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Magnetoelectric characteristics of the structure amorphous alloy AMAG/lead zirconate titanate/amorphous alloy AMAG

© D.A. Filippov,[1] S.O. Sapelnikov,[1] Jitao Zhang[2]

[1] Yaroslav-the-Wise Novgorod State University,
Veliky Novgorod, Russia
[2] College of Electrical and Information Engineering, Zhengzhou University of Light Industry,
450002 Zhengzhou, China
e-mail: dmitry.filippov@novsu.ru, dmitrij_filippov_57@mail.ru

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The field, frequency and load characteristics of the linear magnetoelectric effect of three-layer structures based on the amorphous AMAG alloy and lead zirconate titanate were studied in the low-frequency region and in the region of electromechanical resonance. The influence of the magnetostrictive layer thickness on the magnetoelectric characteristics of these structures was considered. It was shown that the value of the magnetoelectric voltage coefficient and the resonance frequency increase with increasing thickness of the magnetostrictive layer. The magnitude of the bias magnetic field corresponding to the maximum of the magnetoelectric response also increases with increasing magnetostrictive layer thickness. The experimental results are in good agreement with theoretical predictions.

Keywords: composite structure, magnetostriction, piezoelectricity, magnetoelectric effect, magnetoelectric voltage coefficient.

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Introduction

Composite structures combining magnetostrictive and piezoelectric components offer ample opportunities for device engineering in the field of straintronics, which is a rapidly developing branch of modern electronics [1]. The key feature of such heterogeneous systems is the emergence of a magnetoelectric (ME) effect, which is lacking in each of the separate phases. This effect is driven by mechanical interaction at the interface between magnetostrictive and piezoelectric materials [2]. When exposed to an alternating magnetic field, the magnetostrictive phase generates deformations that are transmitted mechanically to the piezoelectric phase. This induces electrical polarization in the latter due to the direct piezoelectric effect. The unique characteristics of ME structures turn them into promising structural components for a new generation of devices: electrically and magnetically controlled inductive elements [3,4]; energy harvesters [5]; current/voltage converters (magnetoelectric gyrators) [6,7]; high-sensitivity magnetometric sensors [8–14]; and compact ME antennas [15,16].

The ME response magnitude is proportional to the product of piezoelectric coefficient d and the piezomagnetic coefficient:

$$q = \frac{\partial\lambda(H)}{\partial H}\Big|_{H=H_{bias}},$$

where λ is magnetostriction and H_{bias} is the bias field. In measurements, the bias field is chosen in such a way as to correspond to the maximum of the piezomagnetic coefficient; i.e., $H_{bias} = H_{opt}$. Amorphous alloys based on FeBSiC (Metglas — United States, Germany; AMAG — Russia) have an advantage over other magnetics in that the maximum piezomagnetic coefficient is observed in bias fields H_{opt} on the order of several kA/m [17,18], while this bias field for permendur is several dozen kA/m [19]. The problem with using an amorphous alloy to fabricate ME structures is that it is produced in the form of ribbons $20-25\,\mu$m in thickness, and the maximum ME response is observed when the thickness of a magnetic material is approximately equal to the thickness of a piezoelectric, which is normally on the order of several hundred micrometers. For this ratio to be fulfilled, one needs to use a magnetostrictive layer consisting of several layers of amorphous alloy foil, which makes the process of structure fabrication significantly more complex. The ME effect in Metglas-based structures has been examined earlier in [20–23]. The authors of [20] have studied the field and frequency characteristics of the ME response of a structure with a piezoelectric ceramic based on lead zirconate titanate (PZT). The ME properties of the symmetric Metglas/PZT/Metglas structure were investigated as a function of thickness ratio of the magnetostrictive layer and the sample at a fixed piezoelectric material thickness of 0.5 mm. It was determined experimentally that the field dependence of the ME voltage coefficient (MEVC) has a maximum in a bias field varying from 2.8 to 13.5 kA/m with a change in thickness ratio from 0.107 to 0.590 (one and 12 Metglas layers with a thickness of $30\,\mu$m on either side of the piezoelectric). The MEVC value in an optimal bias field initially increases from 275 mV/A at a thickness ratio of 0.107, reaches a maximum of 680 mV/A at a thickness ratio of 0.519, and then decreases to 440 mV/A at a thickness ratio of 0.590. The frequency dependence of the MEVC has a maximum at the frequency of electromechanical resonance, which increases nonlinearly with increasing thickness ratio. The ME response in a three-layer Metglas/PZT/Metglas structure has also been investigated in [21]. It was demonstrated that the MEVC value increases by a factor of approximately 1.5–1.75 as the magnetostrictive layer thickness increases from 50 to 150 μm. An ME magnetic field sensor based on a Metglas/quartz/Metglas structure has been examined in [22]. The sensor was demonstrated to be sensitive to fields on the order of several pT at a frequency of 1 Hz. The resonance frequency of such a structure decreases with an increase in the number of Metglas layers, while the resonant MEVC value initially increases with an increase in layer number, reaches a maximum at 14 layers, and then decreases. The characteristics of an ME transformer based on a Metglas/PZT/Metglas structure with the number of Metglas layers on either side of the piezoelectric material varying from 1 to 3 have been studied in [23]. While the directions of magnetization and polarization were perpendicular to each other in [20,21], these directions in [23] were collinear in the plane of the sample. It was demonstrated that the value of H_{opt} increases from 1.2 kA/m to 2.4 kA/m as the layer number increases from 1 to 3, while the resonant frequency varies from 50.0 to 50.9 kHz.

