New way the determination of magnetostrictive parameters composite multiferroics using the magnetoelectric response

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Abstract. The new way the determination of the thin films magnetostriction characteristics using the magnetoelectric response of composite multiferroic is supposed. The analytical expression for the magnetostriction coefficient using the field dependency of the magnetoelectric voltage coefficient is obtained. The experimental data for the bulk composite lead zirconate titanate – nickel ferrite is presented. It is shown, that the data, obtained by supposed way, are in good agreement with previous data, obtained by another way.

1. Introduction

Composite multiferroics are the perspective materials for the using in the recently developed new direction of electronics – straintronics [1]. The composite multiferroics are of particular interests due to their potential applications for dual H- and E-tunable electronic devices [2]. Dual control in such devices is due to the presence of a magnetoelectric effect (ME) in composite multiferroics, which consists in changing the polarization of the sample in a magnetic field (direct ME effect) or, vice versa, changing the magnetization under the action of an electric field (converse or inverse ME effect). The magnitude of ME effect in composite multiferroics is than several orders higher that its magnitude in monocrystals. The magnetostriction and the piezoelectric modulus are the more important parameters of these structures. ME effect in composites arises as a result of the elastic interaction between the magnetostriction and piezoelectric subsystems; therefore, the magnitude of the effect directly depends on the magnetostriction and piezoelectric modulus. These parameters are determined the characteristics of devices based on the ME effect. For example, the change of inductance of the coil by applied electric field is proportional to the product of the piezoelectric coefficient \( d \) and the magnetostriction coefficient \( \lambda \) [3–4]. The shift of the magnetic resonance line in an electric field is also proportional to the product \( d \lambda \) [5]. The magnitude of the piezoelectric modulus is determined quite accurately, for example, by the resonance-antiresonance method. The determination of magnetostriction has certain difficulties. They arise due to the smallness of the effect in the region of small fields.
region is especially interesting because in this region a nonlinear ME effect appears, on the basis of which it is possible to create a whole range of magnetic field sensors with sensitivity much higher than the sensitivity of Hall sensors [6, 7]. To measure the magnetostriction, as a rule, strain gauges are used, which introduce an additional error in the magnitude of the deformation that occurs under the action of a magnetic field. The difficulties of determining the magnitude of magnetostriction are especially pronounced when measuring the magnetostriction of thin films grown on a piezoelectric substrate, which have proven themselves in devices based on electrical control of magnetic resonance [8, 9]. To estimate the effect in such structures, the value of the magnetostriction coefficient for bulk samples is used. However, as shown by the results of experiments [10] the ME effect in magnetostrictive – piezoelectric structures obtained by electrolysis deposition nickel thin film on piezoelectric substrate has some differences compared to the effect in bulk structures. These features are due to different field dependency of magnetostriction thin films and bulk samples. In this paper, a new method is proposed for determining the magnetostriction of composite multiferroics based on measuring the ME response of the system.

2. Theory: magnetostriction from magnetoelectricity

Magnetoelectric (ME) effect in composite multiferroics is caused by the elastic interaction of magnetostrictive and piezoelectric subsystems. Under the action of a magnetic field, mechanical deformations occur in the magnetostriction phase, which pass through a mechanical coupling into the piezoelectric phase and cause a change in the polarization of the sample, which leads to the appearance of an electrical voltage between the electrodes. As a parameter that numerically determines the ME response of the system, they use the ME voltage coefficient (MEVC) \( \alpha_E \) defined as the ratio of the average electric field strength in the sample \( \langle E \rangle \) to the magnitude of the magnetic field \( H \) that caused it, i.e. \( \alpha_E = \langle E \rangle / H \). The average field value is calculated using the ratio \( \langle E \rangle = U / t \), where \( U \) is the measured electrical voltage, which occurs on the plates when the sample is placed in an alternating magnetic field \( H \) and the constant magnetizing field \( H_{bias} \), and \( t \) is the sample thickness. The theory of ME effect in bulk composites at the electromechanical resonance region was first presented in [11, 12]. Using the effective parameters method authors was obtained the expression for MEVC at low frequency region in following form:

