

Influence of the composition of a magnetoelectric composite on the „self-bias“ effect in hybrid structures

© N.N. Poddubnaya¹, D.A. Filippov², V.M. Laletin¹

¹ Institute of Technical Acoustics, National Academy of Sciences of Belarus, Vitebsk, Belarus

² Yaroslav-the-Wise Novgorod State University, Novgorod, Russia

E-mail: poddubnaya.n@rambler.ru

Received September 27, 2023

Revised November 14, 2023

Accepted November 14, 2023

The results of an experimental study of the dependence of the magnetoelectric response of ferromagnetic/magnetoelectric/ferromagnetic hybrid structures on the composition of the magnetoelectric composite at zero bias field are presented. Structures obtained by electrolytic deposition of Ni layers and alternating Ni/Co/Ni layers onto a magnetoelectric bulk composite based on nickel ferrite–lead zirconate titanate are considered. It was found that in a zero bias field the magnitude of the effect in structures with a Ni layer is significantly greater than in Ni/Co/Ni structures. It was concluded that the „self-bias“ effect is not associated with a stepwise change in magnetization, but is caused by a residual magnetic moment.

Keywords: magnetostriction, piezoelectricity, compositional structure, magnetoelectric effect, „self-bias“ effect.

DOI: 10.61011/TPL.2024.02.57988.19743

Searching for magnetoelectric (ME) materials applicable for the development of applied devices for electronics based on direct magnetic field transformation into electrical voltage is one of the practical problems of the condensed matter physics. Layered magnetostriction-piezoelectric structures, in which interconnection of magnetic and electrical properties is provided via mechanical interaction, which enables using them for creation of straintronics devices [1], in which the control of magnetic properties of the substance is performed under effect of electrical field, are the most promising for it. For creation of devices, one must use materials with the maximum efficiency of ME-transformation, especially in the regions of low frequencies, where the value of ME-coefficient is virtually independent on the frequency. For that purpose, the studies of ME-materials with different degree of connectivity and shape, obtained via different technologies, were carried out [2,3]. One of the factors that restrain application of ME-effect is the need for additional bias magnetic field for its emergence H_{bias} . It is because of the value of the effect is pro rata to the production of piezoelectric coefficient d by the piezomagnetic coefficient $q = d\lambda/dH$, where λ — magnetostriction. At the point, where magnetization is zero, the magnetostriction has an extremum, so $q = 0$. Because the magnetostriction is the function of magnetization, in order to obtain $q \neq 0$, one need to create magnetization, for which an additional bias magnetic field is used. However, „self-bias“ effect is observed in several structures, i.e. ME-effect occurs with zero magnetic bias field [4–10]. The works [4,7] present a theory, according to which the „self-bias“ effect is associated with the presence of the magnetization spike or, according to the terminology adopted in these works,

„the magnetization gradient“. However, the „self-bias“ effect occurs also in the absence of the magnetization spike [8,9]. The work [10] proposed a theory, according to which the „self-bias“ effect is associated with the residual magnetization M_r . In the present work, for validation of these theories, we performed experimental studies in hybrid structure of ferromagnetic metal (Ni)/magnetic electrical composite (PZT:NFO)/Ni and in the structure with stepped change of magnetization, consisting of alternating layers Ni/Co/Ni/PZT:NFO/Ni/Co/Ni. The volume ME-composite was made of lead zirconate-titanate powder PZT 23 and nickel ferrite $\text{NiFe}_{1.98}\text{Co}_{0.02}\text{O}_4$ (NFO) by conventional piezoceramic technology with PZT content by weight in the composite $m_{\text{PZT}}/m_{\text{ME}}$ (m_{PZT} — weight of PZT, m_{ME} — total weight of ME-composite) equal to 50, 60, 70, 80, 90%. The fabricated samples had the diameter of 8.7–8.8 mm and the thickness of $t^{\text{ME}} = 0.4$ mm. Electrodes were applied to the samples by chemical nickel metallization from chloride solution, and then were polarized perpendicularly to the plane.

