\text{cb}}(l_{\text{ch}} - l_{\text{c}})}{2(l_{\text{ch}} t_p + 2l_{\text{c}})},
\]

where \(A\) is the transformation/amplification factor depending on the geometric parameters of the PMN-PT-brass cymbal transducer (~28.125 in our case). Equations (1) and (2) provide an important insight into the physics about the enhancement of \(\alpha_{ty}(=dV_{\text{ME}}/dH_{\text{ac}})\) in our heterostructure. The typical piezoelectric parameters were determined by the quasistatic Berlincourt \(d_{31}\) meter and Agilent 4294A impedance analyzer according to the IEEE resonance method (see Table I).

The ME voltage coefficient (\(\alpha_{ty}\)) of our heterostructure was measured for various magnetic bias fields (\(H_{\text{bias}}\)) of 0–1200 Oe under an ac magnetic field (\(H_{\text{ac}}\)) of 1 Oe peak and at a frequency of 1 kHz. As shown in Fig. 2, \(\alpha_{ty}\) increases linearly over a relatively broad range of \(H_{\text{bias}}\) from 0 to 200 Oe and reaches the technical maximum value of 440 mV/0e at an optimal \(H_{\text{bias}}\) of 400 Oe, corresponding to the maximization of the magnetostrictive activity in Terfenol-D.\(^12\) This low-field linear range, to a certain extent, suggests that the current heterostructure is a good candidate for use in dc ME sensors besides the generally studied topic on the ac ME effect. The inset demonstrates the ability of the current heterostructure to operate in a time-domain capture mode. It shows the ME voltage (\(V_{\text{ME}}\) (440 mV peak)) induced by \(H_{\text{ac}}\) (1 Oe peak) as a function of time at 1 kHz frequency and 400 Oe bias. Stable signal conversion between \(V_{\text{ME}}\) and \(H_{\text{ac}}\) is evident. Moreover, \(V_{\text{ME}}\) and \(H_{\text{ac}}\) are of opposite phases, suggesting the existence of an opposition motion between the Terfenol-D bar actuator (elongation or shrinkage) and the PMN-PT-brass cymbal transducer (shrinkage or elongation).

| TABLE I. Piezoelectric parameters of PZT ceramics, (001)-oriented PMN-PT crystal, and metal caps/(001)- PMN-PT assembly. |
|------------------|------------------|------------------|------------------|------------------|
|                  | \(d_{31}\) (\(\times 10^{-12}\) C/N) | \(d_{33}\) or \(d_{33}^{\text{eff}}\) \(\times 10^{-12}\) C/N | \(k_{31}\) | \(k_{33}\) or \(k_{33}^{\text{eff}}\) | \(\varepsilon_{33}^{T} / e_0\) |
| PZT\(^8\)        | −125             | 290              | 0.35            | 0.65            | 1035             |
| (001)-PMN-PT     | −867             | 2013             | 0.59            | 0.61            | 4276             |
| (001)-PMN-PT-brass cymbal | 25 000\(^9\), 26 397\(^9\) | 0.46             |

\(^8\) Cited from APC piezoelectric ceramics.
\(^9\) Measured value after assembling.
\(^9\) Calculated value following Eq. (2).
The induced ME voltage ($V_{ME}$) was measured over a broad ac magnetic field ($H_{ac}$) range of $10^{-3} - 10$ Oe under an optimal magnetic bias field ($H_{bias}$) of 400 Oe at a frequency of 1 kHz. From Fig. 3, it is obvious that $V_{ME}$ has an excellent linear response to $H_{ac}$. The observation also indicates the high detection sensitivity nature of our heterostructure even though it is subject to an $H_{bias}$ as small as $10^{-3}$ Oe. An improved detection sensitivity of $10^{-4} - 10^{-5}$ Oe is practically viable by shielding the magnetic noises and improving the heterostructure quality. From the slope of the plot, the ME voltage coefficient $\alpha_{V}$ can also be determined, which coincides reasonably well with $\alpha_{V}$ versus $H_{bias}$, as shown in Fig. 2.

In summary, we have experimentally and theoretically reported a giant ME effect in a heterostructure comprising a Terfenol-D bar actuator and a PMN-PT-brass cymbal transducer clamped in a brass alloy frame. In comparison with the conventional ME laminated composites, the enhanced piezoelectric effect in the PMN-PT-brass cymbal transducer and the improved mechanical stress coupling between the Terfenol-D bar actuator and the PMN-PT-brass cymbal transducer have dramatically increased $\alpha_{V}$ of the heterostructure to 440 mV/Oe. This makes the heterostructure to be a promising material for the direct realization of solid-state ME devices.

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