Magnetoelectric and magnetostriction characteristics of symmetric three layered structures of nickel - lead zirconate titanate – nickel and permendure – lead zirconate titanate – permendure

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Abstract. The magnetoelectric and magnetostriction characteristics of the symmetric three-layered structures are presented. The composite structures were formed like sandwich structures with a piezoelectric layer between two identical magnetostrictive layers using epoxy adhesive. An analytical expression for magnetoelectric voltage coefficient in terms of parameters for magnetostrictive and piezoelectric subsystems is presented. On the basis of this expression determination of magnetostrictive characteristics via magnetoelectric response was offered. This procedure was tested for these three layered structures using the field dependences of the magnetoelectric response. The magnetostrictive characteristics obtained are in good agreement with results, obtained by other methods.

1. Introduction

Composite multiferroics have shown a good magnetoelectric coupling at the room temperature [1, 2]. The magnitude of magnetoelectric (ME) effect in composite multiferroics is several orders of magnitude higher than in single phase multiferroic materials in which the magnetoelectric interaction is due to a change in the spin-orbit interaction of the electron under the action of an external electric field [3]. Since the spin-orbit interaction is a relativistic effect, this leads to small ME coupling. In composite multiferroics, the ME effect is due to the mechanical interaction between magnetostrictive and piezoelectric subsystems or, so called strain induced mechanism [4]. The interconnection of the magnetic and electrical characteristics of multiferroics allows one to create on their basis devices with dual control of both electric and magnetic fields. These are inductors, tunable by both electric and magnetic fields [5, 6], dual electric and magnetic field tunable high-frequency devices [7, 8], gyrators capable of direct conversion of current to voltage [9, 10]. The origin of ME coupling in composites arises...
due to the elastic interaction between the magnetostriction and piezoelectric subsystems; therefore, the magnitude of the ME effect directly depends on the magnetostriction and piezoelectric parameters of the subsystems. Recently in series of works, theoretically shown and experimentally confirmed that the magnitude of the magnetic resonance line shift under the action of an electric field is proportional to the product of the piezoelectric module $d$ by the magnitude of the saturation magnetostriction $\lambda_S$ [11, 12], power conversion efficiency in gyrator is proportional to the piezomagnetic module $q = d \lambda / dH$ [9, 10], the change in the inductance of a coil under the action of an electric field is also proportional to the product $d \lambda$ [13]. Thus, the piezoelectric module and magnetostriction constant are the most important parameters that determine the possibility of using composite multiferroics in straintronics. Methods for measuring a piezoelectric module are well known and fairly accurate. For example, the piezoelectric constant $d_{31}$ is determined quite strong using the resonance - antiresonance method [14]. The magnetostriction of ferromagnetic materials is usually measured using a strain gauge, glued to the surface of the sample. The measurement accuracy in this case is $\sim 10^{-6}$ in the field range up to 5 kOe [15]. This measurement method is not entirely convenient, since it requires gluing a strain gauge and magnetic material, which introduces an additional measurement error. It leads to the fact that this method does not allow measuring the magnetostriction of films whose thickness is commensurate with the thickness of the strain gauge. Besides, due to the smallness of the effect, it does not allow one to measure magnetostriction in the region of small fields. The low-field region is of particular interest because it exhibits a non-linear effect, on the basis of which sensitive magnetic field sensors can be created [15, 16]. Since the ME effect is directly related to the appearance of an electric field in a piezoelectric due to deformations caused by magnetostriction of a magnet in a magnetic field, it is of interest to use the ME response to determine magnetostriction. In earlier works [17, 18] this method was developed to determine the magnetostriction of bulk composites. In this paper, we have expanded this method for determining magnetostriction via the magnetoelectric response for layered composites.

2. Theory: magnetoelectric response and magnetostriction

Magnetoelectric response in composite multiferroics is caused by mechanical coupling between the magnetostrictive and piezoelectric phases by strain. In the case of the direct ME effect mechanical deformations occurs by magnetic field in the magnetostrictive phase and are transferred to the piezoelectric phase by mechanical interaction at the interface, that leads to change the electric polarization and produces electric voltage between electrodes of the sample. In the case of converse ME effect an electric field produces mechanical stresses in the piezoelectric phase that are transmitted through the interface into the magnetostrictive phase, which leads to a change in the magnetization of the sample. This change leads to occurrence of voltage on the coil wound on the sample [19]. The ME response for the direct ME effect is characterized by ME voltage coefficient (MEVC) $\alpha_E$, which is determined by ratio of the average value electric field $<E>$ to the ac magnetic field $H$, i.e.

$$\alpha_E = <E>/H = (U/p_t)/H$$ \hspace{1cm} (1)

where $U$ is the electric voltage, and $p_t$ is a piezoelectric layer thickness.