Layers of Ni, as well as alternating layers of (Ni/Co/Ni) were applied on both sides of the volume composite for the fabrication of hybrid structures. The layers were obtained by electrochemical deposition from the nickel sulfamate and cobalt sulfate solutions by using the reversible current with the density of 3.5 A/dm^2 at the duration of pulses corresponding to the polarity of deposition/solution, 90/9 s. Measurements of the ME-effect were carried out by measuring the voltage U emerging on the sample under effect of variable H_{ac} and bias H_{bias} magnetic fields. The field H_{ac} was generated by Helmholtz coils and was 80 A/m at the frequency of 1 kHz. The value of ME-voltage

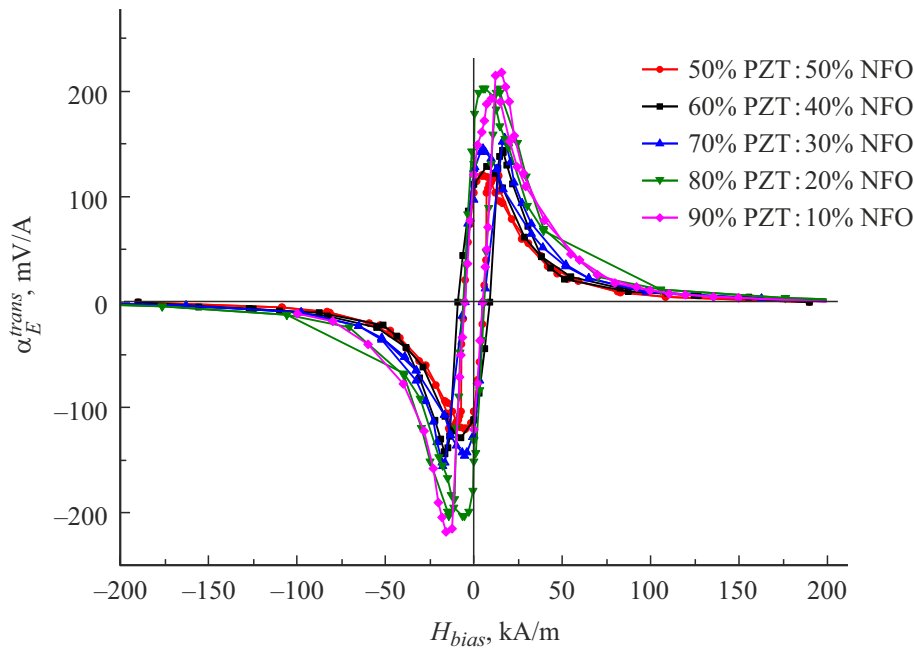


Figure 1. Field-effect dependence of MEVC for the Ni/PZT:NFO/Ni structure on the composition of the PZT:NFO composite. The thickness of Ni layer is equal to $100\ \mu\text{m}$.

coefficient (MEVC) was calculated by using the formula

$$\alpha_E = \frac{U}{t^{\text{ME}} H_{ac}}$$

Fig. 1 and 2 present field-effect dependencies for the hybrid structure Ni/PZT:NFO/Ni and the structure Ni/Co/Ni/PZT:NFO/Ni/Co/Ni with the transverse effect, when the magnetic fields lay within the sample plane, and the electrical field is oriented perpendicularly to the plane. The magnetic fields orientation within the sample plane allowed to ignore the demagnetization effects.

As shown in Fig. 1 and 2, the value of MEVC in the structure Ni/PZT:NFO/Ni is considerably higher than in the structure Ni/Co/Ni/PZT:NFO/Ni/Co/Ni, though the thickness of magnetostriction layers differs only by 20%. It is explained by the fact that Ni and ME-composite has a negative magnetostriction, and Co — a positive one. Carrying out the calculations similar to those in the work [11], for MEVC of hybrid structure caused by planar oscillations, with the transverse orientation of the fields, the following expression is obtained:

$$\alpha_E^{\text{trans}} = \frac{Y^{\text{ME}} d_{31}^{\text{ME}}}{\varepsilon_{33} \bar{Y} t} \times \frac{[Y^{\text{ME}} t^{\text{ME}} (q_{11}^{\text{ME}} + q_{12}^{\text{ME}}) + Y^{\text{Ni}} t^{\text{Ni}} (q_{11}^{\text{Ni}} + q_{12}^{\text{Ni}}) + Y^{\text{Co}} t^{\text{Co}} (q_{11}^{\text{Co}} + q_{12}^{\text{Co}})]}{[1 - 2k_p^2 (1 - Y^{\text{ME}} t^{\text{ME}} / \bar{Y} t)]} \quad (1)$$

