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MAGNETISM

Direct Magnetoelectric Effect in Two-Layer Composite Structures $Tb_{0.12}Dy_{0.2}Fe_{0.68}$–$PbZr_{0.53}Ti_{0.47}O_3$ at Bending and Longitudinal Vibrations

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Abstract—The direct magnetoelectric effect has been studied in samples of two-layer composites containing $8 \times 6 \times 0.3$-mm layers of the piezoelectric material $PbZr_{0.53}Ti_{0.47}O_3$ and $6 \times 6 \times A$-mm layers ($A = 0.3$, 0.6, 0.9, 1.2, and 1.5) of the ferromagnet $Tb_{0.12}Dy_{0.2}Fe_{0.68}$ and epoxy adhesive in the frequency range of 10–253 kHz at room temperature. It has been found that the magnetoelectric effect significantly increases at resonance frequencies (13.2–61.1 kHz) of the first harmonic of bending vibrations along the sample length, at resonance frequencies (39.5–90.7 kHz) of the first harmonic of bending vibrations along the sample width, and at resonance frequencies (123.3–141.0 kHz) of the first harmonic of longitudinal vibrations along the sample length. The magnetoelectric effect magnitudes at the resonance frequencies of the bending vibrations is found to be greater than that at the resonance frequencies of the longitudinal vibrations of the sample.

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1. INTRODUCTION

In the last decade, magnetoelectric composites have attracted more and more attention of researchers due to perspectives of their application in devices in which magnetic field is used to control electrical parameters (direct magnetoelectric effect) and external electric field is used to control magnetic parameters (inverse magnetoelectric effect) [1]. It is natural that, for application in practice, of interest are composite structures with high magnetoelectric coefficients. Because the magnetoelectric effect in composites is due to the coupling between the magnetostrictive and piezoelectric subsystems through the elastic strains, the magnetoelectric coefficients sharply increase near the electromechanical resonance [2]. When longitudinal vibration modes are excited in magnetoelectric composite samples, the resonance frequencies become very high and reach several hundred kilohertz in samples with a length of approximately 10 mm. These resonance frequencies can be unusable in designing composite-based magnetoelectric devices operating at lower frequencies. In order to decrease frequencies of the electromechanical resonance of samples with specified sizes or to retain the same frequencies with decreasing composite sample size for miniaturization of electronic devices, it is common practice to use bending vibration modes [3]. In this case, when longitudinal and bending vibrations are excited in composite samples, the magnetoelectric coefficients can be different. For example, the magnetoelectric coefficient of the two-layer composite $Tb_{0.30}Dy_{0.7}Fe_2$–$PbZr_{0.52}Ti_{0.48}O_3$ is higher upon resonance at the frequency of the first harmonic of longitudinal vibrations than that at the frequency of the first harmonic of bending vibrations [4]. At the same time, according to [5], similar composites demonstrate the opposite picture: at the frequency of the first harmonic of bending vibrations, the magnetoelectric coefficient is higher than that at the frequency of the first harmonic of longitudinal vibrations.

Therefore, the aim of this work is to perform a comparative analysis of the direct magnetoelectric effect in two-layer $Tb_{0.12}Dy_{0.2}Fe_{0.68}$–$PbZr_{0.53}Ti_{0.47}O_3$ composite samples, in which longitudinal and bending vibrations are excited.

