INFLUENCE OF TEMPERATURE ON MAGNETOELECTRIC EFFECT IN A STRUCTURE CONTAINING LANGATATE

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Abstract. The effect of temperature on the linear magnetoelectric response in a three-layered composite structure comprising a single crystal of langatate with thin films of an amorphous ferromagnetic alloy deposited on either side was investigated. Measurements were conducted in the temperature range of 220–340 K. As a result, a linear reduction in the magnetoelectric coupling coefficient and sensitivity to a magnetic field with an increase in temperature was observed.

Keywords: *magnetoelectric effect, temperature dependence, langatate, amorphous magnetic alloy, magnetron sputtering*

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INTRODUCTION

Among multiferroic materials possessing magnetoelectric (ME) effects, the most interesting are layered composite structures consisting of interconnected piezoelectric (PE) and ferromagnetic (FM) layers. The ME effect is a change in the electric polarization of the sample when it is placed in a magnetic field and arises in such composites as a result of a combination of the magnetostriction of the FM layer and the piezoelectric effect in the PE layer [1 -4]. The active study of ME effects has led to a transition from samples with an adhesive layer to thin-film structures fabricated by a variety of sputtering techniques.

This transition is caused by the necessity to increase the manufacturability of devices with functional elements based on ME layered composites, first of all, sensors of magnetic fields. Transition to thin-film coatings leads to improvement of reproducibility of properties of ME composites, increase of acoustic goodness and, as a consequence, sensitivity of sensors operating in the resonant mode, as well as reduction of device sizes. Critical for mass production of devices based on ME effects is their temperature stability and compatibility of used materials with existing technologies [5].

To date, most of the studies have been carried out at room temperature, and a number of papers have been published to investigate the effect of temperature on the ME characteristics of composite structures. In [6-8], theoretical and experimental studies of the ME effects in PZT-5A/TbDyFe, PMN-PT/nickel, and langatate/nickel composite structures at temperatures from 150 to 400 K were carried out. It was shown in [8,9] that in the structure with a nickel layer electrolytically deposited on the langatate surface, the generated ME voltage depends on the combined contribution of the variation of the sample goodness of fit and the piezomagnetic modulus of the nickel layer. At the same time, the adhesive layer for structures in which PE and FM layers are bonded using adhesive degrades strongly with temperature change, which leads to a significant (or unpredictable) decrease in the ME coefficients at temperatures other than room temperature [8-10].

The use of piezoelectric ceramics is impractical for application in precision sensors of magnetic fields, due to the strong dependence of their electrical and mechanical properties on the ambient temperature and low acoustic goodness of fit [11]. Langatate monocrystals show high thermostability of electrical and mechanical properties [12] and are compatible with MEMS technologies, making them candidates for the role of piezoelectric component of ME composites for magnetic field sensors. In this work, the ME effect was investigated in structures based on piezoelectric langatate single crystal, which has thermostable properties [10-12]. The influence of temperature on the ME effect in such composite structures has not been studied before.

SAMPLE DESCRIPTION

In this work, the ME effect in a three-layer composite structure based on a single crystal of langatate ($La_3Ga_{5.5}Ta_{(0.5)\ O14}$) X - slice manufactured by JSC "Fomos-Materials", Moscow [13] has been investigated. On both sides of the single crystal were deposited layers of thin amorphous ferromagnetic $Fe_{77}Co_4Si_{(8)\ B11}$with thickness~ 1.5 μm. The dimensions of the structure were 21.3×4.7×0.4 mm. FM layers of magnetostrictive material were deposited in NITU MISIS (Department of Materials Science of Semiconductors and Dielectrics) by high-frequency magnetron sputtering of the target. The target had the composition: $Fe_{(7)\ (0)}Co_8Si_{(12)\ B(1)\ (0)}$. Deposition was carried out in a SUNPLA-40TM vacuum process chamber without additional heating of the substrate holder. Synthesis of films was carried out in argon atmosphere with an operating pressure of 0.5 Pa at a magnetron power of 200 W. The detailed method of fabrication of thin amorphous ferromagnetic $Fe_{77}Co_4Si_{(8)}B_{11}$is described in [14,15].

