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High magnetoelectric effect at low magnetic biasing in heterostructure rod of magnetostrictive fibers, piezoelectric tube, and epoxy binder

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A three-phase heterostructure rod of 35 mm length and 8 mm diameter is developed by embedding 18 pieces of length-magnetized $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.92}$ magnetostrictive continuous fibers in a wall-thickness-polarized $\text{Pb}(\text{Zr}, \text{Ti})\text{O}_3$ piezoelectric long-thin tube using an epoxy binder in order to study its magnetoelectric (ME) effect for passive sensing of axial magnetic fields. The heterostructure rod exhibits a high non-resonance ME voltage coefficient (α_V) of 38 mV/Oe for frequencies up to 10 kHz and an ultrahigh resonance α_V of 680 mV/Oe at 32 kHz, both under a reasonably low magnetic bias field (H_{Bias}) of 120 Oe. The simultaneously high axial ME responses at the reasonably low H_{Bias} , as confirmed theoretically and experimentally, can be ascribed to the realization of the relatively true magnetostrictive 33-mode and piezoelectric 31-mode of operation in the magnetostrictive continuous fibers and piezoelectric long-thin tube, respectively, as well as the reduction in the effect of demagnetization fields in the high aspect-ratio magnetostrictive continuous fibers.

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I. INTRODUCTION

The magnetoelectric (ME) effect extrinsic in magnetostrictive–piezoelectric laminates has been widely investigated for passive magnetic field sensing over the past decade.^{1–5} One of the largest obstacles to practically realize ME sensors is the need of a high magnetic bias field (H_{Bias}), reaching 400–600 Oe in the well-known L – T laminated plates and even ~ 5000 Oe in the T – T laminated plates, for maximizing the magnetostrictive response for the piezoelectric phase.^{1,2} The need of a high H_{Bias} implies the need of big and strong permanent magnets in the resulting sensors, which in turn increases the size, mass, magnetic influence inside and outside the sensors, and packaging difficulty.⁶ Today, reducing H_{Bias} without reducing sensitivity remains a challenging topic to the ME community. Among several published works, the one reporting heterostructures of metglas and piezoelectric fibers has demonstrated a very high ME voltage coefficient (α_V) of 1.5 V/Oe at a non-resonance frequency of 1 kHz under an extremely low H_{Bias} of ~ 8 Oe.⁷ However, this extremely low H_{Bias} not only constrains the heterostructures to small-signal applications but also is easily affected by environmental noises.

In this paper, we report theoretically and experimentally the existence of a high axial ME effect at a reasonably low H_{Bias} of 120 Oe in a three-phase heterostructure rod, comprising 18 pieces of length-magnetized $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.92}$ magnetostrictive continuous fibers bonded with an epoxy binder in a wall-thickness-polarized $\text{Pb}(\text{Zr}, \text{Ti})\text{O}_3$ piezoelectric long-thin tube. The use of the magnetostrictive continuous fibers and piezoelectric long-thin tube are aimed to enable the relatively true magnetostrictive 33-response and piezoelectric 31-

response in the heterostructure rod, respectively. Besides, the high length-to-width aspect ratio (35:1 in our case) of the magnetostrictive continuous fibers can effectively reduce the effect of demagnetization fields, thereby providing a lower field of operation. The use of the epoxy binder is to provide a good mechanical mediation for the magnetostrictive continuous fibers and the piezoelectric long-thin tube. From another view, our heterostructure rod can be considered as a “rod-in-tube” ME rod in which an epoxy-bonded magnetostrictive continuous fiber composite rod is situated in the middle of a piezoelectric ceramic long-thin tube. The creation of the epoxy-bonded magnetostrictive continuous fiber composite rod can also mitigate the eddy-current losses and mechanical brittleness intrinsic in most of the monolithic magnetostrictive materials. As a result, our ME rod exhibits simultaneously high non-resonance and resonance axial ME responses at a reasonably low H_{Bias} of 120 Oe.