Although regarded as an alternative to the Metglas alloy, the amorphous soft magnetic alloy (AMAG) produced in Russia (Mstator, Borovichi) differs slightly [24] in its parameters [25]. In the present study, we examine the field, frequency, and load characteristics of the ME response of three-layer AMAG/PZT/AMAG structures with different numbers of amorphous alloy layers. It is demonstrated that ME structures based on the AMAG alloy are not inferior in their characteristics to ME structures based on foreign counterparts and are well suited for devices utilizing the ME effect, such as magnetic field sensors, gyrators, power dividers, magnetoelectric antennas, etc.

1. Materials and methods

The following symmetric three-layer structures were fabricated for studies: magnetostrictive amorphous alloy/piezoelectric/magnetostrictive amorphous alloy. This symmetric three-layer configuration was preferred over a two-layer design, since it lacks competing vibration modes. In an asymmetric two-layer structure, planar vibrations are superimposed on bending vibrations, weakening the ME conversion [26]. In contrast, the symmetric three-layer structure excited by a magnetic field in the sample plane induces exclusively planar vibrations, providing a higher ME conversion coefficient.

The magnetostrictive piezoelectric structure consisted of a PZT-19 piezoceramic wafer ($20 \times 5 \times 0.3$ mm in size) with silver electrodes applied by high-temperature annealing (Piezopribor, Russia) and magnetostrictive elements made of AMAG 212N foil (PAO Mstator, Russia; thickness, 20 μm; size, 18×5 mm), which were secure to each side of it with Loctite 435 glue. To enhance the ME response, which is achieved when the thicknesses of the piezoelectric layer (t_p) and the magnetostrictive layer (t_m) are comparable, the latter was formed from foil packs. (Here ans elsewhere, subscripts p and m denote the material type: p — piezoelectric, m — magnetic.) Four versions of structures with magnetostrictive layer thicknesses of t_m = 40, 80, 120, and 160 μm (*1–4* foil layers on each side, respectively) were fabricated. The schematic diagram of these heterostructures is shown in Fig. 1.

MEVC α_E was used as the main parameter characterizing the ME response. It was determined using the following relation:

$$\alpha_E = \left(\frac{V}{t_p h}\right), \tag{1}$$

where h is the alternating magnetic field and V is the output voltage.

