Magnetoelectric effect in self-bias gradient structure
CoFe$_2$O$_4$/Ni/BaTiO$_3$ with 0-3 connectivity

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Abstract. The paper is devoted to the study of mechanism of internal magnetic fields occurrence in the magnetostrictive-piezoelectric gradient structure CoFe$_2$O$_4$/Ni/BaTiO$_3$. The insertions of CoFe$_2$O$_4$ and Ni in this gradient structure in the matrix BaTiO$_3$ is located. The connectivity type of this composite is 0-3 and a modified model of the cell in the form of a parallelepiped is used. This parallelepiped consists of two cubic cells of BaTiO$_3$ matrix. One cubic cell contains of CoFe$_2$O$_4$ insertion and another cell contains of Ni one. The corresponding magnetic circuit by using the magnetostatics equations was considered. Internal magnetic fields which arise in sublattices of CoFe$_2$O$_4$ and Ni, taking into account the known parameters of their magnetization curves were found and equal of 860 Oe and of 50 Oe for CoFe$_2$O$_4$ and Ni, respectively. Further, the pseudo-piezomagnetic constants that correspond to the resulting internal magnetic fields were found by using of the existing dependencies of the pseudo-piezomagnetic coefficients of CoFe$_2$O$_4$ and Ni on the bias magnetic field. The effective parameters of the structure in the form of a parallelepiped was found.

1. Introduction
The presence of dc magnetic field is required in the practical application of the ME effect. The patent RU2363074C1 shows that in a two-phase magnetostrictive-piezoelectric material with gradients of magnetization and polarization, internal magnetic and electric fields can be created [1]. The use of such gradient materials will make it possible to exclude from the design of magnetoelectric devices the source of an external magnetic field. Hence, the developed devices will have the weight and dimensions are smaller than counterparts.

Earlier, cubic cell model was used at considering the composite material of 0-3 connectivity [2]. A modified model of the cell in the form of a parallelepiped consisting of two cubic cells of the BTO matrix, one of which contains of the CFO cubic inclusion and the other of the Ni cubic inclusion was used in this paper, figure 1.

The appearance of the internal magnetic field in gradient materials BaCo$_x$Ti$_y$Fe$_{12-2x}$O$_{19}$ and NZFO as theoretically and experimentally was investigated [1,3]. The gradient of magnetization was related with
the creation with the help of special technology almost linear concentration gradient of Zn and Co, Ti, Fe, respectively.

ME effect in the bending mode for two-layer composite consisting of a functionally graded NZFO and PZT in the low frequency region and in the electromechanical resonance was investigated [4]. Also important conclusions about the influence of the positivity or negativity of the gradient on the value of ME coefficient was made.

The layered magnetostrictive-piezoelectric composites with gradient material NZFO were investigated [5]. Stepped gradient pseudo-piezomagnetic properties were created through stepwise changes in the concentration of zinc in NZFO. It was studied the ME effect at zero bias DC magnetic field through the use of such a gradient magnetostrictive material. Also, it was obtained a higher value of the maximum of the ME voltage coefficient dependence on the bias field with compared to the respective no-gradient magnetostrictive materials.

The layered magnetostrictive-piezoelectric composites, where as magnetostrictive phase was used stepped gradient material formed from layers of Ni and Metglas theoretically and experimentally was investigated [6,7]. ME effect for bending mode, in low-frequency region and EMR region was discussed. Nonzero ME voltage coefficient without applying an external constant magnetic field was found.

ME effect at low frequencies for bending mode for three-layer composite PZT/Ni0.7Zn0.3Fe2O4/Ni was theoretically and experimentally investigated [8]. Hysteretic dependence of the magnetoelectric voltage coefficient on the external magnetic field was obtained.

The aim of this work is the study of mechanism of bias magnetic fields occurrence in the magnetostrictive-piezoelectric gradient structure CoFe2O4/Ni/BaTiO3.