\[
\alpha_E = \frac{Y d_{32} q_{33}}{\varepsilon_{33} \varepsilon_0},
\]

where \( Y, d_{33}, q_{33} \) and \( \varepsilon_{33} \) are the effective values of the Young's modulus, piezoelectric, piezo-magnetostriction coefficients, and dielectric permeability of the composite, \( \varepsilon_0 \) is the dielectric constant.

The piezomagnetostriction coefficient \( d \) in Exp. (1) is determined by the relation

\[
q_{33} = \frac{d \lambda_t}{dH} \bigg|_{H=H_{bias}},
\]

where \( \lambda_t \) is the longitudinal magnetostriction. Since the magnetostriction, and consequently, the piezo-magnetostriction coefficient, depends on the magnetic field, the dependence of the ME response of the system on the bias magnetic field, the so-called field dependence of MEVC, is observed. This dependence allows us to determine the magnetostriction of composite multiferroic. Using equation (1) and equation (2) we get the following expression for magnetostrictive coefficient:

\[
\frac{d \lambda_t}{dH} = \frac{\varepsilon_{33} \varepsilon_0 \alpha_E(H)}{Y d_{33}}.
\]
Integrating equation (3) with respect to $H$ and using the fact that the parameters $Y, d_{33}$ and $e_{33}$ are independent of the magnetic field, for the magnetostriction coefficient we obtain the following expression:

$$\lambda(H) = \int_0^H C \alpha_E(H')dH', \quad (4)$$

where $C = \frac{\varepsilon_{33} \varepsilon_0}{Y d_{33}}$ is the constant coefficient for given samples. This coefficient is independent of the magnetic field, but its value depends on the percentage of ferrite and piezoelectric in the composite.

Thus, using relation (3) and the field dependence of MEVC, we can obtain the dependence of the magnetostriction of the composite multiferroic. The value of this coefficient can determine using the measure the value Young’s modules, piezoelectric modules and dielectric permeability of composite. Using equation (4), we get for the saturation value of magnetostriction the expression in the form:

$$\lambda_\infty = \int_0^\infty \alpha_E(H')dH'. \quad (5)$$

If the value of the saturation magnetostriction is known, then the dependence of the magnetostriction on the magnetic field is easy to determine using the relation:

$$\lambda(H) = \lambda_0 A(H)/A_s, \quad (6)$$

where $\lambda = \int_0^H \alpha_E(H')dH'$ and $A_s = \int_0^\infty \alpha_E(H')dH'$ are the integral characteristics of MEVC [13].

Thus, the integral characteristic of MEVC can to obtain the magnetostriction characteristics of composite multiferroics.

3. Details of experiment
Bulk composite ME materials were fabricated by sintering the mixture of nickel ferrite (NFO) and piezoelectric lead zirconate titanate (PZT) powders. With this aim, the samples were prepared in $(1 - x)\text{PZT23—}x\text{NiFe}_2\text{O}_4$, where parameter $x$ is a weight fraction and equal to 0.1, 0.3, 0.5 and 0.7. The samples were sintered in crucibles with a lead-containing filling for 2 h at a temperature of 1240°C. The cooling rate was not higher than 50°C/h. The electrodes were deposited by means annealing of silver paste at a temperature of 650°C for 20 min. All the samples were shaped as disks with a diameter of 8.8 mm and thickness of 0.8 mm. For polarization of the samples five modes were used: modes 1–4, polarization in the fields with intensity of 1, 2, 3, and 4 kV/mm at room temperature; and mode 5, polarization in the field with intensity of 4 kV/mm at a temperature of 100°C. The polarization of materials was carried out at a temperature of 60-100 °C for two hours in an electric field of 4 kV/mm, followed by cooling in this field to room temperature for half an hour. The piezoelectric module $d_{33}$ was measured with a YE2730A $d_{33}$ METER instrument. The Young's modulus was defined using the resonance – antiresonance method. The dielectric permeability was determined from measure of the capacitance, which was measured at a frequency of 1 kHz.

