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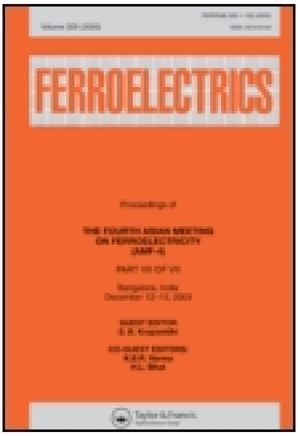

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Magnetoelectric Properties of Two-Layered Composites $Tb_{0.12}Dy_{0.2}Fe_{0.68}$ - $PbZr_{0.53}Ti_{0.47}O_3$

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Magnetoelectric Properties of Two-Layered Composites $Tb_{0.12}Dy_{0.2}Fe_{0.68} – PbZr_{0.53}Ti_{0.47}O_3$

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At frequencies of the measuring magnetic field of 9.9–253 kHz over a temperature range from 79 K to 289 K the magnetoelectric effect in two-layered ferromagnetic-piezoelectric composites $Tb_{0.12}Dy_{0.2}Fe_{0.68} – PbZr_{0.53}Ti_{0.47}O_3$ with ferromagnetic $Tb_{0.12}Dy_{0.2}Fe_{0.68}$ and piezoelectric $PbZr_{0.53}Ti_{0.47}O_3$ layers of $6 \times 6 \times A$ mm^3 (A = 0.6, 0.9, and 1.2) and $8 \times 6 \times 0.3$ mm^3 in size, respectively, has experimentally been studied. Resonant frequencies of bending and longitudinal oscillations of composite samples of $Tb_{0.12}Dy_{0.2}Fe_{0.68}$ – $PbZr_{0.53}Ti_{0.47}O_3$ have been calculated. We have studied dependences of the magnetoelectric effect in composites on a frequency of the measuring magnetic field, a thickness of a $Tb_{0.12}Dy_{0.2}Fe_{0.68}$ layer, the bias magnetic field strength, and temperature.

Keywords Magnetoelectric effect; two-layered composite; lead zirconate titanate; terfenol-D

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1. Introduction

Today magnetoelectric (ME) materials, possessing simultaneously electrical and magnetic orderings, are the focus for modern materials science as a possibility to vary electrical properties of these materials by the magnetic field (the direct ME effect) and magnetic properties by the electric field (the inverse ME effect) [1] gives an opportunity to create a new generation of microelectronic devices [2].

Among well-known ME materials, the most attractive ME materials are ME composites which in comparison with single-phase ferroelectromagnetic media demonstrate strong ME interactions and the ME effect exists at temperatures higher room temperature [3]. Investigators generally distinguish three groups of ME composites: layer, bulk, and rod ones. Layered ME composites, thanks to the absence in them of the mutual phase doping as well as to a possibility to polarize piezoceramic plates before the manufacture of composites, have better ME properties in contrast to bulk ME composites [4]. Besides, layered ME composites as compared with rod ME composites are easier to manufacture. That is why this work is devoted to layer ME composites.

Most of layered ME composites are alternating layers of magnetostriction and piezoelectric materials stuck together with the epoxy [5–7]. For raising the efficiency of ME interactions in such composites, composite components must have large magnetostrictive and piezoelectric coefficients [3]. Therefore, in the present work magnetostrictive layers were made of the terfenol-D $Tb_{0.12}Dy_{0.2}Fe_{0.68}$ (TDF) and piezoelectric layers were made

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Table 1
Properties of TDF and PZT materials

Material	$s_{11} \cdot 10^{12}$, m^2/N	$\lambda_{11} \cdot 10^6$ at $H_= = 720$ Oe	ε_{33} at f = 1 kHz	$d_{31} \cdot 10^{12}$, C/N	σ, $Ohm^{-1} \cdot m^{-1}$	$\rho \cdot 10^{-3}$, kg/m^3
TDF	78	25	—	—	$4.9 \cdot 10^{-9}$	4.0
PZT	15	—	1495	150	$3.9 \cdot 10^{-3}$	7.1