The MEVC was determined by recording the sample voltage in a combined magnetic field: alternating field h and bias field H_{bias}, which was set equal to H_{opt} and varied with sample thickness. These fields were co-directional, lay in the plane of the sample, and were directed along its long axis and perpendicular to the polarization vector of the piezoelectric material. ME voltage measurements were carried out using a GDM 79061 voltmeter. Since the input resistance of the voltmeter (100 MΩ) was much greater than the resistance of the magnetostrictive piezoelectric structure, which was 3 kΩ at frequency f = 10 kHz, the open circuit condition was expressly satisfied.

The field dependence of MEVC at a fixed frequency f = 10 kHz of the alternating magnetic field and its amplitude h = 64 A/m was examined first. After that, the H_{bias} value corresponding to the maximum of ME response (i.e., $H_{bias} = H_{opt}$) was set, and the frequency and load characteristics were measured for each sample.

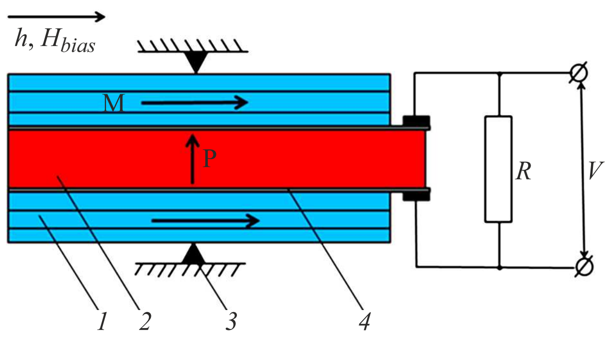

Figure 1. Schematic diagram of the ME heterostructure: *1* — magnetostrictive layer (AMAG 212 N), *2* — piezoelectric material (PZT–19), *3* — supports, and *4* — thin Ag layer.

The frequency characteristic was measured under open circuit conditions, and load characteristics were recorded both at resonance frequencies and at frequencies far away from the resonance at 10 kHz.

2. Results and discussion

Figure 2 presents the results of measurement of field dependences of the averaged (with increasing and decreasing bias field) MEVC value in the low-frequency region at different thicknesses of the magnetostrictive layer.

It follows from Fig. 2 that the field dependence of MEVC has a typical form representative of the dependence of magnetostriction of the amorphous alloy on the bias field [18]. The MEVC maximum is observed in bias field H_{opt}, and the magnitude of this field increases with increasing thickness of the magnetostrictive layer. Figure 3 shows the dependence of optimal bias field on the magnetostrictive layer thickness.

It reveals a correlation between the magnetostrictive layer thickness and the optimal bias field: as the thickness increases from 40 to 160 μm, the optimal bias field magnitude increases almost linearly from 1.28 to 3.42 kA/m. This trend indicates that the MEVC maximum shifts toward stronger fields as the magnetic material grows thicker.

This may be attributed to the fact that, instead of being a function of magnetic field strength, magnetostriction is actually a function of magnetization, which is related to the sample shape by the demagnetization coefficients. These coefficients change with an increase in thickness of the magnetostrictive layer, altering the dependence of magnetization on the magnetic field strength. As was demonstrated in [27], demagnetization factor N of a rectangular wafer is determined by its relative length $l_{rel} = L/t$ (L and t are the wafer length and thickness, respectively): it decreases with increasing l_{rel}. As the magnetostrictive layer grows thicker, l_{rel} decreases, which entails an increase in N. This, in turn, explains the observed shift of optimal field H_{opt} (corresponding to the ME response maximum) toward larger values.

