Magnetoelectric effect in a bi-rectangular structure composed of negative/positive magnetostrictive and piezoelectric flakes

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Magnetoelectric (ME) effect has been studied in bi-rectangular structure made up of epoxy-bonded negative/positive magnetostrictive and piezoelectric flakes. The ME effect is affected by negative and positive magnetostrictive flakes. The ME voltage coefficient at resonance frequency shows a nearly constant plateau behavior with the bias magnetic field increased from 1 to 3.5 kOe. There is no interface between magnetostrictive and piezoelectric flakes required to achieve ME coupling, which provides a new choice to make ME devices.

I. INTRODUCTION

Magnetoelectric (ME) materials, which can display an induction of magnetization by an electric field or of polarization by a magnetic field, have stimulated tremendous fundamental and practical interests due to their potential applications in multifunctional devices such as sensors, actuators, and transducers.[1–4] In the past decades, ME materials have been developed in single-phase materials and multiphase bulk composites. Single-phase ME materials are not suitable for practical applications owing to their weak ME response and low working temperature.[5] Bulk ME composites, especially layered ME composites made by combining magnetostrictive and piezoelectric layers, exhibit excellent ME effect at room temperature. To enhance the ME voltage coefficient, layered ME composites, with Ni, Fe-Co alloys, or rare-earth alloys as the magnetostrictive phase and with lead zirconate titanate (PZT), lead magnesium niobate-lead titanate as the piezoelectric phase, have been made into various structures.[6–14]

The ME effect in layered ME composites is a product property of the magnetostrictive and piezoelectric effects. A magnetic field applied to the layered ME composites will induce strain in the magnetostrictive layer which is passed along to the piezoelectric layer, where it induces an electric polarization.[15] The ME coupling of layered ME composites mainly arises from the magnetic–mechanical–electric transform through the stress-mediated transfer in the interface. Therefore, the interfacial coupling is the key to the ME coupling of layered composites. However, in practice, there is no ideal interface for layered ME composites, which reduces the ME coupling.[16]

Besides utilizing the product property of magnetostrictive and piezoelectric effects mentioned above, the extrinsic ME effect can also be realized in multiphase magnet-piezoelectric composites using the direct coupling of the magnetic attractive/repellent force effect in the magnet phase with the piezoelectric effect in the piezoelectric phase, which is not influenced by interface condition.[17–20] In this work, we report a large ME coupling obtained from a very simple bi-rectangular structure made up of epoxy-bonded negative/positive magnetostrictive and piezoelectric flakes.

II. EXPERIMENTAL

The bi-rectangular ME composite structure of negative/positive magnetostrictive and piezoelectric flakes is shown illustratively in Fig. 1. The commercial PZT flakes used in this work were sliced with various dimensions of $8 \times 6 \times 0.7$, $10 \times 6 \times 0.7$, $15 \times 6 \times 0.7$, and $18 \times 6 \times 0.7$ mm^3 ($l \times$ w $\times$ t). It was polarized along the thickness direction and its dielectric, piezoelectric, and electromechanical coupling coefficients were $\varepsilon_{33} = 2400$, $d_{33} = 400$ pC N^{-1}, and $k_{33} = 0.70$, respectively. The $TbFe_2$ flakes were prepared by arc melting under argon atmosphere. Its dimensions were $6 \times 6 \times 2.5$, $8 \times 6 \times 2.5$, $10 \times 6 \times 2.5$, and $12 \times 6 \times 2.5$ mm^3 ($l \times w \times t$), respectively.

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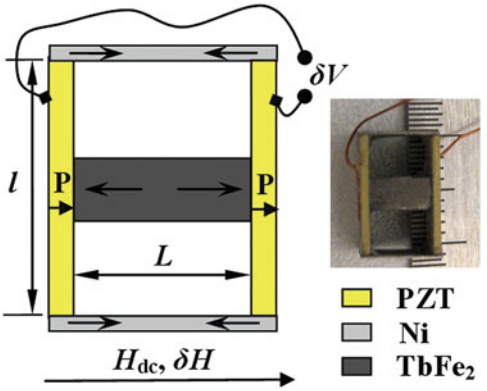


FIG. 1. Schematic illustration and photograph of a bi-rectangular magnetoelectric (ME) composite structure of negative/positive magnetostrictive and piezoelectric flakes.