II. STRUCTURE AND WORKING PRINCIPLE

Figure 1(a) illustrates the schematic diagram of the proposed three-phase heterostructure rod with an axial magnetic field sensing mode configuration. The heterostructure rod, having 35 mm length and 8 mm diameter, consists of a total of 18 pieces of length-magnetized $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.92}$ magnetostrictive continuous fibers inserted inside a wall-thickness-polarized $\text{Pb}(\text{Zr}, \text{Ti})\text{O}_3$ piezoelectric long-thin tube and bonded using an epoxy binder. It can also be considered as a “rod-in-tube” ME rod in which the central portion of magnetostrictive continuous fibers and epoxy binder essentially form an epoxy-bonded magnetostrictive continuous fiber composite rod. In fabrication, the piezoelectric long-thin tube was used as the mold, and the epoxy-bonded magnetostrictive continuous fiber composite rod was prepared inside

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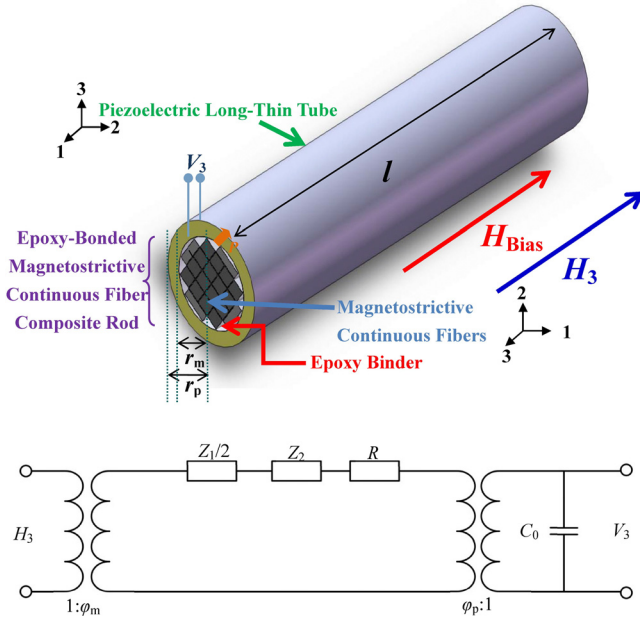


FIG. 1. (a) Schematic diagram of the proposed three-phase heterostructure rod (or ME rod) with an axial magnetic field sensing mode configuration. (b) Dynamic magneto-mechano-electric equivalent circuit of the proposed ME rod.

the mold under vacuum to ensure a good structural integrity. In more detail, a Ferroperm Pz26 hard $\text{Pb}(\text{Zr}, \text{Ti})\text{O}_3$ piezoelectric long-thin tube, with dimensions of 35 mm length (l) \times 8 mm outer diameter ($2r_p$) \times 6 mm inner diameter ($2r_m$), full-fired silver electrodes on the inner and outer circumferential surfaces normal to the wall-thickness direction, and an electric polarization (P) along the wall-thickness direction, was commercially acquired. Magnetostrictive continuous fibers with a square cross-section of 35 mm long and 1 mm width and the highly magnetostrictive [112] crystallographic axis oriented along their length direction were cut along the length of a Baotou Rare Earth [112]-textured monolithic $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.92}$ plate using a wire electrical discharge machining technique. With the piezoelectric long-thin tube mold being sealed at one end, a total of 18 pieces of magnetostrictive continuous fibers were placed inside the mold under a dc magnetic alignment field of ~ 3000 Oe set up by a pair of NdFeB magnets. The predegassed Araldite LY564/HY2954 epoxy was transferred into the mold and degassed again inside the mold, both under vacuum. The other end of the mold was sealed, and the epoxy was allowed to cure at 80°C for 9 h inside an oven. The volume fraction of the magnetostrictive continuous fibers was theoretically determined to be ~ 0.63 in the epoxy-bonded magnetostrictive continuous fiber composite rod, based on the rule-of-mixture formulation for density. In fact, epoxy-bonded $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.92}$ -based composites with $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.92}$ volume fraction in excess of 0.5 have been found to be optimal for magnetostrictive and ME applications by taking the balance between the maximization of magnetostrictive response and the minimization of eddy-current losses and material cost into consideration.^{8,9}

The axial magnetic field sensing in the ME rod in Fig. 1(a) can be described as follows. An ac magnetic field

