¹³ Circumferential magnetoelectric effect in cylindrical piezoelectric-magnetostrictive composites with transverse bias magnetic field

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Received June 14, 2022 Revised August 1, 2022 Accepted September 22, 2022

Strong circumferential magnetoelectric (ME) effect has been observed in cylindrical PZT/Terfenol-D composite with transverse bias magnetic field. With dc bias magnetic field along axial direction and ac magnetic field along circumferential direction, strong ME voltage is obtained as $0.93 \text{ V} \cdot \text{cm}^{-1} \cdot \text{Oe}^{-1}$ at off-resonance frequency 1 kHz and as high as $32 \text{ V} \cdot \text{cm}^{-1} \cdot \text{Oe}^{-1}$ at resonance frequency $f_r = 46.0 \text{ kHz}$, which is much higher than that observed in planar ring-type ME laminate. The enhanced circumferential ME effect is attributed to improved interface mechanical coupling via normal stress. At room temperature, induced voltage from this cylindrical ME composite exhibits a near-linear response to applied ac vortex magnetic field amplitude over a wide magnetic field range of $10^{-8} < H_{ac} < 10^{-5}$ T both at low frequency and resonance frequency. Moreover, induced voltage has an excellent flat response to applied ac magnetic field frequency between Hz and kHz. The strong and stable circumferential ME effect in cylindrical ME laminate offers potential in vortex magnetic field sensor applications.

Key words: ME effect, piezoelectric, frequency response, magnetic sensor.

DOI: 10.21883/TPL.2022.12.54945.19280

Magnetoelectric (ME) effect in multiferroic composite structures has received continually increasing interest in past years, driven by its promising application for magnetic sensors, energy harvest devices, transducers and highpower miniature transformers [1,2]. It is known that the ME effect in laminated multiferroic composites is much stronger than in single phase multiferroics due to the product effect between piezoelectric and magnetostrictive phases [2]. Previous investigations mainly focused on ME composites with some simple configurations, such as rectangular, square, or disk shapes. Magnetic sensors based on ME effect in these geometries can only be used to detect magnetic field of constant direction. However, in some applications, one needs to probe vortex magnetic field excited by wires carrying current I, such as in power integrated circuits or superconducting films [3]. Thus it is necessary to develop ME laminate that can be applied in detection of vortex magnetic field. Dong etc. developed a vortex magnetic field sensor based on a ring-type ME composite composed of a circumferentially magnetized magnetostrictive Terfenol-D ring and a circumferentially polarized piezoelectric $Pb(Zr_{1-x}Ti_x)O_3$ (PZT) ring, which has high sensitivity of $< 10^{-9}$ T to a vortex magnetic field [4]. However, the prototype proposed by Dong is difficult to prepare due to the requirement of circumferential polarization and magnetization. In addition, the interfacial mechanical coupling between the piezoelectric and magnetostrictive phases by shear stresses in their sample is relatively weak, restraining the output of ME voltage.

Recently, ME composites with complex structures such as cylindrical or toroidal configurations have attracted increasing attention due to their inherent unique properties including low shape-induced anisotropy, self-bound effect, improved interface mechanical coupling and closed magnetic circuit. Zhang et al. developed a tri-layer toroidal ME gyrator consisting of circumferential magnetized ferrite and transversely polarized piezoelectrics and realized excellent non-reciprocal voltage-current and bidirectional impedance conversion properties [5]. Savelev et al. proposed a new ring-type of ME inductors with inductance tunability up to 412% by an electric field or 1270% by a magnetic field [6]. Very recently, we reported giant circumferential ME effect at a low optimum bias field in PZT/Mn-Znferrite cylindrical composite with both dc bias and ac magnetic fields along circumferential direction [7]. In this work, we will present strong circumferential ME effect in PZT/Terfenol-D cylindrical composite with ac circumferential excitation and transverse bias (dc bias field parallel or perpendicular to the axis of the cylinder), which is expected to find potential application as a vortex magnetic field sensor.