MEASUREMENT TECHNIQUE

The ME effect was investigated by the dynamic method on the setup developed in the Research Center "Magnetoelectric materials and devices" of RTU MIREA [11] in the temperature range from 220 to 340 K. The composite structure was placed in a fluoroplastic cell between Helmholtz rings, which created a homogeneous constant magnetic field of up to 350 E. The composite structure was placed in a fluoroplastic cell between Helmholtz rings, which created a homogeneous constant magnetic field with an intensity up to 350 Å. An alternating magnetic field with amplitude *h* up to 0.22 Å was created using modulating coils powered by an Agilent 33210A generator (Agilent Technologies, Inc., Santa Clara, CA, USA). The voltage *u*(*t*) generated by the DOE structure was fed to an SR-560 preamplifier with an input impedance of 100 megohms and gain k = 1, which acted as an upper-pass filter with a cutoff frequency $f_{(c)\,(-)\,(o)}$ = 100 Hz. The voltage from the preamplifier output was measured with a voltmeter AKIP-2401 (AO "PriST", Moscow) with an input impedance of 10 megohms.

The temperature in the working chamber was set by changing the flow of vaporized liquid nitrogen heated to a certain temperature and maintained at the same level by proportional-integral-differential (PID) control. The temperature was measured using a Pt1000 type thermistor placed near the sample. Temperature measurements were performed according to the following protocol. The structure was cooled to an initial temperature of 220 K, and the voltage amplitude-frequency response as well as the voltage dependencies of the DC and AC magnetic fields at the resonance frequency were measured. The temperature was changed in steps of 10 K with the external magnetic field switched off, and at each temperature value the measurements were repeated. On the basis of the obtained amplitude and frequency characteristics (AFC), the dependences of the resonance frequency and goodness of fit on temperature were plotted.

MEASUREMENT RESULTS

The results of the study of the linear ME effect at room temperature are presented in Figure 1. Fig. *1a* shows the dependence of *the* ME voltage u generated by the structure on the frequency of the alternating magnetic field f with amplitude h = 0.06 Å. The measurements were carried out at a constant field H_m= 3 Å. The peak at frequency $f \approx$ 115.43 kHz corresponds to the first mode of longitudinal acoustic oscillations and has an amplitude $u \approx$ 420 mV. The goodness of fit of the resonance is $Q = \Delta f/f \approx 4800$, where Δf is the peak width at 0.7 of the resonance height.

Fig. *1b* shows the dependence of the ME voltage generated by the structure on the constant magnetic field measured at the resonance frequency at the amplitude of the alternating magnetic field h = 0.06 E. It can be seen that the voltage grows up to the maximum value ($u \approx$ 420 mV)achieved in the optimal magnetic field H_m= 3 E. With further increase of the constant magnetic field, the voltage monotonically decreases. The shape of the obtained curve is determined by the shape of the field dependence of the piezomagnetic modulus ($q = \partial\ \lambda/\partial H$, where λ is the dependence of the magnetostriction coefficient on the magnetic field) of the FM layer [10].

The value of the ME coefficient is determined by the formula $\alpha = (u/b)/h$, where *u is the* amplitude of the ME voltage, *h is* the amplitude of the alternating magnetic field, and *b* is the thickness of the PE layer of the structure. The value of the ME coefficient obtained in optimum field was $\alpha = 175$ V·E^{-1} ·cm^{-1}. This is greater than the value of 64 V·E^{-1} ·cm$^{(-1)}$obtained on the Metglas-LGT-Metglas structure fabricated by bonding [14] due to better strain transfer between layers, but ~2.5 times less than the 450 V·E^{-1} ·cm^{-1}coefficient of the LGT-Metglas structure with a 25 µm thick magnetic layer [11], which is primarily due to the small thickness of the FM layer compared to the langatat thickness in our case [17].

Fig. *1c* shows the dependence of the ME voltage generated by the structure on the AC magnetic field amplitude *h* measured in the field H_m. The obtained dependence in the region of small AC magnetic field amplitudes *h* has a form close to linear. The deviation from the linear dependence toward smaller voltage values with *h* growth is due to the occurrence of nonlinear ME effects. The sensitivity of the sample, defined as the tangent of the slope of the linear part of the dependence of *u* on *h* in the linear section up to 0.02 Å, is equal to $s \approx$ 7.5 V·E^{-1}, which is greater than the sensitivity of the sample made by bonding ($s \approx$ 6 V·E^{-1}) [16]. [16].