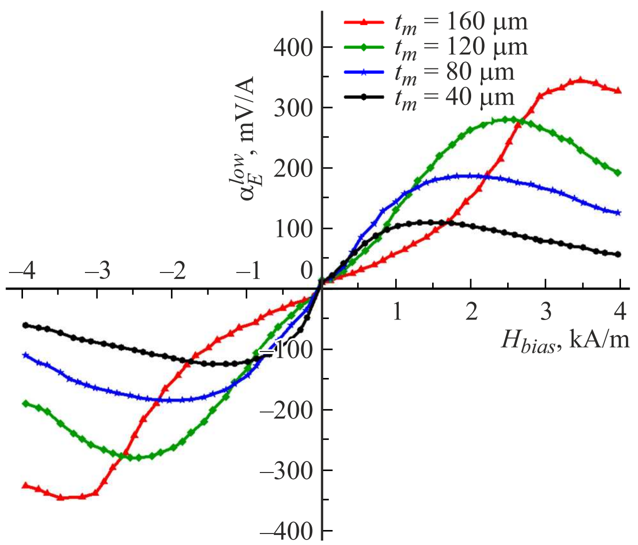

Figure 2. Dependences of the MEVC value on the bias field for four samples with different thicknesses of the magnetostrictive layer at frequency $f = 10$ kHz. Piezoelectric layer thickness $t_p = 300\,\mu$m.

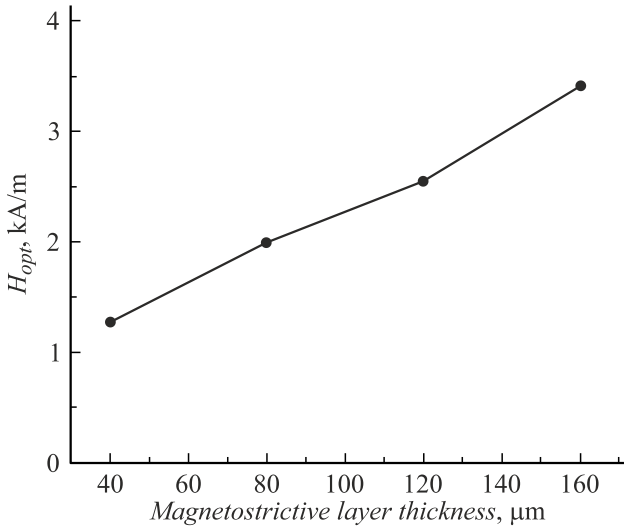

Figure 3. Dependence of optimal bias field H_{opt} on the magnetostrictive layer thickness.

It should be noted that an increase in H_{opt} with an increase in thickness of the magnetostrictive layer of similar structures was noted in [23], where a voltage transformer based on the ME effect was studied. The difference between the present study and [23] is that the ME response is investigated here with a transverse orientation of the electric and magnetic fields, while the ME response in [23] was examined with a parallel orientation of the fields (longitudinal ME effect).

The MEVC maximum in bias field H_{opt} also increases with increasing thickness of the magnetostrictive layer. This result is consistent with theoretical predictions. According to [26], the MEVC value in the low-frequency region is determined by the following expression:

$$\alpha_E^{low} = \frac{Y_p d_{31} q_{11}}{\varepsilon_{33}\varepsilon_0} \frac{Y_m t_m}{(Y_m t_m + Y_p t_p)}. \qquad (2)$$

Here, Y_m, Y_p are the Young's moduli of the magnetic and piezoelectric materials, respectively; d_{31} and q_{11} denote the piezoelectric and piezomagnetic coefficients; ε_{33} is the relative permittivity of the piezoelectric material; and ε_0 is the permittivity of vacuum ($\varepsilon_0 = 8.85 \cdot 10^{-12}$ F/m). At the resonance frequency, the MEVC is enhanced by a factor of Q (Q is the Q factor of the structure). It follows from expression (2) that the MEVC value increases proportionally to t_m at small magnetic thicknesses ($Y_m t_m \ll Y_p t_p$). With a further increase in thickness, the dependence becomes nonlinear, approaching saturation asymptotically at $Y_m t_m \gg Y_p t_p$.