The commercial Ni flakes were sliced with various dimensions of $8 \times 6 \times 0.1$, $10 \times 6 \times 0.11$, $12 \times 6 \times 0.1$, and $14 \times 6 \times 0.1$ mm^3 ($l \times w \times t$). The $TbFe_2$ and PZT flakes were bonded by using silver epoxy. The Ni and PZT flakes were not connected electrically by using epoxy.

In this case, the interface between magnetostrictive and piezoelectric phases is not required to achieve ME coupling. The working principle of the bi-rectangular ME composite structure is as follows. When an external ac magnetic field (H_{ac}) is applied along the length direction of magnetostrictive flakes (Ni and $TbFe_2$ flakes), the $TbFe_2$ flake will produce an extensional strain while the Ni flakes will produce a shrinkage strain because of the difference in magnetostrictive properties of each flake. These strains then transfer to the central and end parts of the PZT flakes. Due to the piezoelectric effect, the PZT flakes will induce a charge output.

The ME effect of the proposed structure was measured by applying both constant (H_{dc}) and alternating (δH) magnetic fields parallel to the length direction of $TbFe_2$ flakes. The induced voltage signal δV across the sample was amplified and measured by an oscilloscope.[8] The ME voltage coefficient was calculated based on $\alpha_E = \delta V/(t_{PZT}\delta H)$, where t_{PZT} is the thickness of PZT flakes. In our experiment, $\delta H = 1.2$ Oe as the amplitude of the AC current through the coil is equal to 1 A.

III. RESULTS AND DISCUSSION

Figure 2(a) shows the bias magnetic field (H_{dc}) dependence of ME voltage coefficient (α_E) for the bi-rectangular ME composite structure. The measurements were carried out at $f = 1$ kHz. The lengths of the $TbFe_2$ and PZT flakes (L and l) are 6 and 10 mm, respectively. Data are shown for both increasing and decreasing H_{dc}. On increasing H_{dc} from zero, it is seen that α_E decreases until a minimum value is reached at a certain H_{dc} and then increases subsequently. With further increase in H_{dc}, α_E reaches a maximum value around $H_{dc} = 3.5$ kOe and then decreases subsequently. The measurements on the ac magnetic field frequency (f) dependence of the ME coupling are also performed. For such studies, the bias magnetic field H_{dc} was set at 200 Oe and α_E was measured as f was varied from 1 to 150 kHz. As shown in Fig. 2(b), one observes that there are four obvious resonance peaks which are related to the electromechanical resonance.[9] And a maximum ME voltage coefficient $\alpha_E = 1.8$ V cm^{-1} Oe^{-1} can be obtained at resonance frequency of $f_r = 80.2$ kHz. The maximum ME voltage coefficient α_E observed here is much larger than that of layered ME Ni/PZT/Ni, $TbFe_2$/PZT/$TbFe_2$, and Ni/PZT/$TbFe_2$ composites with the same Ni, $TbFe_2$, and PZT reported in Ref. 8, which indicates that the proposed structure is more efficient than traditional ones.

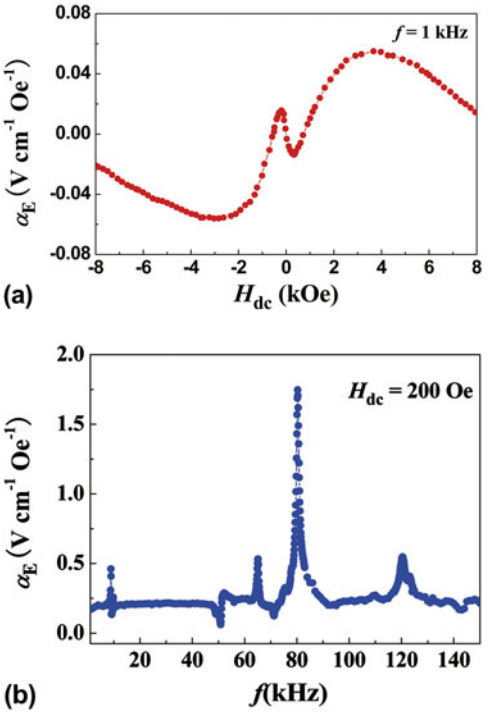


FIG. 2. (a) The bias magnetic field (H_{dc}) at $f = 1$ kHz and (b) frequency at $H_{dc} = 200$ Oe dependences of ME voltage coefficient (α_E) for the bi-rectangular ME composite structure with $L = 6$ mm and $l = 10$ mm.

