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High magnetoelectric tuning effect in a polymer-based magnetostrictive-piezoelectric laminate under resonance drive

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A polymer-based magnetoelectric (ME) laminate was fabricated by sandwiching one layer of thickness-polarized, length-stretched polyvinylidene fluoride (PVDF) piezoelectric polymer between two layers of length-magnetized, epoxy-bonded $Tb_{0.3}Dy_{0.7}Fe_{1.92}$ (Terfenol-D) pseudo-1–3 magnetostrictive particulate composite in the thickness direction, and its resonance ME effect was investigated, both experimentally and theoretically, as a function of magnetic bias field (H_{Bias}). The laminate showed a high ME voltage coefficient (α_V) of 233 mV/Oe at the fundamental resonance frequency (f_r) of 60.6 kHz under a relatively low H_{Bias} of 0.6 kOe. By controlling H_{Bias} in the range of 0.02–1.5 kOe, nonlinear tunabilities as high as 1382 and 8.6% were achieved for α_V and f_r, respectively, as a result of the reduced eddy-current losses and enhanced non-180° domain-wall motion-induced *negative-ΔE* effect in the Terfenol-D composite layers as well as the increased compliance contribution from the PVDF polymer layer to allow the motion of non-180° domain walls in the Terfenol-D composite layers. This improved resonance ME tuning effect, together with the durable and tailorable natures, makes the laminate great promise for developing into tunable ME devices. © *2012 American Institute of Physics*. [doi:10.1063/1.3678321]

The magnetoelectric (ME) effect in materials has attracted much research attention in recent years. Laminated composites based on $Tb_{0.3}Dy_{0.7}Fe_{1.92}$ (Terfenol-D) magnetostrictive alloy and $0.7Pb(Mg_{1/3}Nb_{2/3})O_3$–$0.3PbTiO_3$ (PMN–PT) piezoelectric crystal have been a main focus because of their generally high extrinsic ME effect arisen from the mechanically mediated magnetostrictive and piezoelectric effects.[1,2] When operating at resonance, these alloy-crystal-based laminates often yield an enhanced resonance ME effect of ∼10 times higher in ME voltage coefficient (α_V) compared to their nonresonance α_V counterparts.[2] Recently, ME tuning has become a physically interesting and technologically important topic for realizing tunable ME devices.[1–4] From the aspect of magnetically ME tuning, the highest tunable α_V of ∼10 times (i.e., α_V tunability of ∼1000%) and tunable resonance frequency (f_r) of ∼2 kHz range (i.e., f_r tunability of ∼3%) in a magnetic bias field (H_{Bias}) range of 0.02–0.8 kOe have been observed in the Terfenol-D/PMN–PT laminates.[2]

In this paper, we report a high H_{Bias}-controllable nonlinear resonance ME tuning effect in a polymer-based ME laminate formed by a polyvinylidene fluoride (PVDF) piezoelectric polymer film and two epoxy-bonded Terfenol-D pseudo-1–3 magnetostrictive particulate composite plates. The use of Terfenol-D composite, instead of Terfenol-D alloy, is to alleviate its intrinsic weaknesses of high eddy-current losses at elevated frequencies, negligibly small *negative-ΔE* effect (or magnetomechanical tuning effect) caused by limited non-180° domain-wall motion under zero stress bias, and

mechanical brittleness. The adoption of the relatively compliant PVDF polymer, in place of PMN–PT crystals or $Pb(Zr, Ti)O_3$ (PZT) ceramics, not only relieves the elastic clamping effect on the Terfenol-D composite for best possibly exhibiting the built-in non-180° domain-wall motion, the large *negative-ΔE* effect, and hence the high ME tuning effect, but also improves the durability of the whole laminate.

Figure 1(a) shows the schematic diagram of the proposed polymer-based ME laminate in the Cartesian coordinate system. Two Terfenol-D composite plates were bonded on a PVDF polymer film in the thickness direction using a silver-loaded epoxy adhesive. The Terfenol-D composite plates, which were prepared in house using Terfenol-D particles with randomly distributed sizes of 10–300 μm and Spurr epoxy as the active and passive phases, respectively, had a cross-sectional area of 12×6 mm^2, a thickness of 1 mm, a particulate volume fraction of 0.51, and a magnetization (M) along the length direction.[5] The PVDF polymer film was commercially acquired to have the same cross-sectional area as the Terfenol-D composite plates, a different thickness of 0.1 mm, full-fired silver electrodes on the two major surfaces, a polarization (P) along the thickness direction, and a mechanical stretch in the length direction.[6]