Here Y^{ME} , Y^{Ni} , Y^{Co} , t^{ME} , t^{Ni} , t^{Co} — Young's moduli and thickness of PZT:NFO composite and the layers of Ni and Co, respectively, d_{31}^{ME} , q_{1i}^{ME} and ε_{33} — effective values of piezoelectric constants and dielectric permittivity of the

composite, q_{1i}^{Ni} , q_{1i}^{Co} — the values of piezoelectric constants of Ni and Co respectively, k_p^2 — square of electromechanical coupling factor, $t = t^{\text{ME}} + t^{\text{Ni}} + t^{\text{Co}}$ — total thickness of hybrid structure, $\bar{Y} = (Y^{\text{ME}} t^{\text{ME}} + Y^{\text{Ni}} t^{\text{Ni}} + Y^{\text{Co}} t^{\text{Co}}) / t$ — mean value of the Young's modulus of the structure. In the structure Ni/PZT:NFO/Ni ($t^{\text{Co}} = 0$) the first two members in square brackets at the numerator of the equation (1) have the same signs, which results in amplification of the ME-effect occurring in the volume ME-composite due to the layers of Ni, which results in increase of the effect versus that in the pure composite PZT:NFO. For the structure Ni/Co/Ni/PZT:NFO/Ni/Co/Ni the third member in square brackets at the numerator of the equation (1) has the opposite sign to that of the first two members, which results in decrease of the effect relative to the structure Ni/PZT:NFO/Ni.

It follows from Fig. 1 and 2 that the „self-bias“ is observed in both structures, i.e. MEVC is not zero at $H_{\text{bias}} = 0$. Fig. 3 presents dependences of the maximum MEVC α_E^{max} and MEVC at zero bias field α_E^0 on the ratio of weight of PZT m_{PZT} to the total weight of ME-composite m_{ME} .

According to Fig. 3, for the structure Ni/PZT:NFO/Ni, behavior of α_E^0 depending on the composition of the composite generally coincides with the behavior of α_E^{max} . The principal difference is that the maximum falls not within the composition of $m_{\text{PZT}}/m_{\text{ME}} = 90\%$, but within the composition of $m_{\text{PZT}}/m_{\text{ME}} = 80\%$. It is explained by the fact that the increase of $m_{\text{PZT}}/m_{\text{ME}}$ in the composition of the composite up to 90% results in considerable decrease of residual magnetization of M_r , which leads to significant decrease of piezomagnetic coefficient.

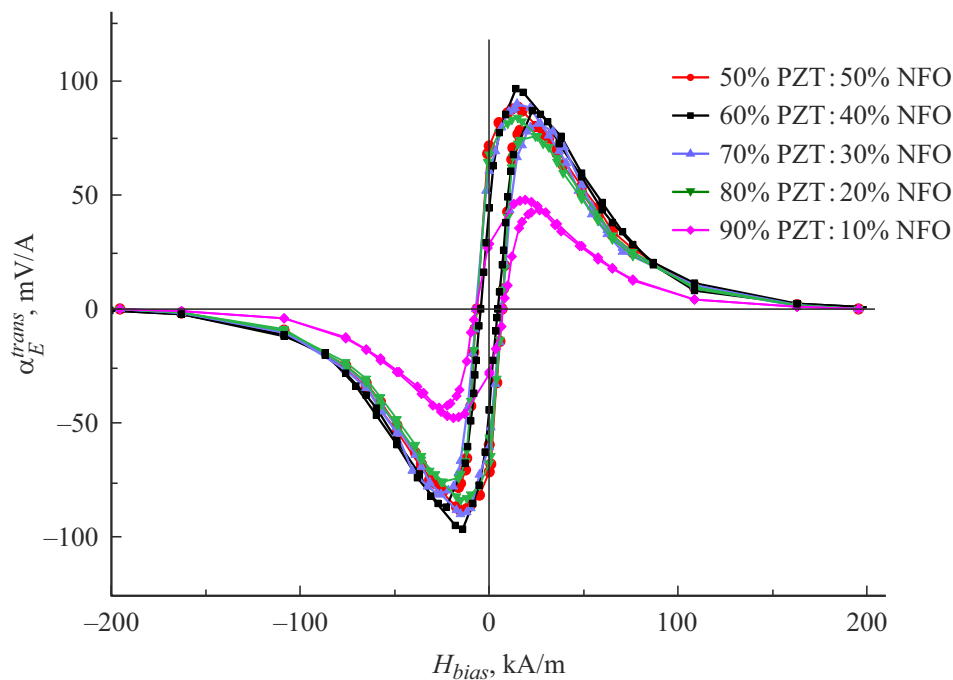


Figure 2. Field-effect dependence of MEVC for the Ni/Co/Ni/PZT:NFO/Ni/Co/Ni structure on the composition of the PZT:NFO composite. Thickness of metallic coatings: Ni — 20 μm , Co — 40 μm .