Figure 4 presents the experimentally obtained dependence of MEVC on the thickness of the magnetic material at frequency $f = 10$ kHz and the dependence calculated theoretically using formula (2) with bias field $H_{bias} = H_{opt}$. The following values of the piezoelectric and magnetic parameters were used in calculations: $Y_p = 67$ GPa, $Y_m = 110$ GPa, $\varepsilon_{33} = 1750$, $d_{31} = -175 \cdot 10^{-12}$ m/V, and $q_{11} = 0.95 \cdot 10^{-9}$ m/A. In addition, it was assumed in calculations that the shape of the magnetostriction curve does not vary with magnetization of the sample (i.e., the maximum piezomagnetic coefficient was observed at one and the same magnetization values regardless of the sample thickness). Thus, it could be assumed that the piezomagnetic coefficient remains unchanged at different sample thicknesses.

It can be seen from Fig. 4 that the experimental values agree closely with the theoretical results. The MEVC value increased by a factor of 3 (from 110 to 340 mV/A) as the magnetostrictive layer thickness increased from 40 to 160 μm. As the number of magnetostrictive Metglas layers increased from 1 to 4 in [20], the MEVC value increased by a factor of 2.3; it reached its maximum at 9 layers and decreased when 12 Metglas layers were used. This reduction is apparently attributable to the fact that the quality of mechanical coupling of layers deteriorates when the number of layers becomes too large.

Figure 5 shows the frequency dependences of MEVC for four samples measured in bias fields corresponding to the maximum ME response.

It can be seen that the MEVC peak is found in the region of electromechanical resonance (EMR). A resonant increase in MEVC is observed at frequencies of 80.0, 82.2, 84.5, and 85.9 kHz with a magnetostrictive layer thickness of 40, 80, 120, and 160 μm, respectively; the corresponding resonant MEVC values are 10, 18, 23, and 26 V/A with a structure Q factor of 80, 95, 84, and 82. The Q

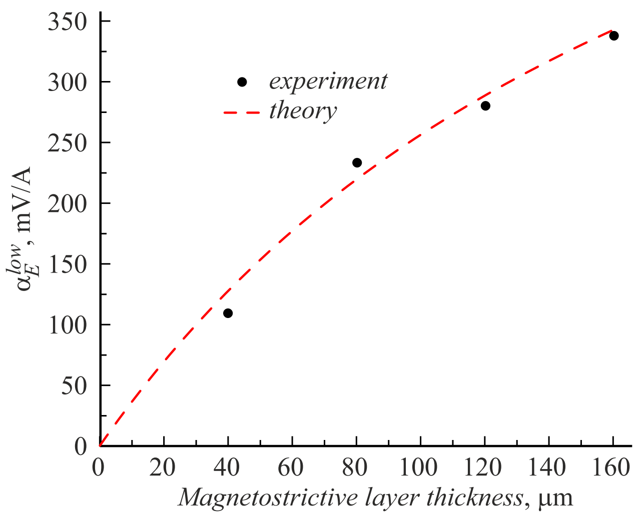

Figure 4. Dependence of the low-frequency ($f = 10$ kHz) MEVC on the magnetostrictive layer thickness at optimal bias fields. Piezoelectric layer thickness $t_p = 300\,\mu$m.

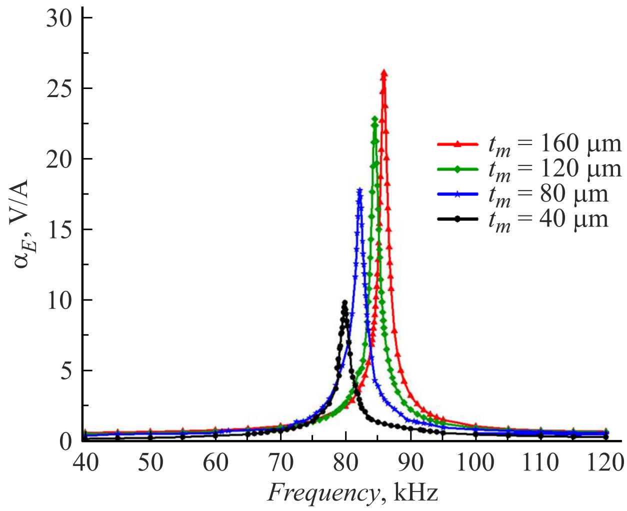

Figure 5. Frequency dependences of the MEVC value for samples with different thicknesses of the magnetostrictive layer. Piezoelectric layer thickness $t_p = 300\,\mu$m.