The longitudinal ME effect was studied, when the direction of the electric polarization of the sample and magnetic fields were parallel to each other. The linear ME effect was investigated by measuring the voltage arising on the sample when placed in an alternating magnetic field and a bias magnetic field. The amplitude of the alternating field was 1 Oe at a frequency $f = 1$ kHz. The magnitude of the bias field varied from 0 to 5000 kOe.

4. Results and discussion
The measurement results of physical parameters of composite for different ferrite content are presented in Table 1. As can see from these data, the physical parameters demonstrate nonlinear dependencies on ferrite content. Young’s modules increase with increasing ferrite content from...
65 GPa for pure PZT to 155 GPa for pure nickel ferrite. The dielectric permeability value for PZT equals 1750, with increasing ferrite content this value decreases from 660 for 10% NFO content to 60 for 70% NFO content. The piezoelectric module value $d_{33}$ for PZT equals 340 pC/N; with increasing ferrite content this value decreases from 200 pC/N for 10% NFO content to 3 for 70% NFO content.

Table 1. The dependence of the parameters of the composite multiferroic on the content of ferrite and piezoelectric.

| Contain of ferrite, % | Young’s modules, GPa | Piezoelectric modules, $d_{33}$, pC/N | Dielectric permeability, $\varepsilon$ |
|----------------------|----------------------|-------------------------------------|----------------------------------|
| 10                   | 93                   | 198                                 | 660                              |
| 30                   | 115                  | 79                                  | 340                              |
| 50                   | 134                  | 22                                  | 140                              |
| 70                   | 147                  | 3.1                                 | 60                               |

In figure 1 the fields dependencies of MEVC and in figure 2 of the curves of magnetostriction, obtained by integration of the MEVC dependencies for different content of the composite, are presented. The integration of MEVC dependencies was performed using the Origin software product.

Figure 1. The fields dependencies of MEVC for different ferrite content.

Figure 2. The magnetostriction of NFO-PZT composite multiferroic for different ferrite content.

As can see from figure 1, field dependency of MEVC has a maximum. The value of magnetic field $H_{\text{max}}$, corresponding maximum MEVC, depends on the content ferrite in composite and increase with increasing this content. The maximum value of MEVC $\alpha_{E,\text{max}}$ depends also from the content ferrite in composite. The maximum of this value $\alpha_{E,\text{max}}=0.125$ V/cmOe corresponding to composite with 50% content of nickel ferrite. The curves of magnetostriction, which are shown on figure 2, have a typical dependency for nickel contain composite. The magnetostriction has a negative sign and increase with increasing the content ferrite in composite. The values of saturation magnetic field are also increase from 1.8 kOe to 2.8 kOe with increasing the content ferrite in composite from 10% to 70%. The saturation magnetostriction is increase with increasing the ferrite content in composite from $-2$ ppm at 10% NFO in composite to $-17$ ppm for 70% NFO in composite. It is good agreement with previously experiments, which shown that for pure polycrystalline nickel ferrite the saturation magnetostriction equals $\lambda_s=-26$ ppm.

5. Conclusion
The new way determination of magnetostriction of composite multiferroics is suppossed. It is shown, that the field dependence of MEVC makes it possible to obtain the dependence of the magnetostriction of a composite multiferroic in a wide range of magnetic fields. The experimental results, obtaining
with using this way for determination of nickel ferrite-PZT composite magnetostriction are in good agreement with data, obtained previously another way. This way is a perspective for determination magnetostriction thin films on piezoelectric substrate.

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