Notations: s_{11} is the elastic compliance, ε_{33} is the dielectric permittivity, λ_{11} is the magnetostriction coefficient at an inflection point on the dependence $\lambda_{11}(H_=)$, d_{31} is the piezoelectric modulus, σ is the electrical conductivity, and ρ is the density.

of the lead zirconate titanate (PZT). However, TDF is very brittle and conductive that leads to the large eddy current losses. To overcome above a disadvantage, bulk TDF was mixed with the epoxy. A particulate composite based on TDF and the epoxy was applied to piezoelectric plates $PbZr_{0.53}Ti_{0.47}O_3$. We have studied ME properties of manufactured two-layered composites $Tb_{0.12}Dy_{0.2}Fe_{0.68}$ – $PbZr_{0.53}Ti_{0.47}O_3$ (further TDF – PZT) with various a thickness of the $Tb_{0.12}Dy_{0.2}Fe_{0.68}$ layer over a wide frequency range of the measuring magnetic field, strengths of the bias magnetic field, and temperatures.

2. Experimental

Magnetoelectric composites $Tb_{0.12}Dy_{0.2}Fe_{0.68}$ – $PbZr_{0.53}Ti_{0.47}O_3$ were manufactured using carefully mixed the TDF powder with an average particle size of ~54 μm and mass of 1.66 g. and the epoxy with mass of 0.36 g. A stiff mixture consistency was applied in the form of magnetic layers to piezoelectric PZT plates pre-polarized in industrial conditions. For 24 h the magnetic layer had cured. In fact, the magnetic layer represents a particulate composite consisting of an epoxy matrix with a filler in the form of TDF particles randomly distributed in the matrix.

Composites, used for ME studies, have contained plane-parallel layers of the ferromagnetic material on basis of TDF with thicknesses of 0.6 mm, 0.9 mm, and 1.2 mm and the piezoelectric ceramics PZT with a thickness of 0.3 mm. A length to width ratio for TDF plates has been 6 mm : 6 mm and that for PZT plates has been 8 mm : 6 mm.

Mechanical, dielectric, piezoelectric, and electrical parameters of composite components TDF and PZT at room temperature are given in Table 1.

A structure of two-layered ME composite samples of TDF – PZT is displayed in Fig. 1.

The ME effect has been studied by measuring a voltage generating across a sample under both measuring and bias magnetic fields whose directions are shown in Fig. 1. The bias magnetic field was generated by an electromagnet while the measuring magnetic field

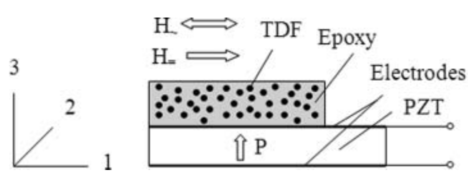

Figure 1. Model of composite samples of TDF – PZT and its direction in measuring $H_\sim$ and bias $H_=$ magnetic fields.

was produced using Helmholtz coils. A value of the measuring magnetic field strength was 5 Oe. A choice of the transverse ME effect, when $H_{\sim}$ и $H_{=}$ are directed perpendicular to a direction of the polarization P in a PZT layer (Fig. 1), is due to the fact that a value of the ME response at $H_{=} \perp P$ exceeds a value of the ME response at $H_{=} \| P$ [8].

In order to characterize the transverse ME effect, we have used a ME voltage coefficient

$$\alpha_{31} = \frac{U}{b \cdot \mathrm{H}_{\sim}}, \tag{1}$$

where b is a thickness of the piezoelectric layer of PZT.

3. Results and discussion

Results of the transverse ME effect study in a two-layered composite consisting of the ferromagnetic layer of TDF with a thickness of 0.6 mm and the piezoelectric layer PZT with a thickness of 0.3 mm (further the 0.6 TDF – 0.3 PZT) at the bias magnetic field strength of 720 Oe and the measuring magnetic field of 5 Oe over an frequency interval of 10.1–175.2 kHz are shown in Fig. 2. In the dependence $\alpha_{31}(f)$ we can see three peaks at frequencies f_{r1}, f_{r2}, and f_{r3} which correspond to electromechanical resonance frequencies of the composite at its different types of oscillations. Similar dependences $\alpha_{31}(f)$ have been observed for composites 1.2 TDF – 0.3 PZT and 0.9 TDF – 0.3 PZT. Values of f_{r1}, f_{r2}, and f_{r3} and corresponding to them values of $\alpha_{31(1)}$, $\alpha_{31(2)}$, and $\alpha_{31(3)}$ for composites 0.6 TDF – 0.3 PZT, 0.9 TDF – 0.3 PZT, and 1.2 TDF – 0.3 PZT are presented in Table 2.