2. SAMPLE PREPARATION AND EXPERIMENTAL TECHNIQUE

We studied layered magnetoelectric composites owing to the unique combination of their high magnetoelectric coefficients and the simplicity of fabrication, which is not characteristic of mixture-type and rod-type magnetoelectric composites. The composite components were the $Tb_{0.12}Dy_{0.2}Fe_{0.68}$ (TDF) ferromagnet and $PbZr_{0.53}Ti_{0.47}O_3$ (PZT) piezoelectric material with an extremely high saturation magnetostriction ($\lambda_s \sim 10^{-3}$) and high piezoelectric coefficients (for example, $d_{31} \sim 140$ pC/N), respectively. There-

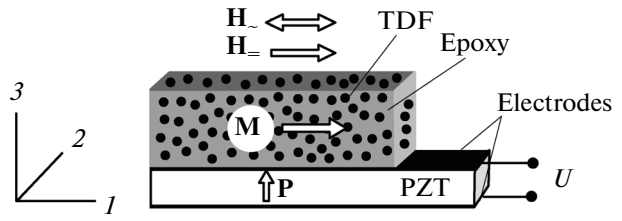

Fig. 1. Schematic drawing of the TDF–PZT composite sample and its orientation in ac and dc magnetic fields.

fore, we can expect high magnetoelectric responses in samples of TDF–PZT composites.

To obtain samples of two-layer TDF–PZT composites, we prepared a statistical mixture from the carefully mixed TDF ferromagnet powder with an average grain size of ~54 μm and a mass of 1.66 g with the epoxy adhesive 0.36 g in mass, which was deposited as ferromagnetic layers of several sizes on piezoceramic PZT plates that were preliminarily polarized in industrial conditions. The ferromagnetic layers of the composite samples were solidified for 24 h at room temperature. Then, using an abrasive paper, the ferromagnetic layers were brought to sizes $6 \times 6 \times A$ mm ($A = 0.3$, 0.6, 0.9, 1.2, and 1.5). The PZT plates have sizes $8 \times 6 \times 0.3$ mm. The ferromagnetic layers were magnetized along their lengths, and the piezoelectric layers were polarized along the thickness. The structure of the TDF–PZT composite sample is shown in Fig. 1.

The cantilever mount of the samples provided the lowest resonance frequencies of bending vibrations [6].

The experimental study of the direct magnetoelectric effect was carried out by measuring the ac voltage generated on the electrodes of the piezoelectric layer of the TDF–PZT composite sample when the sample was placed into ac and dc magnetic fields. The ac magnetic field with strength $H_{\sim} = 5$ Oe was induced by the Helmholtz coils, and the dc magnetic field $H_{=} =$ 720 Oe was obtained using an electromagnet. Strength 720 Oe corresponds to the strength of dc magnetic field at which piezomagnetic coupling coefficient $q_{11} = d\lambda_{11}/dH_{=}$ proportional to the value of the magnetoelectric effect becomes maximal. Because, in

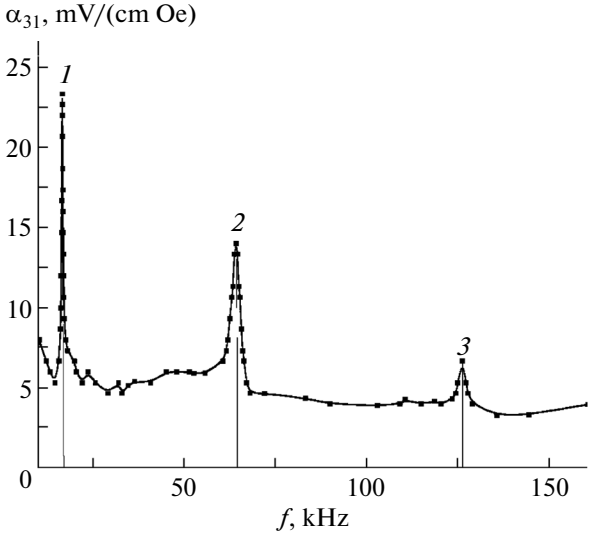

Fig. 2. Frequency dependence of the coefficient α_{31} for the 0.9TDF–0.3PZT composite sample.

composites, the magnetoelectric effect at the perpendicular orientation of magnetization vector **M** and polarization vector **P** (transverse magnetoelectric effect) is higher than the magnetoelectric effect at the parallel orientation of vectors **M** and **P** (longitudinal magnetoelectric effect) [7, 8], we studied in this work the transverse magnetoelectric effect.