factor decreases with increasing number of layers of the amorphous alloy due to an increase in thickness of the adhesive assembly, which has a negative effect on the mechanical coupling of layers. According to [28], the frequency dependence of MEVC for a symmetric three-layer structure is written as

$$\alpha_E = \frac{Y_p d_{31} q_{11}}{\varepsilon_{33}\varepsilon_0} \frac{Y_m t_m}{(Y_m t_m + Y_p t_p)} \frac{1}{\Delta_a} \frac{\tan(\kappa)}{\kappa}, \quad (3)$$

where $\kappa = k L_{\text{eff}}/2$ is a dimensionless parameter; $k = \sqrt{\frac{\rho_{\text{eff}}}{Y_{\text{eff}}}}\,\omega$ is the wave number; $L_{\text{eff}} = (L_m t_m + L_p t_p)/t$, $\rho_{\text{eff}} = (\rho_m t_m + \rho_p t_p)/t$, and $Y_{\text{eff}} = (Y_m t_m + Y_p t_p)/t$ are the effective length, density, and Young's modulus of the heterostructure, respectively; L_m, L_p are the magnetostrictive and piezoelectric layer lengths;$t = t_m + t_p$ is the total thickness of the sample; and $\omega = 2\pi f$ is the angular frequency.

The introduced dimensionless parameter Δ_a characterizes the condition for achieving the peak MEVC value and is given by

$$\Delta_a = 1 - k_p^2 \left(1 - \frac{Y_p t_p}{Y_{\text{eff}}\, t} \frac{\tan(\kappa)}{\kappa}\right), \quad (4)$$

where $k_p^2 = (Y_p (d_{31})^2)/(\varepsilon_{33}\varepsilon_0)$ is the electromechanical coupling coefficient squared. It follows from Eq. (3) that the MEVC peak is observed at the so-called antiresonance frequency when dimensionless parameter $\Delta_a = 0$. As a first approximation, the resonant increase in MEVC occurs when dimensionless parameter $\kappa \approx \pi/2$. The corresponding resonance frequency value is

$$f_{res} \approx \frac{1}{2L_{\text{eff}}} \sqrt{\frac{Y_{\text{eff}}}{\rho_{\text{eff}}}}. \quad (5)$$

The observed increase in resonance frequency with increasing magnetic layer thickness (t_m) has a dual mechanism. The first mechanism is geometric: since the

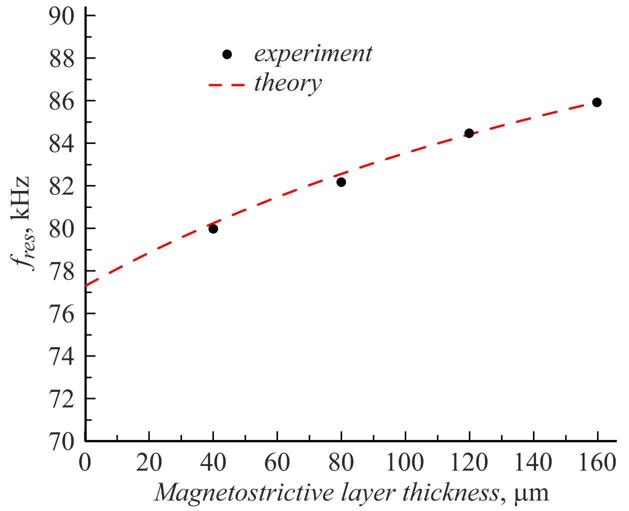

Figure 6. Dependence of the resonance frequency on the magnetostrictive layer thickness.