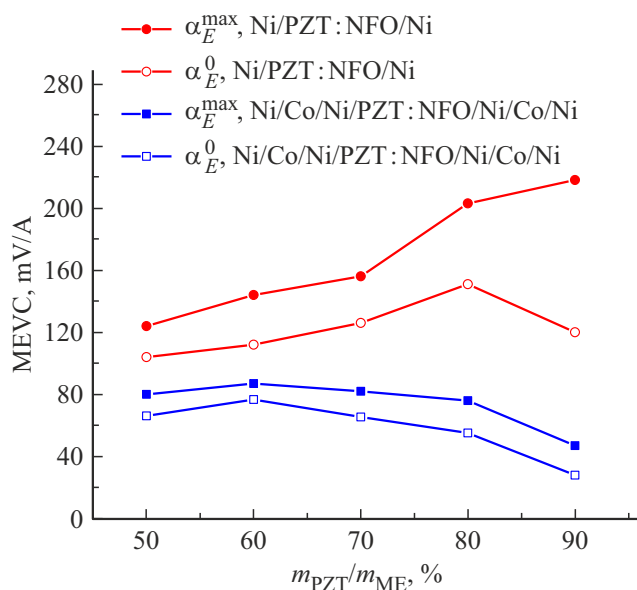


Figure 3. Dependence of the maximum value of MEVC α_E^{\max} and the value of MEVC at zero bias field α_E^0 on the composition of the composite.

It should be noted that for the composition $m_{\text{PZT}}/m_{\text{ME}} = 80\%$ the ratio is $\alpha_E^0/\alpha_E^{\max} = 74\%$, which enables using that structure in devices without the bias field. No expected effect increase in the structure with stepwise change of magnetization observed. Therefore, we may conclude that the „self-bias“ effect is not associated

with stepwise change of magnetization, and is caused by the presence of residual magnetization.

Funding

The paper was supported by the Belarusian Fundamental Scientific Research Foundation under grant F20MC-006. In the Yaroslav-the-Wise Novgorod State University the study was carried out against the grant from the Russian Science Foundation № 22-19-00763 (<https://rscf.ru/project/22-19-00763/>).

Conflict of interest

The authors declare that they have no conflict of interest.

References

- [1] A.A. Bukharaev, A.K. Zvezdin, A.P. Pyatakov, Yu.K. Fetisov, Phys. Usp., **61** (12), 1175 (2018). DOI: 10.3367/UFNe.2018.01.038279.
- [2] J. Yu, L. Bai, R. Gao, Process. Appl. Ceram., **14** (4), 336 (2020). DOI: 10.2298/PAC2004336Y
- [3] C.-W. Nan, M.I. Bichurin, S. Dong, D. Viehland, G. Srinivasan, J. Appl. Phys., **103** (3), 031101 (2008). DOI: 10.1063/1.2836410
- [4] U. Laletin, G. Sreenivasulu, V.M. Petrov, T. Garg, A.R. Kulkarni, N. Venkataramani, G. Srinivasan, Phys. Rev. B, **85** (10), 104404 (2012). DOI: 10.1103/PhysRevB.85.104404
- [5] Y. Zhou, D. Maurya, Y. Yan, G. Srinivasan, E. Quandt, S. Priya, Energy Harvest. Syst., **3** (1), 1 (2016). DOI: 10.1515/ehs-2015-0003

- [6] S. Liu, S. Liao, K. Wei, L. Deng, L. Zhao, H. Zou, *Battery Energy*, **2** (5), 20230005 (2023). DOI: 10.1002/bte2.20230005
- [7] M.I. Bichurin, O.V. Sokolov, V.S. Leontiev, R.V. Petrov, A.S. Tatarenko, G.A. Semenov, S.N. Ivanov, A.V. Turutin, I.V. Kubasov, A.M. Kislyuk, *Phys. Status Solidi B*, **257** (3), 1900398 (2020). DOI: 10.1002/pssb.201900398
- [8] Z. Ou, C. Lu, A. Yang, H. Zhou, Z. Cao, R. Zhu, H. Gao, *Sensors Actuators A*, **290**, 8 (2019). DOI: 10.1016/j.sna.2019.03.008
- [9] J. Zhang, D. Chen, K. Li, D.A. Filippov, B. Ge, Q. Zhang, X. Hang, L. Cao, G. Srinivasan, *AIP Adv.*, **9** (3), 035137 (2019). DOI: 10.1063/1.5078716
- [10] Y. Liu, J. Zhang, P. Zhou, C. Dong, X. Liang, W. Zhang, T. Zhang, N.X. Sun, D. Filippov, G. Srinivasan, *J. Appl. Phys.*, **126** (11), 114102 (2019). DOI: 10.1063/1.5112024
- [11] D. Filippov, Y. Liu, P. Zhou, B. Ge, J. Liu, J. Zhang, T. Zhang, G. Srinivasan, *J. Compos. Sci.*, **5** (11), 287 (2021). DOI: 10.3390/jcs5110287

Translated by D.Kondaurov