To reveal a type of composite oscillations at f_{r1}, f_{r2}, and f_{r3}, resonant frequencies for bending and longitudinal oscillations have been estimated using respectively equations

$$f_{pn}^{B} = \frac{\pi h}{2\sqrt{12}L^2} \cdot \frac{1}{\sqrt{<\rho> \cdot <s_{11}>}} \cdot \left(n + \frac{1}{2}\right)^2 \tag{2}$$

and

$$f_{pn}^{L} = \frac{n}{2L} \cdot \frac{1}{\sqrt{<\rho> \cdot <s_{11}>}}. \tag{3}$$

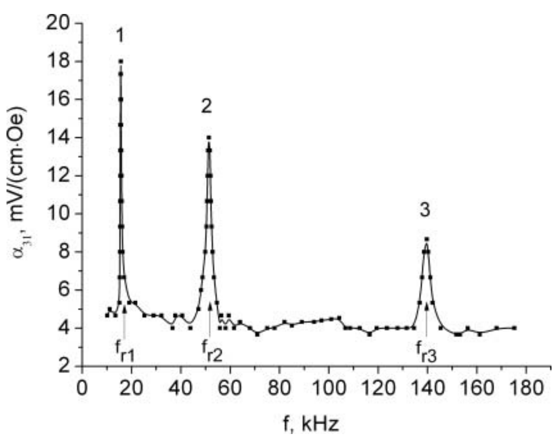

Figure 2. Frequency dependence of α_{31} for the ME composite 0.6 TDF – 0.3 PZT at room temperature.

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Table 2
Resonant frequencies f_{r1}, f_{r2}, and f_{r3} and corresponding to them $\alpha_{31(1)}$, $\alpha_{31(2)}$, and $\alpha_{31(3)}$ for various composites

Composite	f_{r1}, kHz	f_{r2}, kHz	f_{r3}, kHz	$\alpha_{31(1)}$, mV/(cm·Oe)	$\alpha_{31(2)}$, mV/(cm·Oe)	$\alpha_{31(3)}$, mV/(cm·Oe)
0.6 TDF – 0.3 PZT	15.5	51.1	139.7	18.0	14.0	8.6
0.9 TDF – 0.3 PZT	15.7	64.2	125.6	23.4	14.0	6.7
1.2 TDF – 0.3 PZT	19.8	87.8	216.4	20.0	12.0	7.3

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Here h is the thickness of a TDF – PZT composite sample; $n = 1$, 2, and 3 is the number of nodes along a length of a TDF – PZT composite sample for the 1st, the 2nd, and the 3rd harmonic oscillations; L is the length of a PZT plate; $< \rho > = \rho_m V_m + \rho_p V_p$ is the average density of a TDF – PZT composite sample; ρ_m and ρ_p are the densities of TDF and PZT, respectively; V_m and V_p are respectively volume contents of TDF and PZT in a composite.

In Eq. (2) and Eq. (3) $<s_{11}>$ is the effective elastic compliance of a TDF – PZT composite sample calculated as

$$< s_{11} > = \frac{s_{11}^m \cdot s_{11}^p}{V_m \cdot s_{11}^p + V_p \cdot s_{11}^m}, \tag{4}$$

where s_{11}^m and s_{11}^p are the elastic compliances of TDF and PZT.

Substituting into (2) and (3) parameters of components constituting the composite 0.6 TDF – 0.3 PZT (Table 1), as well as a length of the piezoelectric plate of $8 \cdot 10^{-3}$ m and the thickness of a composite sample of $0.9 \cdot 10^{-3}$ m, we have calculated resonant frequencies f_{pn}^B and f_{pn}^L. The 1st, the 2nd, and the 3rd peaks in Fig. 2 are found to fall on resonant frequencies of the 1st harmonic of bending oscillations of a composite sample, the 2nd harmonic of bending oscillations of a composite sample, and the longitudinal oscillations of a composite sample, respectively. The same considerations are right for composites 1.2 TDF – 0.3 PZT and 0.9 TDF – 0.3 PZT.