The working principle of the polymer-based ME laminate in Fig. 1(a) is as follows. An ac magnetic field (H_3) applied along the length direction of the laminate causes the two length-magnetized Terfenol-D composite plates to produce magnetostrictive strains in their length direction due to the magnetostrictive effect. Since the PVDF polymer film is mechanically bonded and sandwiched between the two Terfenol-D composite plates, these magnetostrictive strains subsequently stress the PVDF polymer film to generate

piezoelectric voltages (V_3) across its thickness owing to the piezoelectric effect. Using the equation of motion to couple the magnetostrictive and piezoelectric constitutive relations,[7,8] applying Newton's second law of motion to the laminate,[7] taking free-free mechanical boundary conditions,[9] considering mechanical losses,[10] and finding analogous electrical equivalent parameters,[9] a dynamic magneto-elasto-electric equivalent circuit of the proposed laminate in Fig. 1(a) was obtained and is shown in Fig. 1(b). The ME voltage coefficient (α_V) of the laminate can be expressed as

$$\alpha_V = \left| \frac{dV_3}{dH_3} \right| = \left| \frac{\varphi_m \varphi_p}{\varphi_p^2 + j\omega C_0 R + j\omega C_0 Z} \right|, \tag{1}$$

$$\varphi_m = \frac{A d_{33,m}}{s_{33}^H}, \quad \varphi_p = \frac{w d_{31,p}}{s_{11}^E}, \quad C_0 = \frac{lw\varepsilon_{33}^S}{t_p},$$

$$R = \frac{\pi \sqrt{n s_{11}^E + (1-n) s_{33}^H \rho A}}{8 Q_{mech} \sqrt{s_{11}^E s_{33}^H}},$$

$$Z = -\frac{1}{2} j \sqrt{\frac{n\rho}{s_{33}^H} + \frac{(1-n)\rho}{s_{11}^E}} A \cot\left(\frac{kl}{2}\right),$$

where φ_m and φ_p are the magnetoelastic and elastoelectric coupling coefficients, respectively; C_0 is the clamped capacitance of the PVDF polymer film; R is the mechanical damping resistance of the laminate caused by internal frictions between material phases; and Z is the total mechanical impedance of the laminate. Other symbols have their usual meanings. The resonance frequency (f_r) can be written as

$$f_r = \sqrt{\frac{n}{S_{33}^H} + \frac{1-n}{S_{11}^E}}. \tag{2}$$

Figures 2(a) and 2(b) plot the measured and calculated α_V of the polymer-based ME laminate driven by an H_3 of 1 Oe peak in the f range of 1–100 kHz under various H_{Bias} of 0.02–1.5 kOe, respectively. The α_V–f curves in Fig. 2(b) were calculated based on Eq. (1), with the material properties given in Fig. 3(b) and Refs. 5 and 6 and the geometric parameters described in Fig. 1. It is clear that the measured and calculated α_V agree reasonably well with each other for all H_{Bias} levels

with no remarkable frequency dispersion, except for the variations associated with the resonance frequency range of 50–70 kHz. The observation indicates that the eddy-current losses are insignificant in the laminate for f up to 100 kHz. Moreover, the largest resonance α_V is found at a relatively low H_{Bias} level of 0.6 kOe for both measurement and calculation. In particular, the largest resonance α_V is detected to be 233 mV/Oe at $f_r = 60.6$ kHz by measurement and 228 mV/Oe at $f_r = 60.9$ kHz by calculation. These resonance α_V are ~20 times larger than their nonresonance α_V of ~12 mV/Oe. This resonance amplification of α_V is even more significant compared to the best known alloy-crystal-based Terfenol-D/PMN–PT laminates of ~10 times as a result of improved coupling between the Terfenol-D composite plates and the PVDF polymer film in our laminate.[2] When H_{Bias} is reduced to 0.02 kOe, the measured and calculated resonance α_V undergo a large reduction of ~15 times to 16 and 17 mV/Oe, respectively, while their corresponding f_r exhibit an up-shift of ~2 kHz to 62.5 and 62.2 kHz, respectively. At an elevated H_{Bias} level of 1.5 kOe, the measured and calculated resonance α_V have ~1.2 times reduction to 193 and 204 mV/Oe, respectively, but their corresponding f_r exhibit a large up-shift of ~4 kHz to 65.7 kHz, respectively. The observations indicate that besides the presence of a high resonance ME effect in our laminate, there also exist an interestingly high H_{Bias}-controllable nonlinear resonance ME tuning effect in the laminate.