At the second stage, the temperature dependences of the linear magnetoelectric effect were investigated. The temperature dependences of the ME coefficient, acoustic goodness of resonance, and resonance frequency were obtained on the basis of the measured frequency dependences of the ME voltage and are presented in Fig. 2.

Fig. *2a* shows the dependence of the value of the ME coefficient obtained in the optimal magnetic field H_m, the value of which was selected for each point, on the temperature *T*. The value of the ME coefficient decreases linearly from the value $\alpha \approx$ 250 V·E^{-1} ·cm^{-1}at 220 K to the value $\alpha \approx$ 150 V·E^{-1} ·cm^{-1}at 340 K. All values of the ME coefficient are obtained in the optimal magnetic field $H_m$at the amplitude of the alternating magnetic field h = 0.22 E. The total change of the ME coefficient $\Delta\alpha/\alpha$ is 40%. This behavior of the ME effect is due to the decrease in the acoustic goodness of the resonance from a value of ~8000 to ~5100 with increasing temperature, which can be seen in Fig. 2b. The overall change in the goodness of fit was also about 40%, which can be attributed to the increase in the internal mechanical losses of the resonator from temperature. In addition, Fig. 2a shows the dependence of the sensitivity of the structure to magnetic field on temperature. It can be seen that the sensitivity drops linearly with increasing temperature ~ 40% from $s \approx 9.8$ V/E (at $T =$ 220 K) to $s \approx 6$ V/E (at $T =$ 340 K).

Finally, Fig. *2c* shows the dependence of the resonant frequency f on temperature, where a slight increase (0.1%) of the resonant frequency with increasing temperature is observed, which is due to the temperature stability of Young's modulus ($f \propto \sqrt{E_{22}}$) of the langatate single crystal [18].

CONCLUSION

Thus, we have investigated the influence of temperature on the characteristics of the magnetoelectric effect in a structure fabricated on the basis of a plate of langatate single crystal with layers of thin amorphous ferromagnetic alloy sputtered on both its sides. At room temperature, the magnetoelectric coefficient was $\alpha \approx 175$ V· E^{-1} ·cm^{-1}. It is shown that the magnetoelectric coefficient decreases linearly from a value of 350 V· $E^{(-1)}$ ·$cm^{-1}$by 40 % in the temperature range from 220 to 340 K. The resonant frequency of the structure is almost independent of temperature. The results obtained can be promising for the creation of highly sensitive sensors of magnetic fields based on single crystals operating in a wide temperature range.

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FIGURE CAPTIONS

Fig. 1. Characteristics of the linear magnetoelectric effect: amplitude-frequency response of the structure (a) measured at a constant magnetic field H_m= 3 E, at the amplitude of the alternating magnetic field h = 0.06 Å; dependence of the ME voltage generated by the structure on the constant magnetic field (b), measured at the resonance frequency at the amplitude of the alternating magnetic field h = 0.06 Å; dependence of the ME voltage on the amplitude of the alternating magnetic field (c), measured at the resonance frequency at the constant magnetic field H_m= 3 Å.

Fig. 2. Temperature dependences of the linear ME effect: dependence of the ME coefficient and sensitivity on temperature (a), dependence of the acoustic goodness of resonance on temperature (b), dependence of the resonant frequency of the structure on temperature (c).