(H_3) applied along the axial (or length) direction of the ME rod causes the inner epoxy-bonded magnetostrictive continuous fiber composite rod to produce axial magnetostrictive strains in its length direction due to the magnetostrictive 33-mode effect. Since the epoxy-bonded magnetostrictive continuous fiber composite rod is mechanically bonded inside the piezoelectric long-thin tube, these axial magnetostrictive strains will subsequently stress on the outer piezoelectric long-thin tube to generate an ac voltage (V_3) across its wall-thickness in the wall-thickness direction because of the piezoelectric 31-mode effect. Using the equation of motion to couple the magnetostrictive and piezoelectric constitutive relations,^{10,11} applying Newton's second law of motion to the ME rod,¹⁰ taking free-free mechanical boundary conditions at the two ends of the ME rod,¹² considering mechanical losses and internal stresses,^{13,14} and finding analogous electrical equivalent parameters,¹² a dynamic magneto-mechano-electric equivalent circuit of the proposed ME rod, shown in Fig. 1(a), was obtained, and is shown in Fig. 1(b). Therefore, the ME voltage coefficient (α_V) of the ME rod can be determined as

$$\alpha_V = \left| \frac{dV_3}{dH_3} \right| = \left| \frac{-2\varphi_m\varphi_p\beta}{2\varphi_p^2 + j\omega C_0(Z_1 + 2Z_2 + R)} \right|, \quad (1)$$

$$\varphi_m = \frac{A_m d_{33,m}}{s_{33}^H}, \quad \varphi_p = \frac{2\pi(r_p - r_m)d_{31,p}}{s_{11}^E},$$

$$C_0 = \frac{\pi l (\epsilon_{33}^T - d_{31,p}^2)}{s_{11}^E},$$

$$Z_1 = j \sqrt{\frac{n\rho}{s_{33}^m} + \frac{(1-n)\rho}{s_{11}^E}} A \tan(kl/2),$$

$$Z_2 = \frac{\sqrt{\frac{n\rho}{s_{33}^m} + \frac{(1-n)\rho}{s_{11}^E}} A}{j \sin(kl)},$$

where φ_m and φ_p are the magnetomechanical and mechano-electric coupling coefficients, respectively; ω is the angular frequency; C_0 is the clamped capacitance of piezoelectric long-thin tube; β is the internal stresses acting on the magnetostrictive continuous fibers; R is the mechanical damping resistance of the ME rod; Z_1 and Z_2 are the mechanical impedances of the ME rod; A and l are the cross-sectional area and length of the ME rod, respectively; A_m and r_m are the cross-sectional area and radius of the epoxy-bonded magnetostrictive continuous fiber composite rod, respectively; r_p is the outer radius of the piezoelectric long-thin tube; $d_{33,m}$ and $d_{31,p}$ are the piezomagnetic and piezoelectric coefficients, respectively; s_{33}^H and s_{11}^E are the elastic compliance coefficients at constant magnetic field strength and at constant electric field strength, respectively; ϵ_{33}^T is the dielectric permittivity at constant stress; ρ is the average mass density of the ME rod; $n = r_m^2/r_p^2$ is the cross-section area ratio of the ME rod; and k is the wave number. From Eq. (1), it is noted that α_V of the ME rod depends on the material properties of the epoxy-bonded magnetostrictive continuous fiber composite rod and the piezoelectric long-thin tube

(i.e., $d_{33,m}$, $d_{31,p}$, s_{33}^H , s_{11}^E , ϵ_{33}^T , etc.) as well as the geometric parameters of the ME rod.

III. MEASUREMENTS

The ME effect in our ME rod was characterized at room temperature using an in-house automated ME measurement system.¹⁵ The ME rod was placed between the pole gap of a water-cooled, U-shaped electromagnet (Mytlen PEM-8005 K), and the electromagnet was energized by a dc current supply (Sorensen DHP200–15) to give a dc magnetic bias field (H_{Bias}). An arbitrary waveform generator (Agilent 33210 A) connected to a constant-current supply amplifier (AE Techtron 7796HF) was employed to drive a pair of Helmholtz coils for providing an ac magnetic drive field (H_3) over the prescribed frequency (f) range. H_{Bias} was monitored using a Hall probe connected to a Gaussmeter (F. W. Bell 7030), while H_3 was measured using a pick-up coil connected to an integrating fluxmeter (Walker MF-10D). All qualities were gathered, together with the induced ME voltage (V_3), using a data acquisition unit (Nation Instruments BNC-2110 and NI-PCI6132) under the control of a computer with a Labview program.