Fig. 3 shows the dependence $\alpha_{31}(H_=)$ at the measuring magnetic field strength of 5 Oe and a resonant frequency of 15.5 kHz for the composite 0.6 TDF – 0.3 PZT. Curves, similar to a curve in Fig. 3, have also observed for bending and longitudinal oscillation modes of a 0.6 TDF – 0.3 PZT composite sample with resonant frequencies of 51.1 kHz and 139.7 kHz, respectively, and for 1.2 TDF – 0.3 PZT and 0.9 TDF – 0.3 PZT composite samples at f_{r1}, f_{r2}, and f_{r3}. Following features of the ME effect in composites have been revealed: the dependence $\alpha_{31}(H_=)$ has a maximum at ~720 Oe at any frequency f_{pn}, which is explained by the behavior of the magnetostriction coefficient depending on $H_=$ for TDF [10]; at cycling variation of $H_=$ in the dependence $\alpha_{31}(H_=)$ a hysteresis loop is observed probably related to the presence of defects in a TDF layer; a maximum value of α_{31} at ~720 Oe in $\alpha_{31}(H_=)$ is decreased as a resonant frequency varies from f_{r1} to f_{r3}; the ME hysteresis loop width is reduced with a resonant frequency varying from f_{r1} to f_{r3} (for example, see an inset in Fig. 3 for the composite 0.6 TDF – 0.3 PZT) and, apparently, is due to the fact that with increasing a resonant frequency a number of domains involved in the switching

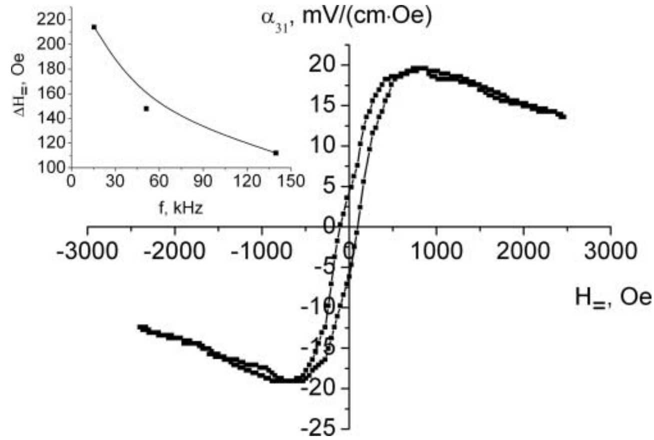

Figure 3. Field dependence of α_{31} for the composite 0.6 TDF – 0.3 PZT at room temperature. Inset: frequency dependence of the ME hysteresis loop width ΔH for the composite 0.6 TDF – 0.3 PZT.

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of TDF becomes smaller, losses on switching of magnetic domains is decreased and, as a consequence, the magnetic hysteresis loop width (and, hence, the ME hysteresis loop width) characterizing the loss is decreased; the ME hysteresis loop width is increased with a thickness of the ferromagnetic layer in the composite TDF – PZT at a certain frequency f_{pn} that is presumably attributed to the fact that in a thick ferromagnetic layer a total surface area of TDF particles is more than that in a thin layer, therefore, more defects are situated on a particle surface which fix magnetic domain boundaries leading to the increase of magnetic hysteresis losses.

Since the transverse ME effect in the composite TDF – PZT is due to a chain of links magnetostriction - elastic strain - piezoelectric effect and a magnetostriction value of λ_{11} for TDF [11] and the piezoelectric effect in PZT [12] depend on temperature, values of α_{31} are supposed to depend on temperature as well.