PZT and Terfenol-D were adopted as piezoelectric and magnetostrictive phases, respectively. The commercially purchased PZT ring with Ag electrodes pasted on the top and bottom planes was polarized along the thickness direction and had dimensions of inner diameter of 15.0 mm, out diameter of 26.0 mm, and thickness of 3.0 mm. The inner surface of the PZT ring was polished slowly by

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Figure 1. Bias filed dependence of the circumferential ME voltage coefficient α_{ME} at f = 1 kHz for C–A (bias parallel to the axis of the cylinder) and C–V modes (bias perpendicular to the axis of the cylinder), respectively. Inset shows the schematic diagram of circumferential ME effect measurement, where gray part shows the PZT ring and white part shows Terfenol-D ring.

sandpaper to attain good size matching between Terfenol-D and PZT rings. The commercially purchased Terfenol-D ring with inner diameter of 8.2 mm was inserted into the PZT ring to form the PZT/Terfenol-D bilayered cylindrical composite. The air gap between PZT and Terfenol-D rings was filled with insulate adhesive. For the ME effect measurement, a harmonic voltage of frequency 1 kHz generated form signal generator was applied to the coil wound around the cylindrical composite with 200 turns to produce ac circumferential magnetic field, which was measured using ac Gauss meter for a counterpart coil with the same input voltage. The tunable dc bias magnetic field generated by Helmholtz coils was applied parallel or perpendicular to the axis of the cylinder. To measure the frequency response of ME effect, a lock-in amplifier (Zurich Instruments, UHF-DEV2031) was used to provide the harmonic voltage with frequency $f = 1 - 100 \,\text{kHz}$ and magnitude U = 1.5 V, and the induced voltage was recorded by the lock-in amplifier with input impedance of $1 M\Omega$. The configuration of the sample and schematic diagram of circumferential ME effect measurement is shown in the inset of Fig. 1.

Fig. 1 shows dependence of the circumferential ME voltage coefficient α_{ME} ($\alpha_{ME} = V_{ME}/t_pH_{ac}$, t_p is thickness of PZT) on the dc bias magnetic field H_{dc} at f = 1 kHz for C–A (bias parallel to axial direction) and C–V modes (bias perpendicular to axial direction), respectively. It can be seen that the H_{dc} dependence of α_{ME} is similar with that in planar ME composites or $\alpha_{E,A}$ and $\alpha_{E,V}$ in cylindrical composites, which is related to the bias field dependent piezomagnetic coefficient of magnetostrictive phase [8]. The difference of the optimum bias field for the two modes, around 1500 and 304 Oe for C–A and C–V mode, respec-

tively, could be attributed to the effect of demagnetization factor [8]. The circumferential ME coefficient at optimum bias field in our cylindrical ME composite at low frequency reaches $0.93 \text{ V} \cdot \text{cm}^{-1} \cdot \text{Oe}^{-1}$, which is much higher than that in ring-type ME laminate [4] (where the maximum of their defined ME voltage coefficient is about $0.26 \text{ V} \cdot \text{Oe}^{-1}$, but the effective thickness of PZT is much larger due to the sample's circumferential polarization direction). The enhanced ME effect can be attributed to improved interface mechanical coupling and self-bound effect due to cylindrical configuration. It is believed that the obtained ME effect could be further enhanced if the gap between PZT and Terfenol-D rings is minimized. The high coefficient indicates that the cylindrical PZT/Terfenol-D composite has a highvoltage sensitivity to small H_{ac} .

Recently, nonlinear ME effect was observed in a heterostructure containing layers of amorphous ferromagnet FeBSiC and PZT when the excitation ac field was directed perpendicular to the bias dc field [10]. In this case of transverse excitation, only even voltage harmonics are generated, and the second harmonic amplitude quadratically depends on the ac excitation field amplitude. It was suggested that the total field change by $\delta H \approx H_{\rm ac}^2/2H_{\rm dc}$ is responsible for the dependence of the second harmonic amplitude on field H_{dc} and excitation field H_{ac} . In contrast, circumferential ME effect with transverse bias magnetic field in the cylindrical composite reported here is a linear effect (see below). In some previous works by other researchers, the linear ME effect under transverse excitation was also observed in layered ferromagnetic-piezoelectric composites [11-13]. The linear transverse ME effect can be understood by the basic principles of magnetostriction of polycrystalline ferromagnetic materials [14]. In general, magnetostrictive materials have a preferred direction in which the magnetostrictive coefficient is relatively large. As they are polycrystals, the materials in other directions will also be magnetostrictive. Once dc magnetic field is applied, extension in one direction usually results in a contraction in the other direction. Nevertheless, there are also simultaneous stretching in all directions for some materials, only varying in size. The magnetostrictive properties of the magnetostrictive material will be affected regardless of which direction the bias magnetic field is applied in. Usually, the magnetostrictive effect is strongest when the dc field takes a certain optimum value. Under the ac magnetic field, magnetostriction occurs in all directions with different phases. Consequently, the optimum bias field for maximum ME coefficient will vary with the angle of bias magnetic field to ac magnetic field. We note that in Ref. 10 the amplitude of ac magnetic field is 2 Oe, comparable to dc bias field $H_{\rm dc} \approx 11$ Oe, which may be the main origin for observed nonlinear ME effect. While the amplitude of ac magnetic field is far less than dc bias filed in our experiment, only linear ME effect can be observed.