2. Theoretical modeling

Figure 1. Model of the cell appears in the form of a parallelepiped, which consists of two cubic cells of the BTO matrix, one of which contains of the CFO cubic insertion (insertion 1) and the other contains of Ni cubic insertion (insertion 5).
It is supposed that a sample of the composite was magnetized along the axis 1 a constant magnetic field up to saturation of Ni and of cobalt ferrite, and then the magnetizing field was reduced slowly to zero, as it shown in figure 1. The residual magnetism will be present as the result of it influence in Ni and CFO. In this case, an internal magnetic field directed at the original magnetizing field in the CFO will be present, in addition the internal magnetic field in Ni will be the opposite directed, because the cobalt ferrite is much more hard magnetic material than nickel.

The appropriate models of the magnetic circuit will calculate using the equations of magnetostatics based on the known parameters of the curves of demagnetization of the CFO and Ni. The internal magnetic fields $\tilde{H}_1$, $\tilde{H}_2$ which appearing in them were found for the case $a_1 \geq a_2$:

$$\tilde{H}_1 = \frac{P_1}{Q_1}$$

$$P_1 = H_1\left[B_{1i}\left((a_i^2 - 1)(\mu_2 - \mu)\alpha_{1z}^2 + (a_i^2 - a_1 + 2)\mu_2\alpha_{1z}^2\right) + B_{1z}(a_i^2 - 1)\alpha_{1z}^2 + B_{1i}(a_i^2 - 1)\alpha_{1z}^2\mu\right]$$

$$Q_1 = B_{1i}\left((a_i^2 - 1)(\mu_2 - \mu)\alpha_{1z}^2 + (a_i^2 - a_1 + 2)\mu_2\alpha_{1z}^2\right) + B_{1z}(a_i^2 - 1)\alpha_{1z}^2 + B_{1i}(a_i^2 - 1)\alpha_{1z}^2\mu$$

$$\tilde{H}_2 = \frac{P_2}{Q_2}$$

$$P_2 = \mu_0 H_e B_{1i}\alpha_{1z}^2(1 - a_i^2) + B_{1z}(\mu_0 H_e - B_{1i})\alpha_{1z}^2(a_i^2 - 1) - B_{1z}\alpha_{1z}^2(a_i^2 - a_2 + 2)$$

for the case $a_1 < a_2$:

$$\tilde{H}_1 = \frac{P_1}{Q_1}$$

$$P_1 = H_1\left[B_{1i}\left(\alpha_{1z}^2 - \mu_2\alpha_{1z}^2(a_i^2 - 1) + (a_i^2 - a_1 + 2)\mu_2\alpha_{1z}^2\right) + B_{1z}(a_i^2 - 1)\alpha_{1z}^2\mu\right]$$

$$Q_1 = B_{1i}\left((a_i^2 - 1)(\mu_2 - \mu)\alpha_{1z}^2 + (a_i^2 - a_1 + 2)\mu_2\alpha_{1z}^2\right) + B_{1z}(a_i^2 - 1)\alpha_{1z}^2 + B_{1i}(a_i^2 - 1)\alpha_{1z}^2\mu$$

$$\tilde{H}_2 = \frac{P_2}{Q_2}$$

$$P_2 = \mu_0^p \mu H_e B_{1i}\alpha_{1z}^2(1 - a_i^2) + \mu_0 \mu H_e B_{1z}\left[a_i^2(a_i^2 - 1) + (a_i^2 - a_2^2)(a_i^2 - a_2 + 2)\right] + B_{1z}\alpha_{1z}^2(a_i^2 - a_2^2 + a_2 - 2)$$

where $a_i$, $a_2$ is the relative edge length of CFO and Ni cubic inclusions, $\mu_0$ is the magnetic permeability, $B_{1i}$, $H_i$ is residual magnetic induction and coercive force of CFO, $B_{1z}$, $\mu_2$ is the residual magnetic induction and the relative reversible magnetic permeability Ni, $\mu^p$ is the relative magnetic permeability of BTO.

The volume fraction of the CFO and Ni is equal to 0.15, and for BTO is 0.7. Then $a_1 = a_2 = 0.67$, and internal fields calculated by the formula (1): $\tilde{H}_1 = 68562 \text{ A/m}$, $\tilde{H}_2 = 4257 \text{ A/m}$.