The transverse magnetoelectric effect is characterized by the magnetoelectric voltage coefficient calculated according to the formula

$$\alpha_{31} = \frac{U}{h_{\mathrm{PZT}} H_{\sim}}, \qquad (1)$$

where h_{PZT} is the thickness of the piezoelectric layer of the TDF–PZT composite sample.

3. RESULTS AND DISCUSSION

Figure 2 shows the dependence of coefficient α_{31} at room temperature on the frequency f of the ac magnetic field for the sample of the two-layer composite with the 0.9-mm-thick ferromagnetic and 0.3-mm-

Table 1. Transverse magnetoelectric voltage coefficients and corresponding resonance frequencies for different samples of the TDF–PZT composites

Composite sample	$\alpha_{31(1)}$, mV/(cm Oe)	f_{r1}, kHz	$\alpha_{31(2)}$, mV/(cm Oe)	f_{r2}, kHz	$\alpha_{31(3)}$, mV/(cm Oe)	f_{r3}, kHz
0.3TDF–0.3PZT	16.0	13.2	10.0	39.5	5.9	141.0
0.6TDF–0.3PZT	18.0	15.5	14.0	51.1	8.6	139.7
0.9TDF–0.3PZT	23.3	15.7	14.0	64.2	6.7	125.6
1.2TDF–0.3PZT	20.0	19.8	12.0	87.8	7.3	216.4
1.5TDF–0.3PZT	12.0	61.1	9.3	90.7	7.0	123.3

thick piezoelectric layers (in what follows, 0.9TDF–0.3PZT).

We can see that coefficient α_{31} as a function of f passes through three peaks (*1–3*) corresponding to the electromechanical resonance frequencies f_{r1}, f_{r2}, and f_{r3} for the 0.9TDF–0.3PZT composite sample at different types and/or vibration harmonics. Similar dependences were obtained for composites 0.3TDF–0.3PZT, 0.6TDF–0.3PZT, 1.2TDF–0.3PZT, and 1.5TDF–0.3PZT. The coefficients $\alpha_{31(1)}$, $\alpha_{31(2)}$, and $\alpha_{31(3)}$ and corresponding frequencies f_{r1}, f_{r2}, and f_{r3} are given in Table 1 for all composite samples.

To determine the type and harmonic of vibrations excited in the TDF–PZT composite samples at frequencies f_{r1}, f_{r2}, and f_{r3}, we calculated the resonance frequencies of the composite samples in the cases of bending and longitudinal vibrations. The resonance frequencies of the bending and longitudinal vibrations along the sample length were found, respectively, by formulas [4]

$$f_{\delta n}^{\mathrm{BL}} = \frac{\pi h}{2\sqrt{12L^2}}\frac{1}{\sqrt{\langle\rho\rangle\langle s_{11}\rangle}}\left(n+\frac{1}{2}\right)^2, \quad (2)$$

$$f_{\delta n}^{\mathrm{LL}} = \frac{n}{2L}\frac{1}{\sqrt{\langle\rho\rangle\langle s_{11}\rangle}}, \quad (3)$$

where h is the thickness of the TDF–PZT composite sample; $n = 1$, 2, and 3 are the numbers of nodes along the sample for the first, second, and third vibration harmonics; L is the sample length (the sample length was taken to be equal to the PZT layer length); $\langle\rho\rangle = \rho_E V_E + \rho_{\mathrm{TDF}} V_{\mathrm{TDF}} + \rho_{\mathrm{PZT}} V_{\mathrm{PZT}}$ is the effective density of the TDF–PZT composite; ρ_E, ρ_{TDF}, and ρ_{PZT} are the densities of the epoxy adhesive, ferromagnet TDF, and piezoelectric PZT, respectively; $V_E = m_E/(m_{\mathrm{TDF}} + m_E)$, $V_{\mathrm{TDF}} = m_{\mathrm{TDF}}/(m_{\mathrm{TDF}} + m_E)$, and $V_{\mathrm{PZT}} = h_{\mathrm{PZT}}/(h_{\mathrm{TDF}} + h_{\mathrm{PZT}})$ are the volume contents of epoxy adhesive, TDF ferromagnetic powder, and PZT piezoceramic in the TDF–PZT composite, respectively; m_E is the epoxy adhesive mass; m_{TDF} is the TDF ferromagnetic powder mass; and h_{TDF} is the ferromagnetic layer thickness in the sample.