Figure 3(a) shows the comparison between the measured and calculated resonance α_V and f_r as a function of H_{Bias}. To enable a quantitative description of the H_{Bias}-controllable nonlinear resonance ME tuning effect in Fig. 2, the tunability
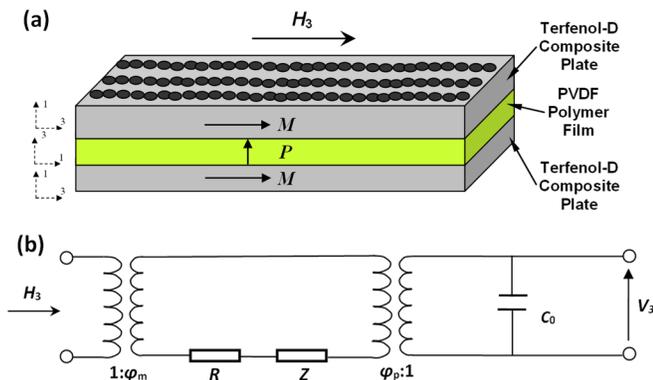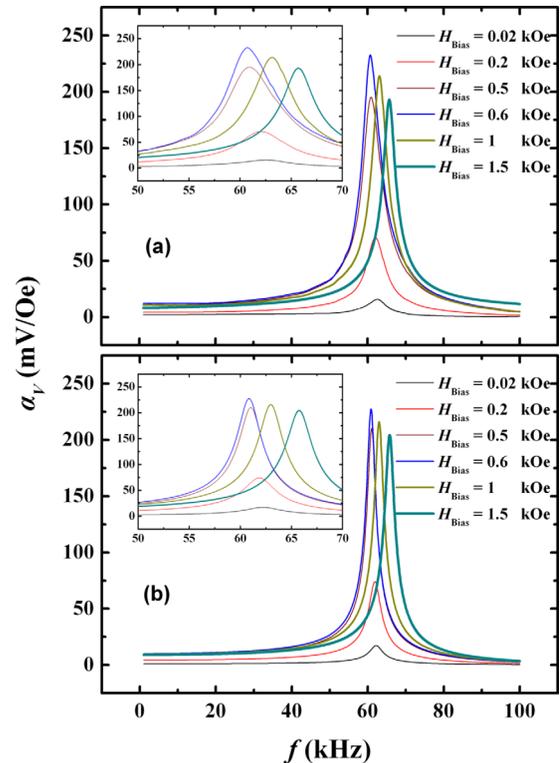

FIG. 1. (Color online) (a) Schematic diagram of the proposed polymer-based ME laminate in the Cartesian coordinate system. (b) Dynamic magneto-elasto-electric equivalent circuit of the laminate.

FIG. 2. (Color online) (a) Measured and (b) calculated α_V of the polymer-based ME laminate driven by an H_3 of 1 Oe peak in the f range of 1–100 kHz under various H_{Bias} of 0.02–1.5 kOe.
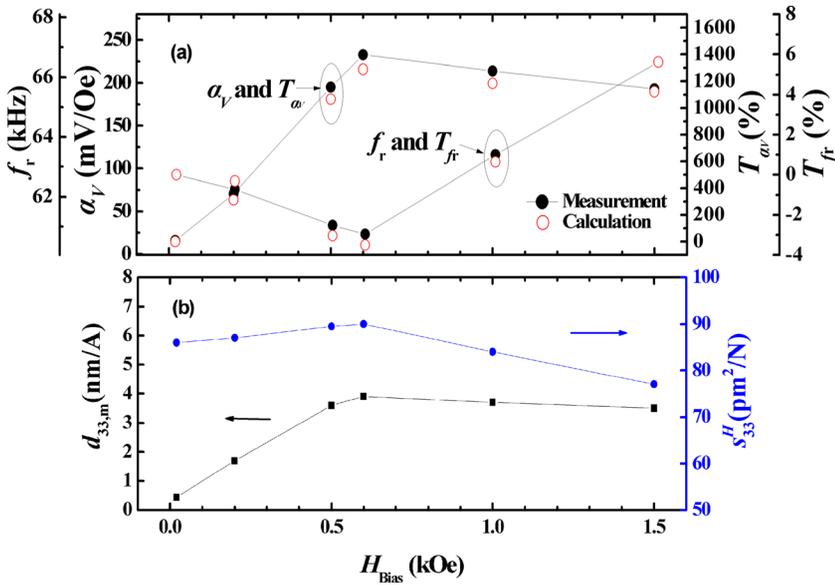
FIG. 3. (Color online) (a) Comparison between the measured and calculated resonance α_V and f_r as well as T_{α_V} and T_{f_r} as a function of H_{Bias}. (b) Variations of $d_{33,m}$ and s_{33}^H with H_{Bias} in the Terfenol-D composite plates.