Figure 3(a) shows the three-dimensional plot of the bias magnetic field (H_{dc}) and frequency (f) dependences of ME voltage coefficient (α_E) for the bi-rectangular ME composite structure with $L = 6$ mm and $l = 10$ mm. At resonance frequency of $f_r = 80.2$ kHz, the α_E increases with H_{dc}, up to 3.0 V cm^{-1} Oe^{-1} at $H_{dc} = 0.5$ kOe, and then decreases slightly as the H_{dc} increases until 1 kOe. With further increasing H_{dc} from 1 to 3.5 kOe, one observes that the α_E shows a nearly constant plateau behavior. But when $H_{dc} > 3.5$ kOe, the α_E decreases as

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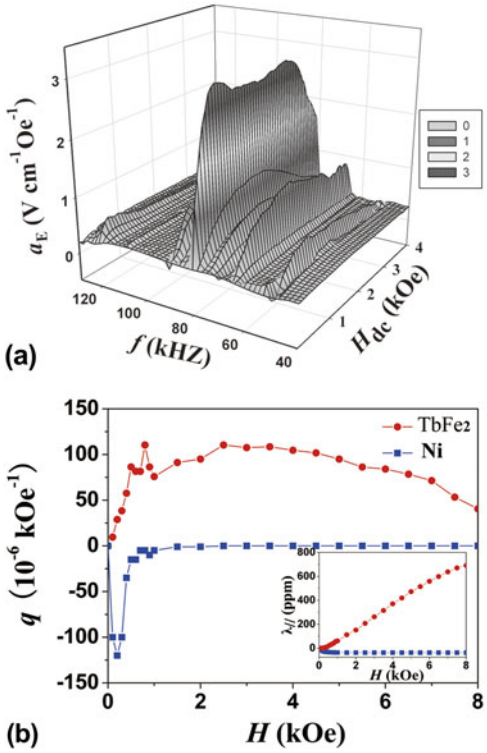


FIG. 3. (a) Three-dimensional plot of the bias magnetic field (H_{dc}) and frequency (f) dependences of ME voltage coefficient (α_E) for the bi-rectangular ME composite structure with L = 6 mm and l = 10 mm and (b) the applied magnetic field dependence of the piezomagnetic coupling coefficient q for $TbFe_2$ and Ni flakes. The inset shows the applied magnetic field dependence of the in-plane parallel magnetostriction $\lambda_{//}$ for $TbFe_2$ and Ni flakes.

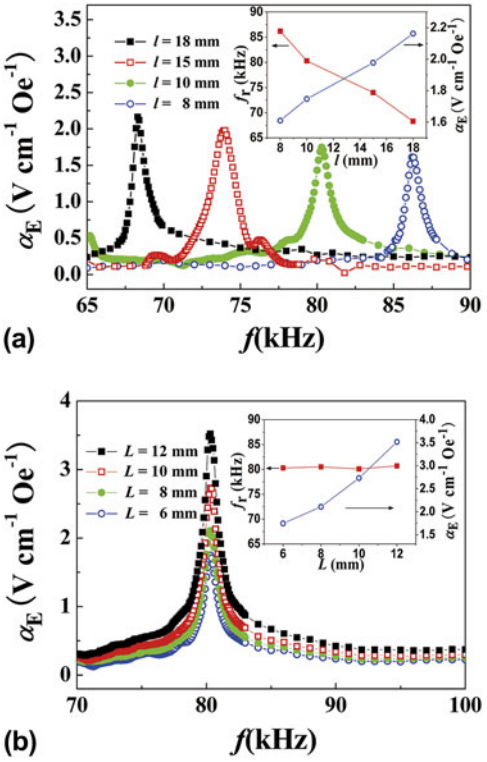


FIG. 4. (a) Frequency (f) dependence of the ME voltage coefficient (α_E) with various l for the bi-rectangular ME composite structure. The inset shows the l dependence of the f_r and the maximum α_E. (b) f dependence of the α_E with various L. The inset shows the L dependence of the f_r and the maximum α_E.