In Eqs. (2) and (3), parameter $\langle s_{11}\rangle$ is the effective elastic compliance of the TDF–PZT composite calculated by formula [9]

$$\langle s_{11}\rangle = \frac{s_{11}^{E/\mathrm{TDF}} s_{33}^{\mathrm{PZT}}}{V_{E/\mathrm{TDF}} s_{33}^{\mathrm{PZT}} + V_{\mathrm{PZD}} s_{11}^{E/\mathrm{TDF}}}, \quad (4)$$

where

$$\langle s_{11}^{E/\mathrm{TDF}}\rangle \geq \frac{s_{11}^{E} s_{11}^{\mathrm{TDF}}}{V_E s_{11}^{\mathrm{TDF}} + V_{\mathrm{TDF}} s_{11}^{E}}, \quad (5)$$

$s_{11}^{E/\mathrm{TDF}}$ is the effective elastic compliance of the ferromagnetic powder with epoxy adhesive; s_{11}^{E} is the elastic

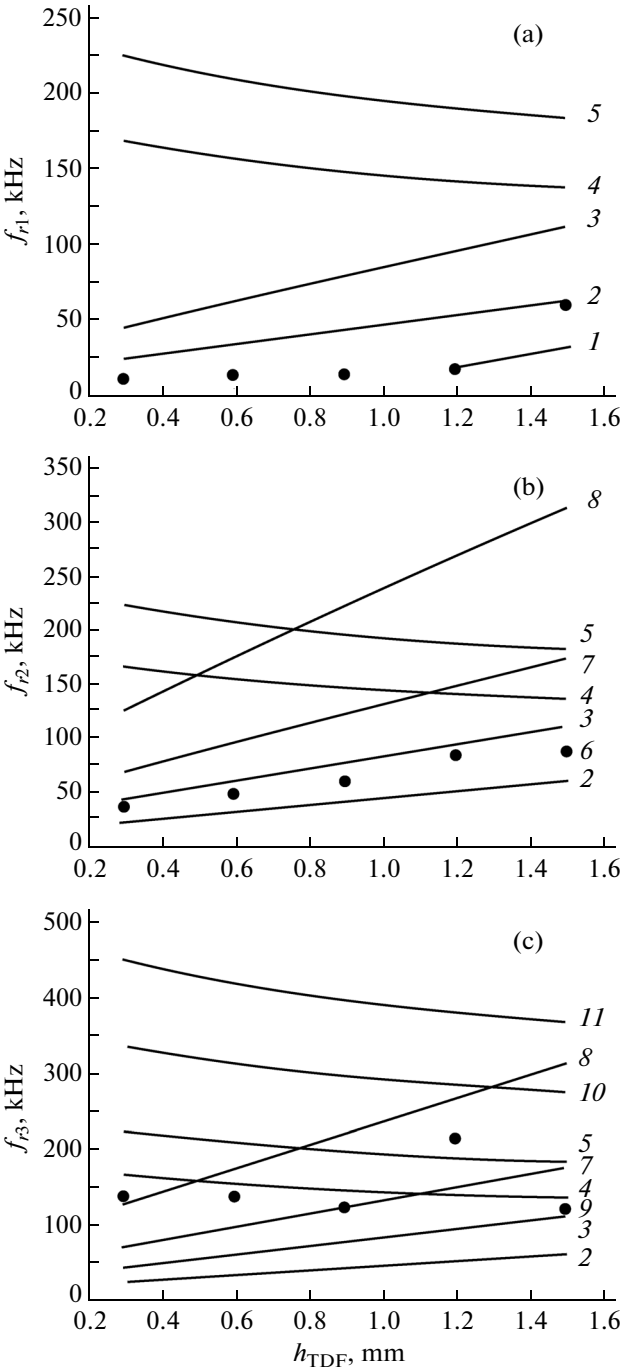

Fig. 3. Dependences of the resonance frequencies of the bending and longitudinal vibrations of the TDF–PZT composite samples on the ferromagnetic layer thickness. Points are the experimental data, and solid lines show the calculated data. (*1*, *6*, *9*) Resonance frequencies corresponding to peaks *1*, *2*, and *3* in the dependence $\alpha_{31}(f)$, respectively; (*2*, *7*) resonance frequencies of the first and second harmonics of the bending vibrations along the sample length, respectively; (*3*, *8*) resonance frequencies of the first and second harmonics of the bending vibrations along the sample width, respectively; (*4*, *10*) resonance frequencies of the first and second harmonics of the longitudinal vibrations along the sample length, respectively; and (*5*, *11*) resonance frequencies of the first and second harmonics of the longitudinal vibrations along the sample width, respectively.

Table 2. Parameters of the TDF–PZT composite components

TDF–PZT composite components	$\rho \times 10^{-3}$, kg/m^3	$s_{11} \times 10^{12}$, m^2/N	$s_{33} \times 10^{12}$, m^2/N
TDF	4.0	78	–
PZT	7.1	–	15
Epoxy adhesive	1.2	0.00033	–

compliance of epoxy adhesive; s_{11}^{TDF} is the elastic compliance of the TDF ferromagnet; s_{11}^{PZT} is the elastic compliance of the PZT piezoelectric; and $V_{E/\mathrm{TDF}}$ is the volume content of the ferromagnetic powder with epoxy adhesive in the TDF–PZT composite.