For the ME effect for three layered symmetric structures the theory predicts the following expression for the low frequency MEVC

$$\alpha_E = \frac{p_Y p_{d31}(m_{q11}+m_{q12})}{p_{E3}q_0} \frac{m_Y m_t}{(1-K_p^2 q_Y^2 m_t)\left(m_t p_t + p_Y p_t^2\right)},$$ \hspace{1cm} (2)

where $p_Y, m_Y$ are the Young’s modules of the piezoelectric and magnetostrictive phases; $p_{d31}, m_{q11}, m_{q12}$ are the piezoelectric and piezomagnetic coefficients; $p_t, m_t$ are the thickness of the
piezoelectric and the two magnetostrictive layers; $\varepsilon_3$ is permittivity, $\varepsilon_0$ is dielectrically constant and

$$K_p^2 = \frac{\rho_Y (p_{d31}^m)^2}{\varepsilon_3}$$

is the squared coefficient of electromechanical coupling.

Using the fact that $K_p^2 \ll 1$, we can rewrite Equation (2) in the first approximation in the form

$$\alpha_E = \frac{\rho_Y (p_{d31}^m)^2}{\varepsilon_3 \varepsilon_0} \frac{m_Y m_t}{m_Y m_t + \rho_Y \rho_f}.$$

(3)

The piezomagnetic coefficients $m_{q11}$ and $m_{q12}$ in Equation (3) are defined as follows:

$$m_{q11}(H_{bias}) = \frac{\partial \lambda_1}{\partial H_1} \bigg|_{H=H_{bias}},$$

(4)

$$m_{q12}(H_{bias}) = \frac{\partial \lambda_1}{\partial H_1} \bigg|_{H=H_{bias}},$$

(5)

where $\lambda_1$ is a longitudinal magnetostriction, $\lambda_1$ is transverse magnetostriction, $H_{bias}$ is bias magnetic field. In Equation (3) only terms $m_{q11}$ and $m_{q12}$ have a strong dependence on magnetic field. The terms, including $\rho_Y$, have a weak dependence on magnetic field. The remaining terms in expression (3) for the MEVC are independent of the bias field. Thus, the field dependence of the MEVC is determined by the dependence of the piezomagnetic modules $m_{q11}$ and $m_{q12}$ on the bias field. This allows us to determine the magnetostriction from the field dependence of MEVC $\alpha_E(H_{bias})$ using the relationship:

$$\lambda_1(H_{bias}) + \lambda_1(H_{bias}) = \int_0^{H_{bias}} m_{q11} + m_{q12} dH = \frac{1}{C} \int_0^{H_{bias}} \alpha_E(H) dH,$$

(6)

where $C = \frac{\rho_Y (p_{d31}^m)}{\varepsilon_3 \varepsilon_0} \frac{m_Y m_t}{m_Y m_t + \rho_Y \rho_f}$ is a constant coefficient for given structures. At the first approximation we can use the fact $\lambda_1 = -\nu \lambda_1$. Using this relation we get finally for the magnetostriction follows expression:

$$\lambda_1(H_{bias}) = \frac{1}{C(1-\nu)} \int_0^{H_{bias}} \alpha_E(H) dH.$$

(7)

Saturation magnetostriction coefficient $\lambda_{1s}$, according to Equation (7) will be determined as:

$$\lambda_{1s} = \frac{1}{C(1-\nu)} \int_0^\infty \alpha_E(H) dH.$$

(8)

Using this determination we get the expression for magnetostriction coefficient in the following form:

$$\lambda_1(H_{bias})/\lambda_{1s} = \int_0^{H_{bias}} \alpha_E(H) dH / \int_0^\infty \alpha_E(H) dH.$$

(9)

Expressions (8) and (9) allow us to determine the magnetostriction of a magnetic phase by the field dependence of the MEVC for the composite structures without any addition devices. It is very important for the thin magnetic films on the piezoelectric substrate in the region of low bias magnetic field. For weak magnetic field the value of magnetostriction is very small; therefore there are many difficulties for its determination using resistance strain gauge. The method for the determination of the magnetostriction using ME response has a very high sensitivity, more than strain gage factor, therefore one can use for the small values of the magnetic field.

3. Details of experiment

The ME effect in structures was studied by measuring the voltage arising on samples in ac magnetic field $H_{ac}=1$ Oe and in bias field, the strength of which $H_{bias}$ changed in the range of $H_{bias}=0 – 2000$ Oe. The samples were three-layer Nickel – Lead Zirconate Titanate (PZT) – Nickel and Permendure
(Pe) – Lead Zirconate Titanate – Permendure structures in the form of a parallelepiped, obtained by the gluing using an epoxy. The length of the samples was 20 mm, the width was 4.5 mm, the thickness of the PZT plate was $p_t = 0.3$ mm, the thickness of magnetic plates was $m_t = 0.15$ mm of each.