H_{dc} increases, which is the same as the phenomena shown in Fig. 2(a). The wide-field behavior of the maximum α_E at resonance frequency, which can provide a stable electrical signal in a wide range of magnetic field, offers the possibility of making multifunctional devices from this kind of structure.

The values of the ME voltage coefficient α_E are proportional to the piezomagnetic coupling coefficient of the magnetostrictive flake $q \sim \delta\lambda/\delta H$,[9] where λ is the magnetostriction. The linear magnetostriction was measured by using a standard strain-gauge technique in directions parallel ($\lambda_{//}$) to applied magnetic fields. The applied magnetic field (H) dependence of the piezomagnetic coupling coefficient (q) for $TbFe_2$ and Ni flakes is shown in Fig. 3(b). The applied magnetic field dependence of the in-plane parallel magnetostriction $\lambda_{//}$ for $TbFe_2$ and Ni flakes is also shown as an inset in Fig. 3(b). The $TbFe_2$ flake shows a positive magnetostriction, while the Ni flake exhibits a negative magnetostriction. When $H > 1$ kOe, the q of Ni is equal to zero due to the magnetostriction of Ni reaches the saturation stage. But the magnetostriction of $TbFe_2$ does not reach the saturation stage even though $H = 8$ kOe. For the bi-rectangular ME composite structure, when $H < 1$ kOe, the magnetostriction of Ni does not reach the saturation stage, both Ni and $TbFe_2$ flakes play a role in α_E, which results in a negative α_E in low field [shown in Fig. 2(a)] and a largest ME voltage coefficient obtained with a certain H_{dc} at resonance frequency. When 1 kOe $< H <$3.5 kOe, the magnetostriction of Ni reaches the saturation stage, $TbFe_2$ flake plays a predominant role in α_E. The q of $TbFe_2$ flake increases as H increases. Hence, the α_E increases slowly until a maximum value is reached [shown in Fig. 2(a)]. When $H > 3.5$ kOe, the q of $TbFe_2$ flake decreases as H increases, so the α_E decreases. In the last two processes, α_E shows a nearly plateau behavior at resonance frequency.

For layered ME composites, the resonance frequency is strongly influenced by the length of piezoelectric layer, $f \propto 1/l$.[8] The resonance frequency decreases as the length of piezoelectric layer increases. Figure 4(a) shows the frequency (f) dependence of the ME voltage coefficient (α_E) with various length (l) of PZT flakes. One observes that the α_E and resonance frequency (f_r) depend strongly on l. With the increasing in l from 8 to 18 mm, the f_r of the bi-rectangular ME composite structure gradually decreases from 86.2 to 68.3 kHz, while the maximum α_E of the composite structure increases from 1.61 to 2.16 V cm^{-1} Oe^{-1}, which is also shown as an inset in Fig. 4(a). The experimental result is in good agreement with the theoretical

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prediction. The dependence of the α_E on f with various lengths (L) of $TbFe_2$ flakes is shown in Fig. 4(b). The L dependence of the f_r and the maximum α_E is also shown as an inset in Fig. 4(b). It can be seen that the value of the maximum α_E increases with L, which can be ascribed to the increasing magnetostriction due to shape demagnetization effect.[21] The resonance frequency is not affected by L, which is attributed to the bi-rectangular structure. Obviously, if the length of PZT and $TbFe_2$ flakes (l and L) is increased, higher values of ME voltage coefficient would be obtained.

IV. CONCLUSIONS

In summary, a large ME coupling has been obtained from a very simple bi-rectangular structure. The experimental results have demonstrated that the ME effect is affected by negative and positive magnetostrictive flakes, which is different from that of other ME structures. At resonance frequency of $f_r = 80.2$ kHz, the α_E shows a nearly constant plateau behavior with H_{dc} increased from 1 to 3.5 kOe, which is believed to be useful for future device application. The resonance frequency of the bi-rectangular ME composite structure gradually decreases as the length of PZT flakes increases. The maximum ME voltage coefficient increases with the length of PZT or/and $TbFe_2$ flakes. The outstanding advantage of the proposed structure is that there is no interface between magnetostrictive and piezoelectric flakes required to achieve ME coupling, which provides a new choice to make ME devices.

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