Note that, when determining the resonance frequencies of the bending and longitudinal vibrations along the sample widths of the TDF–PZT composites, sample length L was replaced by their width W in Eqs. (2) and (3).

Using Eqs. (2) and (3), the geometric sizes, and the parameters of the components of the TDF–PZT composites listed in Table 2, we plotted the dependences of the resonance frequencies of the bending and longitudinal vibrations of the samples on the ferromagnetic layer thickness (Fig. 3).

It is seen that resonance frequency f_{r1} corresponds to the first harmonic of the bending vibrations along the sample length (Fig. 3a), resonance frequency f_{r2} corresponds to the first harmonic of the bending vibrations along the sample width (Fig. 3b), and resonance frequency f_{r3} corresponds to the first harmonic of the longitudinal vibrations along the sample length (Fig. 3c). Frequencies of the bending vibration mode f_{r1} and f_{r2} increase and frequency of the longitudinal vibration mode f_{r3} decreases with an increase in the ferromagnetic layer thickness of the samples. The increase in frequencies f_{r1} and f_{r2} is due to the competition of two factors: the increase in sample thickness h and in product of effective density $\langle\rho\rangle$ by the effective compliance of the composite $\langle s_{11}\rangle$ (Eq. (2)). The decrease in frequency f_{r3} is due to the increase in $\langle\rho\rangle\langle s_{11}\rangle$ (Eq. (3)).

4. CONCLUSIONS

Thus, based on the data of Table 1 and Fig. 3, we can conclude that magnetoelectric responses in the TDF–PZT composite samples at the resonance frequencies of bending vibrations are higher than those at the resonance frequencies of longitudinal vibrations. The lower resonance frequencies of the bending vibrations in the combination with high magnetoelectric responses make the bending vibration modes more preferable from the standpoint of practical applications of the TDF–PZT composites in devices using the magnetoelectric effect.

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