IV. RESULTS AND DISCUSSION

Figures 2(a) and 2(b) show the measured and calculated α_V of the ME rod driven by an H_3 of 1 Oe peak in the f range of 1–60 kHz under various H_{Bias} of 50–500 Oe, respectively. The α_V - f curves in Fig. 2(b) were calculated based on Eq. (1),

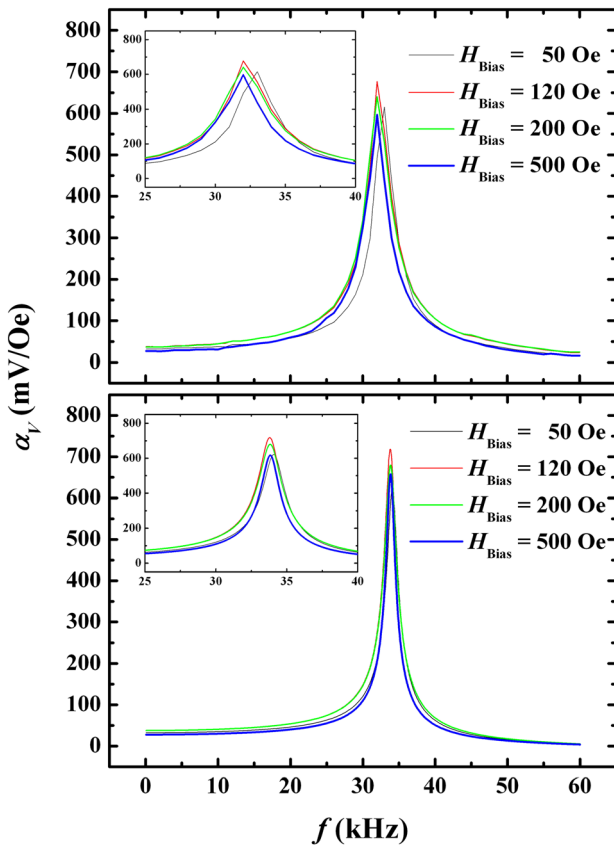


FIG. 2. (a) Measured and (b) calculated α_V of the ME rod driven by an H_3 of 1 Oe peak in the f range of 1–60 kHz under various H_{Bias} of 50–500 Oe.

with the material properties given in Refs. 9 and 16, and the geometric parameters were 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 (f_r) range of 25–40 kHz. The observation indicates that the eddy-current losses are insignificant in the ME rod for f up to, at least, 60 kHz. Moreover, the largest α_V response is found at a low H_{Bias} level of 120 Oe throughout the whole f range for both measurement and calculation. This is mainly due to the reduced effect of demagnetization fields in the high aspect-ratio magnetostrictive continuous fibers of 35:1. In particular, the largest resonance α_V is detected to be 680 mV/Oe at $f_r = 32$ kHz by measurement and 710 mV/Oe at $f_r = 33.8$ kHz by calculation. These resonances α_V , which are caused by the fundamental axial mode resonance with half-wave vibrations along the length of the ME rod, are ~ 20 times larger than their nonresonance α_V of ~ 38 mV/Oe for f up to 10 kHz. This resonance amplification of α_V is even more significant when compared to the widely known ME laminated plates of $\text{Tb}_{0.3}\text{Dy}_{0.7}\text{Fe}_{1.92}$ alloy and $0.71\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3-0.29\text{PbTiO}_3$ (PMN–PT) crystal of ~ 10 times, as a result of good magneto-mechano-electric coupling between the epoxy-bonded magnetostrictive continuous fiber composite rod and the piezoelectric long-thin tube in our ME rod enabled by the relatively true magnetostrictive 33-mode and piezoelectric 31-mode of operation in the magnetostrictive continuous fibers and piezoelectric long-thin tube, respectively. When the H_{Bias} level is reduced to 50 Oe, the measured resonance α_V undergoes a reduction of ~ 0.91 times to 617 mV/Oe, while its corresponding f_r exhibits an up-shift of ~ 1 kHz–33 kHz. At an elevated H_{Bias} level of 500 Oe, the measured resonance α_V has ~ 0.87 times reduction to 590 mV/Oe, but its corresponding f_r exhibits a smaller up-shift of ~ 0.2 kHz–33.2 kHz. The observations indicate that besides the presence of a high resonance ME effect in our ME rod, there also exists an interestingly high low- H_{Bias} -controllable nonlinear resonance ME tuning effect in the ME rod originated from increasing, maximizing, and decreasing the motion of non-180° domain walls in the epoxy-bonded magnetostrictive continuous fiber composite rod for H_{Bias} below, at, and above 120 Oe, respectively.^{5,9} This represents the operation of the ME rod in the magnetostrictive *negative*- ΔE region, at the *negative*- ΔE -*positive*- ΔE transition, and in the *positive*- ΔE region, respectively.^{5,9}

