Observation of the inverse magnetoelectric effect in PZT/FeCuNbSiB two-layered composite structures

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Abstract. In recent years, much attention has been paid to the study of magnetoelectric (ME) effects in solids, which was caused by their potential practical applications. Of particular interest are composite multiferroic materials containing ferroelectric and ferromagnetic phases, since significant ME responses may occur in them. This paper is devoted to the study of the inverse ME effect in a two-layered composite sample consisting of a piezoelectric Pb(Zr, Ti)O$_3$ (PZT) plate and an amorphous FeCuNbSiB ferromagnetic ribbon. It was found that the inverse ME effect depends on the magnitudes of the external magnetic field $H$, the electric voltage $U$ and on the mutual orientation of $H$ and $U$.

1. Introduction

The magnetoelectric (ME) effect is defined as the connection between magnetic and electric fields in a substance [1]. It can be observed as a change of an electric polarization of a material under the magnetic field action (the direct ME effect) or as a change of its magnetization upon application of an electric field (the inverse ME effect). The first theoretical consideration of the ME effect was made by L. Landau and E. Lifshitz in 1956. This effect is described as a member in the expression for Helmholtz free energy $F$, linear in electric, $E$, and magnetic, $H$, fields:

$$ F_{ME} = -\alpha_{ik} E_i H_k $$  \hspace{1cm} (1)

Magnetization and polarization of a material can be expressed as partial derivatives of Helmholtz free energy:

$$ M_k = -\frac{\partial F}{\partial H_k}, \quad P_i = -\frac{\partial F}{\partial E_i} $$  \hspace{1cm} (2)

Thus, when $H = 0$, an electric field induces magnetization in a material:

$$ M_k = \alpha_{ik} E_i $$  \hspace{1cm} (3)

And, when $E = 0$, a magnetic field induces electric polarization:

$$ P_i = \alpha_{ik} H_k $$  \hspace{1cm} (4)
The ME effect may occur only in crystals that possess specific symmetry properties. In particular, such crystal must be invariant to IR (spatial and time) inversion. In 1959 I. Dzyaloshinskiy predicted that the ME effect should take place in a single-crystal Cr2O3 [2]. In 1960 D. Astrov detected the change of magnetization in this crystal under the action of an electric field, that is, the inverse ME effect [3].

The ME effects arising in single-phase crystals are relatively small and occur only at low temperatures, which limits their practical application. To achieve significant ME responses, multiferroic materials are produced by combining ferroelectric and ferromagnetic phases. The concept of two-phase multiferroic materials was proposed by van Suchtelen in 1972 [4]. The mechanism of the direct ME effect in such structures is as follows. When a composite material is placed in an external magnetic field \( H \), its ferromagnetic component is deformed due to the magnetostrictive effect, and this deformation transfers to the piezoelectric component mechanically coupled to the ferromagnetic one. Due to the piezoelectric effect, the electric polarization \( P \) of the piezoelectric component changes. In the case of the inverse ME effect, the ME response is the result of the inverse piezoelectric effect and the inverse magnetostrictive effect. Multiferroic materials in which ME effects take place may find application in highly sensitive field sensors [5], memory storage elements [6], autonomous sources of energy [7].

In this work the amorphous FeCuNbSiB ribbon was used as the ferromagnetic component in the composite material. It is known that amorphous alloys deserve attention due to their unique soft magnetic properties, high mechanical strength and corrosion resistance. The choice of an amorphous FeCuNbSiB composition ribbon was stimulated by the optimal combination of its high saturation magnetization and magnetostriction coefficient [8].

\( \text{Pb(Zr,Ti)}_3 \text{O}_3 \) (PZT) was used as the piezoelectric component. PZT is a ferroelectric material obtained in the 1950’s [9]. Ferroelectrics are a subgroup of piezoelectrics that possess a spontaneous polarization and have hysteresis behavior of polarization in an external electric field. They consist of domains with different local polarizations, which can be aligned by application of an electric field. When the electric field is removed, a ferroelectric has a non-zero remanent polarization. PZT compounds are notable for their high dielectric constants and piezoelectric coefficients. Ferroelectric materials have a large practical application, including in memory devices, pressure sensors, capacitors, etc.