In our previous work, a theoretical model, based on constitutive equations for piezoelectric phase and magnetostrictive phase and equation of motion for continuous medium, was proposed to describe the circumferential ME effect under circumferential excitation in cylindrical composites [7]. The ME voltage coefficient in cylindrical ME composite can be estimated by

$$\alpha_{\rm ME} = Ad_{mn}(q_{31} - q_{11}), \tag{1}$$

where A is a constant, depending on geometric parameters and material parameters of the structure; d_{mn} is piezoelectric coefficient of piezoelectric phase, and $(q_{31}-q_{11})$ is piezomagnetic coefficient of magnetostrictive phase. In previous investigation, the dc bias magnetic field is applied parallel or perpendicular to the axis of the disk-ring or cylindrical composites, superimposed with ac magnetic field in the same direction. The generated ME effects for axial or vertical modes are dominated by piezomagnetic coefficient q_{31} and $q_{11} + q_{12}$, respectively. As $q_{11} + q_{12}$ is usually much larger than q_{31} due to the effect of demagnetization factor, $\alpha_{\rm EV}$ for vertical mode is accordingly larger than $\alpha_{\rm EA}$ for axial mode [15,16]. However, in our investigation the circumferential ME effect for C-A mode is stronger than that for C-V mode at their respective optimum bias fields. The underlying mechanism of this observation is speculated to result from H_{dc} -orientation-dependent dynamic piezomagnetic strain [17]. As indicated in Eq. (1), the ME effect in cylindrical composites under circumferential excitation is determined by piezomagnetic coefficient $(q_{31}-q_{11})$, whether the bias field H_{dc} is parallel or perpendicular to the axial direction. Upon H_{dc} with different orientation, the magnetic domain rotation and demagnetization effect cause the effective magnetic induction B_{μ} to vary, resulting in an anisotropic dynamic strain of magnetostrictive phase. $(q_{31}-q_{11})$ reaches maximum at their respective optimum bias filed for the two modes. However, for C-A mode, the bias field H_{dc} is always perpendicular to the plane of the structure as well as the excitation field $H_{\rm ac}$, leading to different parts of magnetostrictive phase being in the same magnetostrictive state and thus a relatively large effective piezomagnetic coefficient. In contrast, the internal field H_{in} differs greatly in different parts of magnetostrictive phase due to demagnetization, and the angle between H_{dc} and $H_{\rm ac}$ also varies greatly with position in the plane for C-V mode, resulting a weaker effective piezomagnetic coefficient $(q_{31}-q_{11})$ than that for C–A mode, which could be responsible for the stronger circumferential ME effect for C-A mode than that for C-V mode.

Fig. 2 gives the measured frequency response of α_{ME} for C–A mode at optimum $H_{dc} = 1500 \text{ Oe}$ and C–V mode at optimum $H_{dc} = 304 \text{ Oe}$. The obtained resonant α_{ME} for C–A mode reaches the value as high as about $32 \text{ V} \cdot \text{cm}^{-1} \cdot \text{Oe}^{-1}$. It is found that the resonance frequencies for C–A and C–V modes at their respective optimum bias fields are very close, corresponding to the radial vibration mode at electromechanical resonance (EMR) region. The resonance frequency of radial vibration mode