Figure 3 plots the measured V_3 over broad ranges of H_3 of 0.1–10 Oe and H_{Bias} of 20–500 Oe at 1 kHz and f_r . It is seen that V_3 varies essentially linearly with H_3 for different H_{Bias} at both 1 kHz and f_r . The good linearity between V_3 and H_3 confirms the validity of the proposed working principle. The largest V_3 - H_3 response is observed at $H_{\text{Bias}} = 120$ Oe for all f , which is in good agreement with the observations in Fig. 2. From the slopes of plot, α_V can be determined at different H_{Bias} and f . The inset of Fig. 3 shows the measured and calculated α_V as a function of H_{Bias} at 1 kHz and f_r . The measured and calculated α_V agree reasonably well with each other at both f . The relatively larger discrepancy at f_r than at 1 kHz is mainly due to the error in estimating the H_{Bias} -dependent mechanical damping resistance of the ME rod (R) in Eq. (1). Nonetheless, the measured α_V is found to increase in

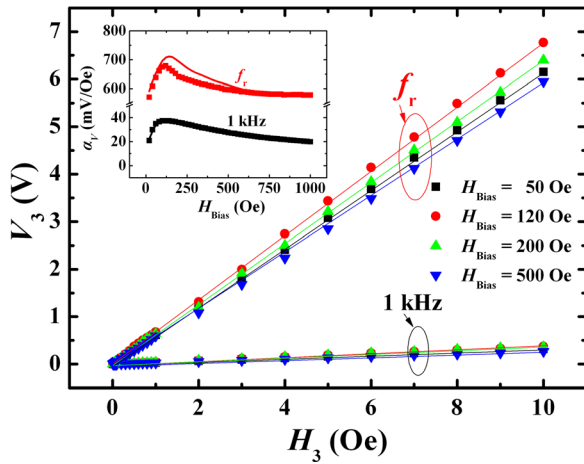


FIG. 3. Measured V_3 over broad ranges of H_3 of 0.1–10 Oe and H_{Bias} of 20–500 Oe at 1 kHz and f_r . The inset shows the measured (symbols) and calculated (lines) α_V as a function of H_{Bias} at 1 kHz and f_r .

the initial region of H_{Bias} of 0–120 Oe, reaching the maximum value of 38 and 680 mV/Oe at 1 kHz and f_r under a low H_{Bias} of 120 Oe, respectively, and then decreasing with increasing H_{Bias} . The observation suggests the presence of an interestingly high low- H_{Bias} -controllable nonlinear resonance ME tuning effect in the ME rod as described in Fig. 2. For comparison, the well-known L - T laminated plates generally exhibit a non-resonance α_V of ~ 60 mV/Oe at a higher H_{Bias} of 400–600 Oe,¹ while the T - T laminated plates demonstrate a quantitatively similar non-resonance α_V of ~ 60 mV/Oe at an even higher H_{Bias} of ~ 5000 Oe.² Therefore, the simultaneously high ME responses in our ME rod at the reasonably low H_{Bias} of 120 Oe can be attributed to the induction of the relatively true magnetostrictive 33-response and piezoelectric 31-response by the magnetostrictive continuous fibers and piezoelectric long-thin tube, respectively, in addition to the reduction in the effect of demagnetization fields through the high aspect ratio magnetostrictive continuous fibers.^{10,11}

V. CONCLUSION

We have found theoretically and experimentally the existence of a high non-resonance α_V of 38 mV/Oe for f up to 10 kHz and an ultrahigh resonance α_V of 650 mV/Oe at

33 kHz, both under a reasonably low H_{Bias} of 120 Oe in a 35 mm long, 8 mm diameter “rod-in-tube” ME heterostructure rod formed by an inner epoxy-bonded magnetostrictive continuous fiber composite rod and an outer piezoelectric long-thin tube. We have also attributed the observed axial ME responses at relatively low H_{Bias} to the deployment of the relatively true magnetostrictive 33-mode and piezoelectric 31-mode of operation in the ME rod, respectively, besides the reduction in the effect of demagnetization fields by the high aspect-ratio magnetostrictive continuous fibers. The single-dimensional design of the ME rod makes it high potential as an axial probe for passive sensing of axial magnetic fields.

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