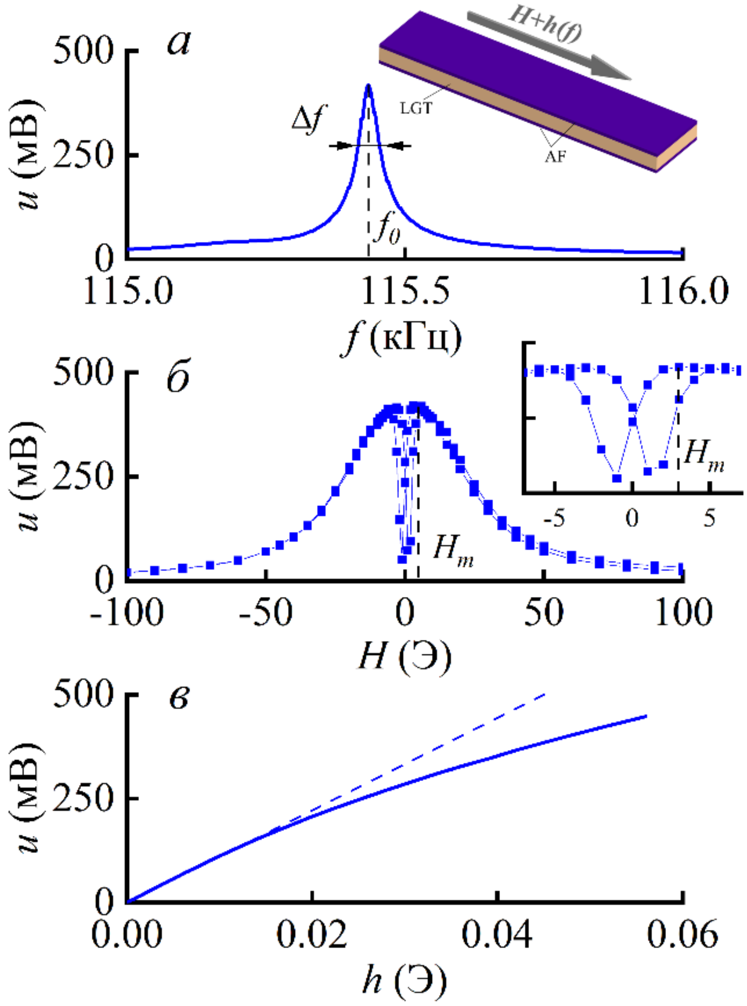

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Fig. 1.

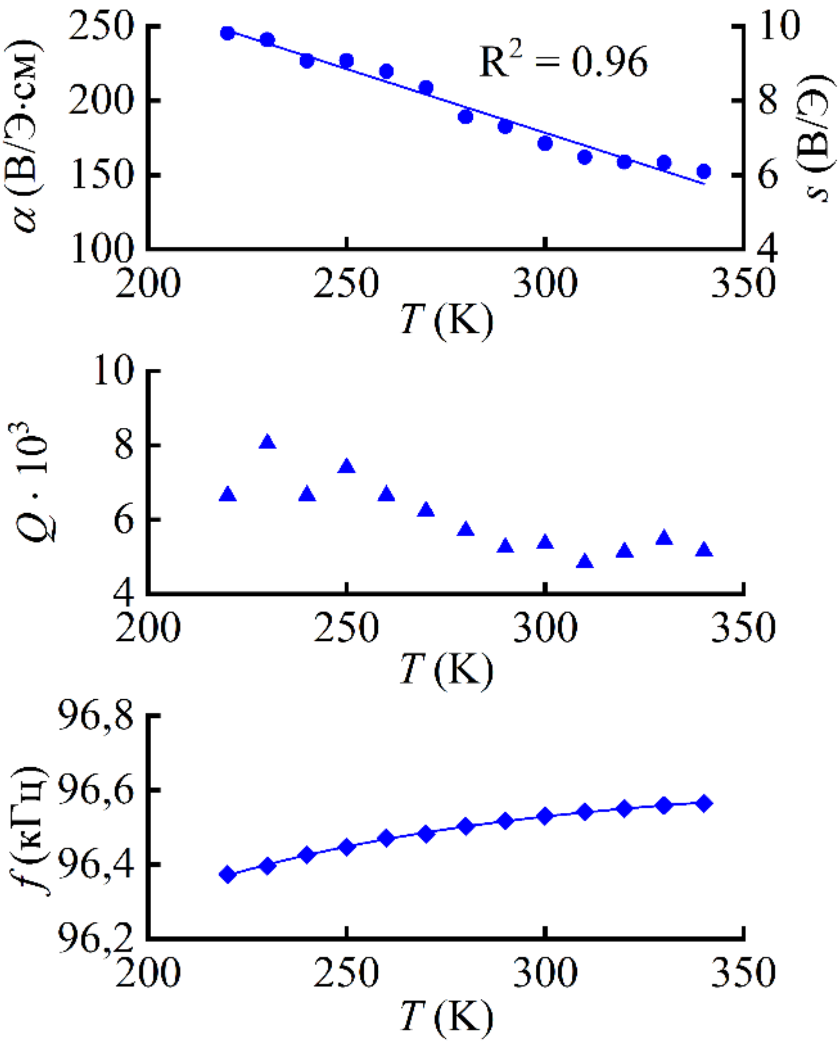
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Fig. 2.