Figure 2. Frequency response of α_{ME} for C–A mode at optimum $H_{dc} = 1500$ Oe and C–V mode at optimum $H_{dc} = 304$ Oe. Inset shows the induced ME voltage V_{ME} as a function of applied H_{ac} amplitude for C–A mode with optimum bias field $H_{dc} = 1500$ Oe at low frequency f = 1 kHz and resonance frequency $f_r = 46$ kHz, respectively.

for the cylindrical composite is given by [16]

$$f_r = \frac{1}{\pi D} \sqrt{\frac{1}{\bar{\rho}\bar{s}_{11}}},\tag{2}$$

where *D* is average diameter, $\bar{\rho}$ is average mass density. \bar{s}_{11} is equivalent compliance coefficient, which can be obtained by

$$\bar{s}_{11} = \frac{p_{S_{11}}m_{S_{11}}}{\nu_m p_{S_{11}} + \nu_p m_{S_{11}}},$$

where v_p and v_m are volume fractions of piezoelectric and magnetostrictive phases, respectively, ${}^{p}s_{11}$ and ${}^{m}s_{11}$ are compliance coefficients of piezoelectric and magnetostrictive phases, respectively. Calculations were performed using the following material parameters: ${}^{p}s_{11} = 15.3 \cdot 10^{-12} \text{ m}^2/\text{N}$, ${}^{p}\rho = 7.8 \cdot 10^3 \text{ kg/m}^3$ (for PZT), ${}^{m}s_{11} = 125 \cdot 10^{-12} \text{ m}^2/\text{N}$, ${}^{m}\rho = 9.2 \cdot 10^3 \text{ kg/m}^3$ (for Terfenol-D) [18,19]. The volume fraction of PZT is about 0.74. The calculated resonance frequency is 46.3 kHz, in good agreement with the experimental value $f_r = 46.0 \text{ kHz}$.

The linear response of output voltage to applied field amplitude is an important parameter for the application of magnetic sensor. The inset of Fig. 2 shows the measured ME voltage as a function of applied H_{ac} in the magnetic field range $10^{-8} < 10^{-5}$ T. The measurement was performed at room temperature and without magnetic shielding. The ME voltage is found to be a linear response to applied ac magnetic field over this range both at low frequency and resonance frequency, which indicates that the cylindrical PZT/Terfenol-D laminate composite has a high sensitivity to small ac vortex magnetic field change.



Figure 3. Induced ME voltage response over low frequency range from 1 Hz to 6 kHz upon constant ac magnetic field $H_{ac} = 0.1$ Oe for C–A and C–V modes, respectively.

Fig. 3 presents the frequency response of ME voltage over the range from 1 Hz to 6 kHz upon constant amplitude $H_{ac} = 0.1$ Oe for C–A and C–V modes, respectively. The induced voltage can be seen to be nearly independent of input ac magnetic field frequency in the frequency range from Hz to kHz, which is advantageous to its application as vortex magnetic field sensor detecting ac magnetic field with low and unstable frequency. While for ac magnetic field sensor based on a reluctance coil, the induced voltage is strongly dependent on frequency, so the signal could not be detected by the coil due to small flux change rate through the coil at low frequency condition.

Giant circumferential ME effect has been reported in PZT/Terfenol-D bilayered cylindrical composite with bias field parallel or perpendicular to the axis of the cylinder and ac magnetic field along circumferential direction. The ME voltage has a high sensitivity of 10^{-8} T to a vortex magnetic field. It is believed that the sensitivity limit could be achieved to $10^{-11} \sim 10^{-12}\,T$ if more accurate device is adopted to produce and measure small ac voltage signal and the size matching between PZT and Terfenol-D rings can be improved. The ME voltage has a linear response to ac magnetic field amplitude in the magnetic field range of $10^{-8} \sim 10^{-5} \,\mathrm{T}$ and is independent on ac magnetic field frequency at low frequency range from Hz to kHz. The upper limit of the operational frequency range is expected to be enhanced to hundreds of kHz by reducing the diameter of the cylindrical ME composite, which is favorable for high power application. All these advantages, combined with the distinguishing feature of easy preparation, make cylindrical PZT/Terfenol-D laminate composite have a bright prospect of application in sensitive vortex magnetic field sensors.

Financial support

This work is financially supported by Priority Academic Program Development of Jiangsu Higher Education Institution (PAPD) and the National Natural Science Foundation of China under Grant N 51277098.

Conflict of interests

The authors declare that they have no conflict of interests.

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