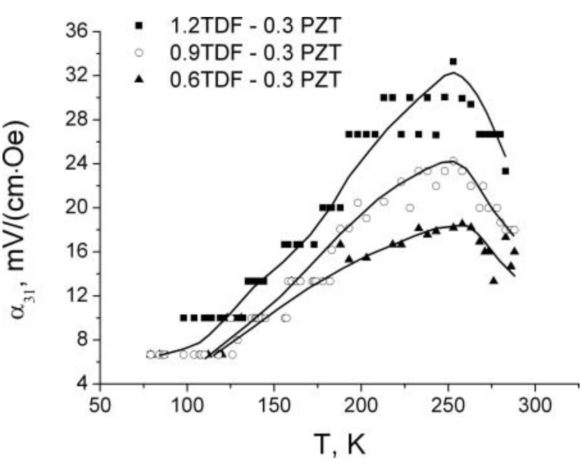

Figure 4. Temperature dependences of α_{31} at f_{r1} for composites 0.6 TDF – 0.3 PZT, 0.9 TDF – 0.3 PZT, and 1.2 TDF – 0.3 PZT.

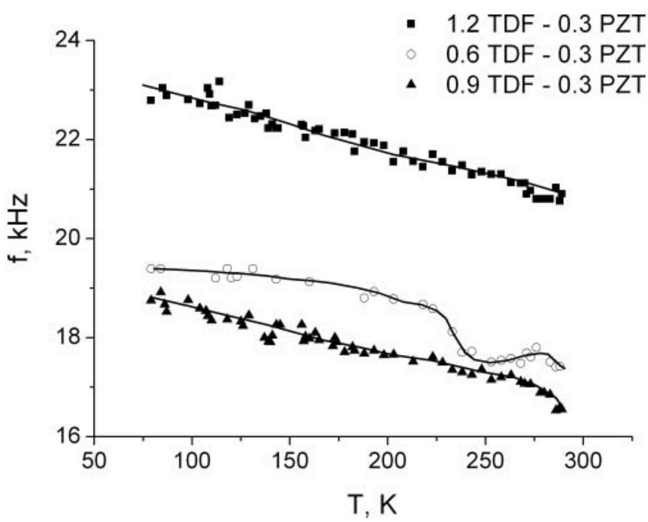

Figure 5. Temperature dependences of the resonant frequency f_{r1} for composites 0.6 TDF – 0.3 PZT, 0.9 TDF – 0.3 PZT, and 1.2 TDF – 0.3 PZT.

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Experimental results demonstrated in Fig. 4 confirm the above assumption. As can see in the figure, for composites 0.6 TDF – 0.3 PZT, 0.9 TDF – 0.3 PZT, and 1.2 TDF – 0.3 PZT the behavior of $\alpha_{31}(T)$ qualitatively coincides with that of $\lambda_{11}(T)$ for TDF [11] that enables us to relate a nature of peaks in $\alpha_{31}(T)$ at 253 K to features of the magnetostriction material TDF in composites.

A possible contribution of the condensed moisture on composite sample surfaces, which is formed during low-temperature measurements, can be excluded as peaks in the dependence $\alpha_{31}(T)$ remain unchanged at the cyclic heating-cooling runs of composite samples over a temperature range from 79 K to 289 K.

Note that for composites the resonant frequency f_{r1} corresponding to a peak in the dependence $\alpha_{31}(f_1)$ shifts to low temperatures (Fig. 5). Such a shift can be explained using Eq. (2). The fact is that with temperature a composite sample is slightly lengthened ($\Delta L/L \sim 10^{-3}$ at 300 K), therefore its density is slightly decreased as well, and the elastic compliance is significantly increased leading to a growth of a denominator in the equation used, and, hence, to the decrease in f_{r1}.

Conclusions

We have manufactured two-layered ME composites 0.6 TDF – 0.3 PZT, 0.9 TDF – 0.3 PZT, and 1.2 TDF – 0.3 PZT using the ceramic technology. The transverse ME effect in composites over a range of the measuring magnetic field frequencies of 9.9–253 kHz and temperatures from 79 K to 289 K has been studied. It is revealed that for composites resonant frequencies f_{r1} over a range from 15.5 kHz to 19.8 kHz correspond to the first mode of bending oscillations, resonant frequencies f_{r2} within limits of 50.1 – 87.8 kHz correspond to the second mode of bending oscillations, and resonant frequencies f_{r3} taking values from 125.6 kHz to 216.4 kHz correspond to the first mode of longitudinal oscillations. For the composite 0.6 TDF – 0.3 PZT the transverse ME voltage coefficient α_{31} at frequencies f_{r1}, f_{r2}, and f_{r3} has values 18 mV/(cm·Oe), 14 mV/(cm·Oe), and 8.6 mV/(cm·Oe), respectively; for the composite 0.9 TDF – 0.3 PZT the coefficient α_{31} at f_{r1}, f_{r2}, and f_{r3} is