The purpose of this work is the magneto-optical study of the inverse magnetoelectric effect in a two-layered composite sample consisting of a piezoelectric \( \text{Pb(Zr,Ti)}_3 \text{O}_3 \) plate and an amorphous FeCuNbSiB ferromagnetic ribbon.

2. Sample and experimental method

2.1. Sample under study

The object of our study was a two-layered composite sample consisting of an amorphous FeCuNbSiB ferromagnetic ribbon rigidly mounted on a PZT piezoelectric plate by using the polymer adhesive BF6. The thicknesses of the ferromagnetic and the piezoelectric layers were 30 and 200 \( \mu \)m, correspondingly. The length and the width of the sample were 8.5 and 4 mm. The schematic picture of the sample is shown in figure 1.

The long side of the amorphous layer coincided with the length of the initial amorphous ribbon used for producing the sample. The magnetostrictive coefficient \( \lambda_S \) of the amorphous ribbon was about \(+2\times10^{-5}\). The piezoelectric plate was manufactured in such a way that, when an electric voltage \( U \) was applied perpendicular to its plane, an arising deformation was directed parallel to its long side.

2.2. Experimental procedure

The technique of the inverse ME effect measuring was as follows. The magnetization curves for the amorphous ribbon were measured at an electrical voltage \( U \) of various values (0, 75, 100, 160, 200 V) applied to the piezoelectric plate. The measurements were performed on the magneto-optical
magnetometer using the magneto-optical transverse Kerr effect (TKE). TKE is a change of the intensity of light reflected by a sample when an external magnetic field is applied parallel to the sample’s surface and perpendicular to the plane of light incidence.

![Figure 1. The sample under study. The arrows denote the magnetic field \( H \) orientations: a – parallel to the long side of the sample, b – parallel to the short side.](image)

To increase the sensitivity of the magneto-optical method, the modulation method of the registration of magneto-optical signals was used. This made it possible to eliminate the effect of fluctuations in the intensity of light and the photocurrent of the optical receiver. Since this method is differential, its sensitivity is 2-3 orders of magnitude higher than the static method’s sensitivity. As a result, it allows detecting changes of the light intensity reflected from a sample during its remagnetization with an accuracy of \( 10^{-5} \). To exclude the impact of edge effects, the measurements of magnetization curves were carried out in the center of the sample. The diameter of the illuminated area of the sample was equal to 1 mm.

The magnetization curves \( M/M_s(H) \), where \( M_s \) is the saturation magnetization of the sample, were measured in an external magnetic field \( H \) applied parallel to the plane of the sample and parallel or perpendicular to its long side. The voltage \( U \) was applied perpendicular to the plane of the PZT plate, and the emerging mechanical deformation was parallel to its length. The preliminary measurements at \( U = 0 \) revealed that the saturation field of the amorphous layer is smaller when the external magnetic field is applied parallel to the long side of the sample. This means that the axis of easy magnetization of the ribbon is parallel to its length, which is caused by the positive value of the magnetostrictive coefficient \( \lambda_s \).

To estimate the inverse ME effect coefficient \( \alpha \), the formula given in [10] was used, according to which:

\[
\alpha = \Delta B / E = \Delta B / (U/d), \text{[G/cm/V]} \tag{5}
\]

where \( \Delta B \) denotes the change of magnetic flux density inside the ferromagnetic ribbon under the electric field action, \( E \) is the electric field strength, \( d \) is the thickness of the piezoelectric plate.

The change of magnetic flux density \( \Delta B \) inside the amorphous ribbon due to the ME effect for each value of the external magnetic field \( H \) was determined as the difference between the values of \( B \) at \( U \neq 0 \) and \( U = 0 \):

\[
\Delta B(H,U) = B(H,U) - B(H,0) \tag{6}
\]

To estimate the value of the magnetic flux density \( B \) inside the ferromagnetic layer of the sample, the hysteresis loop for the amorphous ribbon was measured employing the vibrating sample magnetometer Lake Shore (VSM 7400) with the sensitivity up to \( 10^{-6} \) G·cm\(^3\).