of α_V (T_{α_V}) and tunability of f_r (T_{fr}) expressed, respectively, as follows are also included.

$$T_{\alpha_V} = \left| \frac{\alpha_V - \alpha_{V @ HBias,min}}{\alpha_{V @ HBias,min}} \right| \qquad (3)$$

$$T_{fr} = \frac{f_r - f_{r@HBias,min}}{f_{r@HBias,min}}. \qquad (4)$$

It is noted that $\alpha_{V @ HBias,min}$ and $f_{r@HBias,min}$ in Eqs. (3) and (4) are taken as the values of α_V and f_r at $H_{Bias} = 0.02$ kOe in our case. From Fig. 3(a), the measured data agree well with the calculated data with only ~3% discrepancy. Both α_V and T_{α_V} increase initially up to 233 mV/Oe and 1382%, respectively, at $H_{Bias} = 0.6$ kOe and then decrease to 193 mV/Oe and 1130%, respectively, at an elevated $H_{Bias} = 1.5$ kOe. In the meantime, both f_r and T_{fr} decrease with increasing H_{Bias}, reaching 62.7 kHz and −3%, respectively, at $H_{Bias} = 0.6$ kOe and then increasing to 65.7 kHz and 5.6%, respectively, at $H_{Bias} = 1.5$ kOe. These suggest the high H_{Bias}-controllable nonlinear tunabilities of 1382 and 8.6% for α_V and f_r, respectively. Since an average axial residual compressive stress of ~3 MPa has been developed in our Terfenol-D composite plates through the thermal cure of the Spurr epoxy phase during composite fabrication, this compressive stress essentially exerts on the embedded Terfenol-D particles to create a preferred non-180° domain-wall state in the composite.[5] Thus, the initial increase in α_V and T_{α_V} as well as the initial decrease in f_r and T_{fr} with increasing H_{Bias} can be explained by the H_{Bias}-induced motion of the available non-180° domain walls in the Terfenol-D composite plates. That is, as H_{Bias} is increased near 0.6 kOe, the compliance associated with the increased deformation contribution from this non-180° domain-wall motion is maximized, resulting in a maximum in strain (and hence α_V and T_{α_V}) and a minimum in stiffness (and hence f_r and T_{fr}). The effect is said to be the maximization of *negative*-ΔE effect at $H_{Bias} = 0.6$ kOe.[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 an increase in stiffness. Figure 3(b) illustrates the variations of $d_{33,m}$ and s_{33}^H with H_{Bias} in the Terfenol-D composite plates. The maximization

of $d_{33,m}$ and s_{33}^H at $H_{Bias} = 0.6$ kOe not only confirms essentially our domain-wall motion discussion, but also supports our design of using PVDF polymer to relieve the elastic clamping effect on the Terfenol-D composite for best possibly exhibiting the built-in non-180° domain-wall motion, the large *negative*-ΔE effect, and hence the high ME tuning effect.

We have reported experimentally and theoretically a high H_{Bias}-controllable nonlinear resonance ME tuning effect in a polymer-based ME laminate formed by Terfenol-D composite plates and PVDF polymer film. A high resonance α_V of 233 mV/Oe, which is ~20 and ~2 times larger than its nonresonance α_V and the best known alloy-crystal-based Terfenol-D/PMN–PT laminates, respectively, has been observed at $f_r = 60.6$ kHz under $H_{Bias} = 0.6$ kOe due to the improved magneto-elasto-electric coupling between the Terfenol-D composite plates and the PVDF polymer film. Nonlinear tunabilities as high as 1382 and 8.6% have been achieved for α_V and f_r, respectively, in the H_{Bias} range of 0.02–1.5 kOe because of the reduced eddy-current losses and enhanced non-180° domain-wall motion-induced *negative*-ΔE effect in the Terfenol-D composite plates as well as the increased compliance contribution from the PVDF polymer film to allow the motion of non-180° domain walls in the Terfenol-D composite plates. The laminate has great promise for use in various tunable ME devices.

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