Further, the pseudo-piezomagnetic coefficients that correspond to the resulting internal magnetic fields were found using the dependence of the magnetostriction coefficients of CFO and Ni on the biasing DC magnetic field, which was given in [9] and [10], respectively.
If an alternating magnetic field directed along the axis 1, the electric field in the direction of the axis 3 occurs in this case. The elastostatic, magnetostatics and electrostatics equations allow to find effective parameters of magnetoelectric composites using the modified model.

Longitudinal components of strain tensor the piezoelectric phase are expressed as

\[
\begin{align*}
S_1 &= p_{s11}T_1 + p_{s12}T_2 + p_{s13}T_3 + d_{31}E_3 \\
S_2 &= p_{s12}T_1 + p_{s11}T_2 + p_{s13}T_3 + d_{31}E_3 \\
S_3 &= p_{s13}T_1 + p_{s11}T_2 + p_{s12}T_3 + d_{31}E_3 \\
\end{align*}
\]

\( n = 2, 3, 4, 6, 7, 8 \) \hspace{1cm} (3)

The third components of the electric displacement vector and the first vector components of the magnetic induction piezoelectric phase are expressed as

\[
\begin{align*}
D_3 &= d_{31}(T_1 + T_2) + d_{33}E_3 + p_{e33}\epsilon_0 E_3 \\
B_3 &= \mu_0 \mu H_1 \\
\end{align*}
\]

\( n = 1 \) \hspace{1cm} (4)

The longitudinal components of the strain tensor magnetostrictive phases are expressed as

\[
\begin{align*}
S_1 &= m_{s11}T_1 + m_{s12}T_2 + m_{s13}T_3 + q_{11}H_1 \\
S_2 &= m_{s12}T_1 + m_{s11}T_2 + m_{s13}T_3 + q_{11}H_1 \\
S_3 &= m_{s13}T_1 + m_{s12}T_2 + m_{s13}T_3 + q_{11}H_1 \\
\end{align*}
\]

\( n = 2, 3, 4, 5, 6, 7, 8 \) \hspace{1cm} (5)

The third components of the electric displacement vectors and the first components of magnetic induction vectors of the magnetostrictive phases are expressed as

\[
\begin{align*}
D_3 &= m_{e33}\epsilon_0 E_3 \\
B_3 &= q_{12}(T_2 + T_3) + q_{11}T_1 + \mu_1\mu_0 H_1 \\
D_3 &= m_{e33}\epsilon_0 E_3 \\
B_3 &= q_{12}(T_2 + T_3) + q_{11}T_1 + \mu_1\mu_0 H_1 \\
\end{align*}
\]

\( n = 1 \) \hspace{1cm} (6)

Taking into account the conditions of compatibility for deformations and boundary conditions for the field vectors one gets a system of 40 equations with 40 unknowns components obtained by substituting the expressions for the longitudinal components of the tensor of deformations, the third component of the vectors of the electric displacement and the first component of the vectors of magnetic induction in the rest of the equation. All unknown components are found by numerical-analytical method of expressing through \( T_1, T_2, T_3, E_3, H_1 \).

As the first component of effective deformation tensor is equal to

\[
S_1 = \frac{4S_1 + 8S_3}{2}
\]

will be obtained the expression

\[
S_1 = s_{11}T_1 + s_{12}T_2 + s_{13}T_3 + d_{31}E_3 + q_{11}H_1
\]

\( n = 1 \) \hspace{1cm} (8)
The effective parameters such as the compliance coefficient, piezoelectric coefficient and pseudo-piezomagnetic coefficient are 
\[ s_{11} = 5.8 \times 10^{-12} \text{ m}^2 \text{N}^{-1}, \quad d_{11} = -3.88 \times 10^{-11} \text{ m} \text{V}^{-1}, \quad q_{11} = -2.16 \times 10^{-10} \text{ m} \text{A}^{-1}. \]

The third component of effective electric displacement vector is 
\[ D_3 = \left[ a_1^2 \cdot D_3 + (1-a_1)^4 D_3 + a_1 (1-a_1)^3 D_3 + a_2^2 \cdot D_3 + (1-a_2)^8 D_3 + a_2 (1-a_2)^7 D_3 \right] / 2. \] (9)