23.4 mV/(cm·Oe), 14 mV/(cm·Oe), and 6.7 mV/(cm·Oe), respectively; and for the composite 1.2 TDF – 0.3 PZT the coefficient α_{31} at f_{r1}, f_{r2}, and f_{r3} takes values of 20 mV/(cm·Oe), 12 mV/(cm·Oe), and 7.3 mV/(cm·Oe), respectively. In the bias magnetic field of 720 Oe and the measuring magnetic field of 5 Oe at 19.8 kHz and 253 K the composite 1.2 TDF – 0.3 PZT shows the best ME interaction efficiency with a value of α_{31} equal to 32.2 mV/(cm·Oe).

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References

1. K.-H. Cho and Sh Priya, Direct and converse effect in magnetoelectric laminate composites. *Appl. Phys. Lett.* **98**, 232904 (3pp) (2011).
2. C.-W. Nan, M. I. Bichurin, S. Dong, D. Viehland, and G. Srinivasan, Multiferroic manetoelectric composites: Historical perspective, status, and future directions. *J. Appl. Phys.* **103**, 031101 (35pp) (2008).
3. M. Fiebig, Revival of the magnetoelectric effect. *J Phys D: Appl Phys*. **38**, R123–R152 (2005).
4. J. Ma, J. Hu, Zh Li, and C.-W. Nan, Recent progress in multiferroic magnetoelectric composites: from bulk to thin films. *Adv. Mater.* **23**, 1062–1087 (2011).
5. J. Ryu, A. V. Carazo, K. Uchino, and H.-E. Kim, Magnetoelectric properties in piezoelectric and magnetostrictive laminate composites. *Jpn. J. Appl. Phys.* **40**, 4948–4951 (2001).
6. S. N. Babu, T. Bhimasankaram, and S. V. Suryanarayana, Magnetoelectric effect in metal – PZT laminates. *Bull. Mater. Sci.* **28**, 419–422 (2005).
7. X. W. Dong, Y. J. Wu, J. G. Wan, T. Wei, Z. H. Zhang, S. Chen, H. Yu, and J.-M. Liu, Phase shift of electric-field-induced magnetization in magnetoelectric laminate composite. *J. Phys. D: Appl. Phys.* **41**, 035003 (4pp) (2008).
8. S. A. Gridnev, A. V. Kalgin, A. A. Amirov, and I. K. Kamilov, Magnetic and magnetoelectric properties of particulate (x)$PbZr_{0.53}Ti_{0.47}O_3$ – (1-x)$Mn_{0.4}Zn_{0.6}Fe_2O_4$ composites. *Ferroelectrics*. **397**, 142–150 (2010).
9. J. G. Wan, Z. Y. Li, Y. Wang, M. Zeng, G. H. Wang, and J.-M. Liu, Strong flexural magnetoelectric effect in Terfenol-D/epoxy – $Pb(Zr,Ti)O_3$ bilayer. *Appl. Phys. Lett.* **86**, 202504 (3pp) (2005).
10. D. Wu, W. Gong, H. Deng, and M. Li, Magnetoelectric composite ceramics of nickel ferrite and lead zirconate titanate via in situ processing. *J. Phys. D: Appl. Phys.* **40**, 5002–5005 (2007).
11. G. A. Politova, I. S. Tereshina, S. A. Nikitin, T. G. Sochenkova, V. N. Verbetsky, A. A. Salamova, and M. V. Makarova, Effect of hydrogenation on the magnetic and magnetoelastic properties of the $Tb_{0.27}Dy_{0.73}Fe_2$ and $Tb_{0.27}Dy_{0.73}Co_2$ compounds with compensated magnetic anisotropy. *Physics of the solid state*. **47**, 1909–1913 (2005).
12. E. G. Smazhevskaya and N. B. Feldman, *Piezoelectric ceramics*. (Soviet radio, Moscow 1971).

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