Simultaneously were prepared three-layer Nickel-Quartz-Nickel structures in the form of a parallelepiped, obtained by the method of gluing using epoxy and electrolytic deposition. The length of the samples was 20 mm, the width was 4.5 mm, the thickness of the quartz plate was $p_t = 0.5$ mm. For electrolytic deposition structures, the thickness of nickel plates was $m_t = 0.25$ mm of each; therefore, these structures can be considered as bulk samples. The thickness of the nickel layer for structures obtained by galvanic deposition was equal $m_t = 10 \, \mu m$, therefore, these structures were the thin film structures.

The frequency dependence of MEVC has the resonance character. At the low frequencies MEVC does not depend on the frequency. To measure the field dependency of MEVC ac magnetic field with frequency $f=1 \, kHz$ was used. The ME coefficient is given by the expression:

$$\alpha_E = \frac{u}{p_t H_{ac}} \cdot$$

where $u$ is the induced voltage between the plates of the sample, $p_t$ is the thickness of the piezoelectric layer, $H_{ac}$ is the value of the applied ac magnetic field.

4. Results and discussion

The measured low frequency MEVC for Ni – PZT – Ni structures and Pe – PZT – Pe are presented in figure 1a and figure 1b respectively.

![Figure 1](image1.png)

**Figure 1.** Field dependencies of MEVC for three-layered symmetric structures.

As can be seen from figure 1, both field dependences have a typical characteristic for low frequency MEVC. The maximum value of MEVC for Ni – PZT – Ni structure is $\alpha_{E,max}=0.43 \, V/cm \cdot Oe$ and observed at the bias magnetic field $H_{bias}=100 \, Oe$, while maximum value for Pe – PZT – Pe structure observed at the bias magnetic field $H_{bias}=150 \, Oe$ and its value is $\alpha_{E,max}=0.41 \, V/cm \cdot Oe$. Magnetostriction characteristics for these structures were calculated using Equation 8. For the calculations following values were used for material parameters: $\psi Y= 67 \, GPa$, $p_t=0.3 \, mm$, $p d_{31}=-175 \, pC/N$, $\varepsilon_0=8.85 \cdot 10^{-12} F/m$, $m Y_{NI}= 210 \, GPa$, $m Y_{Pe}=207 \, GPa$, $m t=2*0.25 \, mm$. The calculated magnetostriction curves are presented in figure 2.
As can see from figure 2, presented curves are in good agree with results obtained by using resistance strain gauge [22]. Thus, the procedure proposed here provides one with a unique method for determination of magnetostriction characteristic magnetic phase in the layered multiferroic.

We also utilized this procedure for the determination magnetostriction of thin films, fabricated by galvanic deposition of nickel on quartz piezoelectric substrates. The samples were three-layer Nickel-Quartz-Nickel structures in the form of a parallelepiped, obtained by the method of gluing with using epoxy and electrolytic deposition. The length of the samples was 20 mm, the width was 4.5 mm, the thickness of the quartz plate was \( t_p = 0.5 \) mm. The thickness of nickel plates the gluing structures was equal \( t_m = 0.25 \) mm; therefore, these structures can be considered as bulk samples. The thickness of the nickel layer for structures obtained by galvanic deposition was \( m_t = 10 \) \( \mu \)m, therefore, they were the thin film structures. The field dependency of MEVC for the Ni – Q – Ni structures obtained by epoxy glue and electrolytic deposition is presented in figure 3.

As can be seen from figure 3 MEVC has a maximum for both samples, but for epoxy glue structures this maximum occurs when the value of bias field is \( H_{ac}=150 \) Oe, while for electrolytic deposition structures this maximum is observed at the value of bias field is equal to \( H_{ac}=400 \) Oe. This fact demonstrates the difference of field dependence of magnetostriction between bulk sample and thin film structures. The field dependencies of magnetostriction obtained by using Equation (9) are
presented in Fig. 4. It shows that the magnetostriction for bulk sample has saturation in magnetic field near 500 Oe. These results are in good agreement with those reported in previous work [23]. For the electrolytic deposition sample the magnetostriction tends to saturation at fields above 1500 Oe.

5. Conclusion

The study of the field dependence of MEVC in composite magnetostrictive – piezoelectric structures allows one to estimate the magnetostriction and its field dependence reveal the features expected for ferromagnetic thin films. The magnetostriction coefficient of nickel thin films tends to saturate at the value of magnetic field more than three times higher than the values of saturation magnetic field for bulk samples.

Acknowledgment

The work was jointly supported by the Russian Foundation for Basic Research and Belarusian Foundation for Basic Research (Russian project no. 18-52-00021 and Belarusian project no. F18R-300). The research at Oakland University was supported by grants from the National Science Foundation (DMR-1808892, ECCS-1923732).

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