3. Results and discussion

The hysteresis loop of the amorphous ribbon measured using the vibrating sample magnetometer is shown in figure 2. It was found that the amorphous tape has an extremely low coercive force and almost no hysteretic behavior in an external magnetic field. Given the ribbon size and its saturation...
magnetic moment, the magnitude of saturation magnetic flux density \( B_s \) of the amorphous ribbon was determined. It was found that \( B_s = 10500 \) G.

![Figure 2](image)

**Figure 2.** The hysteresis loop observed for the amorphous ribbon.

The magnetization curves of the amorphous ribbon observed without an electric field \( (U = 0 \) V) and with an external electric field of various magnitudes applied to the piezoelectric plate are shown in figure 3 and figure 4. For clarity, the initial parts of the magnetization curves are given.

![Figure 3](image)

**Figure 3.** The magnetization curves observed for the studied sample at presence of an electric voltage \( U \) of different magnitudes. a – the whole magnetization curves, b – initial parts of the curves. The external magnetic field \( H \) was applied parallel to the long side of the amorphous ribbon.

![Figure 4](image)

**Figure 4.** The magnetization curves observed for the studied sample at presence of an electric voltage \( U \) of different magnitudes. a – the whole magnetization curves, b – initial parts of the curves. The external magnetic field \( H \) was applied perpendicular to the long side of the amorphous ribbon.
Analysis of these data showed that the slope of the magnetization curves decreases/increases with increasing applied voltage $U$, when the magnetic field $H$ is applied parallel/perpendicular to the long side of the sample. These data indicate that in the cases described above the inverse magnetoelectric effect has opposite signs.

The dependences of the change of magnetic flux density $\Delta B$ in the ferromagnetic ribbon on the magnetic field $H$ at presence of an electric voltage $U$, calculated using the formula (6), are presented in figure 5.

The dependences of the change of magnetic flux density $\Delta B$ in the ribbon on the electric field strength $E$ are presented in figure 6. The value of $E$ was calculated as $U/d$, where $d$ is thickness of the PZT plate.

![Figure 5](image1.png)

**Figure 5.** The dependences of the change of magnetic flux density $\Delta B$ in the amorphous ribbon due to the application of an electric voltage $U$ to the piezoelectric PZT plate on the external magnetic field $H$ directed parallel (a) and perpendicular (b) to the long side of the sample.

![Figure 6](image2.png)

**Figure 6.** The dependences of the change of magnetic flux density $\Delta B$ in the ribbon on electric field strength $E$ at different magnitudes of the magnetic field $H$ in cases of $H$ applied parallel (a) and perpendicular (b) to the long side of the sample.

The dependences of the inverse magnetoelectric effect coefficient $\alpha$ calculated using the formula (5) on the magnetic field strength $H$ when the magnetic field is applied parallel and perpendicular to the long side of the sample are illustrated in figure 7.
Figure 7. The dependences of the inverse magnetoelectric effect coefficient $\alpha$ on the external magnetic field $H$ directed parallel (a) and perpendicular (b) to the long side of the sample.

Analysis of the obtained data showed that the maximum absolute values of the inverse ME effect coefficient $\alpha$ are equal to 0.1 G·cm/V and 0.049 G·cm/V at the orientations of magnetic field parallel and perpendicular to the long side of the sample, respectively.

4. Conclusion

- For the first time the study of the features of the inverse magnetoelectric effect in a two-layered composite structure consisting of an amorphous FeCuNbSiB ribbon and a Pb(Zr,Ti)O$_3$ plate has been carried out by using the magneto-optical method.
- It has been found that the electric voltage $U$ applied to the piezoelectric plate affects the magnetic properties of the amorphous ribbon.
- It was established that the change of the orientation of the external magnetic field $H$ from the direction parallel to the long side of the sample to the perpendicular one is accompanied by a change in both the magnitude and the sign of the inverse ME effect.
- It has been found that the absolute value of the coefficient $\alpha$ of inverse ME effect varies from 0 to 0.1 G·cm/V and from 0 to 0.049 G·cm/V when the external magnetic field $H$ is directed parallel and perpendicular to the long side of the sample, respectively.
- The difference in the signs of the inverse ME effect in the cases of magnetization reversal of the sample by a magnetic field applied parallel and perpendicular to its long side can be explained by the anisotropy of the amorphous layer.

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