magnetostrictive layer is shorter than the piezoelectric one, an increase in t_m reduces effective length L_{eff} of the system, increasing frequency f_{res} directly. The second mechanism has to do with the difference in elastic properties: the Young's modulus of AMAG (~ 110 GPa) is significantly higher than that of PZT-19 (~ 67 GPa), while their densities are close. As a result, with an increase in t_m, effective modulus Y_{eff} grows faster than effective density ρ_{eff}, which, according to formula (5), should contribute additionally to an increase in frequency. The $f_{res}(t_m)$ dependence is illustrated in Fig. 6. In contrast to the present study, the authors of [22] have examined the Metglas/quartz/Metglas structure. The Young's modulus of quartz (~ 90 GPa) is comparable to that of Metglas, but its density (2200 kg/m^3) is more than three times lower than the density of Metglas (~ 7180 kg/m^3). Thus, with an increase in Metglas thickness, the numerator in the expression under the radical sign in (5) grows significantly slower than the denominator, and the resonance thickness decreases as a result.

Since the maxima of the ME effect in different samples are achieved at different optimal bias fields (H_{opt}), the question arises as to whether ΔE (the change in Young's modulus of the amorphous alloy under the influence of a magnetic field [29]) contributes to the frequency shift. However, it was established experimentally that the resonance frequency did not change as the bias field was varied from 0 to 4 kA/m. This is attributable to the fact that the magnetic fields used in experiments were not strong enough to alter significantly the elastic properties of the material. For example, a 1% relative change in resonance frequency was achieved in [29] only with a bias field of approximately 40 kA/m, which is ten times greater than the maximum field magnitude used in the present work.

The proposed electrical equivalent circuit of the ME structure features a voltage source with EMF $\varepsilon_{ME} = \alpha_E t_p h$ and two resistances connected in parallel: leakage resistance R_{leak} and capacitive resistance $X_C = 1/\omega C$, where $C = \varepsilon_{33} L_p W / t_p$ is the capacitance of the piezoelectric wafer. Since the leakage resistance is much greater than the capacitive resistance at frequencies on the order of several kHz and higher, output voltage V across load resistance R will be written as

$$V = \frac{\varepsilon_{ME}}{\sqrt{R^2 + X_C^2}} R. \tag{6}$$

Equation (6) suggests that the output voltage depends linearly on R in the region of low load resistances ($R \ll X_C$). In the region of high resistances ($R \gg X_C$), the dependence becomes nonlinear, and voltage reaches the saturation level determined by expression $V = \alpha_E t^p h$. The experimental dependences of output voltage in the resonant mode (Fig. 7) were obtained at EMR frequencies increasing successively (80.0, 82.2, 84.5, and 85.9 kHz for samples Nos. 1–4, respectively) with thickness of the magnetostrictive layer.

It can be seen from Fig. 7 that the output voltage tends to saturation at load resistances $R \geq 5\,\text{k}\Omega$. This is quite consistent with theory, since the capacitance of the piezoelectric wafer is $C = 5.2$ nF, which yields a capacitive resistance $X_C \approx 380\,\Omega$ at resonance frequencies on the order of 80 kHz. Figure 8 presents the dependence of the output voltage on the magnetostrictive layer thickness at resonance frequencies with load resistance $R = 5\,\text{k}\Omega$.

It can be seen that the output voltage first increases significantly (at magnetostrictive layer thicknesses up to 120 μm), but then the variation becomes significantly less pronounced, and the output voltage tends to saturation at thicknesses $t_m \gg t_p$.

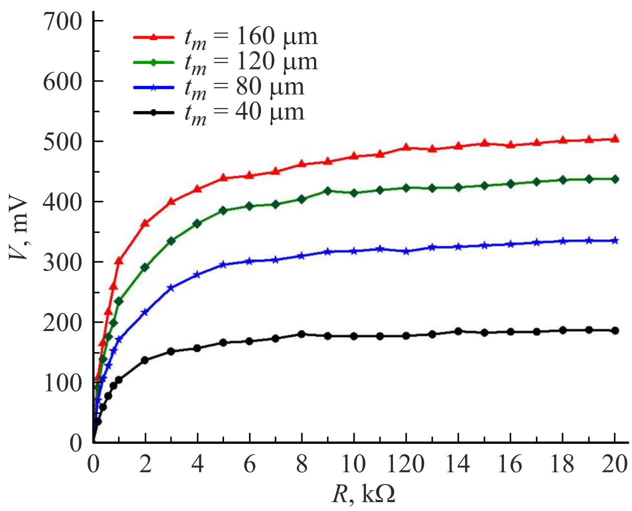

Figure 7. Dependences of the output voltage on the load resistance for samples with different thicknesses of the magnetostrictive layer at resonance frequencies.

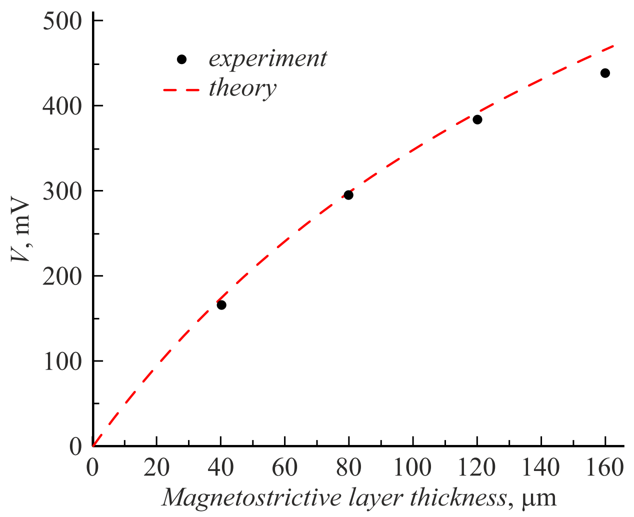

Figure 8. Dependence of the output voltage on the magnetostrictive layer thickness at resonance frequencies with load resistance $R = 5\,\mathrm{k\Omega}$. Piezoelectric layer thickness $t_p = 300\,\mu$m.

Conclusion

Three-layer magnetostrictive piezoelectric structures based on the AMAG 212N amorphous alloy and PZT 19 piezoelectric ceramics were demonstrated to have fine ME properties at low bias fields on the order of several kA/m. The ME characteristics of such structures depend strongly on the magnetostrictive layer thickness. The optimal bias field magnitude, which corresponds to the maximum ME response, increases with increasing thickness of the magnetostrictive layer. Specifically, an increase in thickness of the magnetostrictive layer from 40 to 160 μm is accompanied by an almost linear increase in magnitude of the optimal bias field from 1.28 to 3.42 kA/m. The frequency dependence of MEVC has a wide plateau in the low frequency region, where the MEVC value is virtually independent of frequency. Within this frequency range, the MEVC value increased by a factor of 3 (from 110 to 340 mV/A) as the magnetostrictive layer thickness increased from 40 to 160 μm. In the region of electromechanical resonance, the ME conversion coefficient has a peak at a structure Q factor on the order of $Q \cong 90$. The increase in resonance frequency with increasing thickness of the magnetostrictive layer is attributable to two mechanisms: a reduction in effective length of the structure and an increase in effective Young's modulus. Both mechanisms contribute to the mentioned increase in resonance frequency. The load characteristics were analyzed, and it was found that the output voltage depends linearly on load in the region of low resistances. However, with a further increase in load resistance (when its value becomes commensurate with the capacitive resistance of the structure), the dependence becomes nonlinear, asymptotically approaching saturation at load resistances much greater than the capacitive resistance. The saturation voltage increases nonlinearly with increasing thickness of the magnetic material: a sharp rise is observed at first, but then the growth rate decreases, and the voltage tends to saturation when the thickness of the magnetostrictive layer becomes much greater than the piezoelectric layer thickness. This allows us to conclude that ME structures based on a PZT 19 wafer and the AMAG 212N magnetostrictive amorphous alloy, which are produced in Russia, are on par with structures based on foreign alloys. Therefore, they are well suited for fabrication of devices utilizing the ME effect, such as gyrators, voltage dividers, and magnetic field sensors.

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Conflict of interest

The authors declare that they have no conflict of interest.

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