Unknown components that have previously been found, we substitute in the formula (9) and thus we find 
\[ D_3 = -\tilde{a}_{31} T_3 + \tilde{a}_{32} T_2 + \tilde{a}_{33} T_1 + \varepsilon_{33} \varepsilon_0 E_3 + m_{33} H_1. \] (10)

The effective relative permittivity and effective magnetoelectric susceptibility are 
\[ \varepsilon_{33} = 232.8, \quad m_{31} = -1.61 \times 10^{-9} \text{ S} \text{m}^{-1}. \]

The longitudinal oscillations in the magnetoelectric sample investigated above structure by the action of an alternating magnetic field directed along the axis 1 will be discussed below. Composite length is equal \( l=10^{-2} \text{ m} \) and directed along the axis 1, the width and thickness much less than the length. The metal electrodes deposited on the surface of the material. AC voltage is generated on the metal electrodes due to the ME effect. The normal line to the electrodes plane is directed along the axis 3.

In this case, we assume that \( T_2=T_3=0 \), then 
\[ S_1 = s_{11} T_1 + \tilde{d}_{11} E_3 + q_{11} h_1, \] (11)
whence 
\[ T_1 = \frac{1}{s_{11}} \left( S_1 - \tilde{d}_{11} E_3 - \frac{q_{11}}{s_{11}} h_1 \right). \] (12)

The equation of longitudinal oscillations of the membrane 
\[ \rho \frac{\partial^2 u_1}{\partial t^2} = \frac{\partial T_1}{\partial x_1}, \] (13)
where \( \rho = \rho' + \varepsilon_0 \varepsilon_m' + m_1 \varepsilon_m + m_2 \varepsilon_m \rho = 0.7 \cdot 6017 + 0.15 \cdot 5200 + 0.15 \cdot 8902 = 6327.2 \text{ kg} / \text{ m}^3 \) is the effective density of the composite.

The first component of strain tensor 
\[ S_1 = \frac{\partial u_1}{\partial x_1}, \] (14)

The solution to the equation of longitudinal oscillations 
\[ u_1 = A \cos \left( k x_1 \right) + B \sin \left( k x_1 \right), \]
\[ k = \omega \sqrt{\rho s_{11}} \] (15)

From the boundary conditions for the free sample 
\[ T_1 \left( -\frac{l}{2} \right) = 0 \]
\[ T_1 \left( \frac{l}{2} \right) = 0 \] (16)
follows
\[ A = 0 \]
\[ B = \frac{\tilde{d}_{31} E_i + q_{11} h_k}{k \cos \left( \frac{kl}{2} \right)}. \]  \hspace{1cm} (17)

Using the open circuit condition
\[ \int \frac{j}{3} D_i dx_i = 0, \]  \hspace{1cm} (18)

ME voltage coefficient is
\[ \alpha_E = \frac{E_i}{h_k} = \frac{\tilde{d}_{31} q_{11} (1 - w) - m_{31} s_{11}}{\varepsilon_{33} \varepsilon_0 s_{11} + \tilde{d}_{31}^2 (w - 1)}. \]  \hspace{1cm} (19)

The dependence of ME voltage coefficient on the frequency of the alternating magnetic field is shown in figure 2. To account for losses in the calculation, the following formula is used
\[ \omega = 2\pi \left( 1 + \frac{1}{2Q} \right) f, \]  \hspace{1cm} (20)

where the quality factor of the resonance \( Q = 100. \)

\[ \text{Figure 2.} \quad \text{The dependence of ME voltage coefficient on the alternating magnetic field frequency. The dimension of ME structure is } 10 \times 5 \times 0.5 \text{ mm. The effective relative permittivity } \varepsilon_{33} = 232.8, \text{ and effective magnetoelectric susceptibility } m_{31} = 1.61 \times 10^{-9} \text{ s/m. The compliance coefficient, piezoelectric coefficient and pseudo-piezomagnetic coefficient are } s_{11} = 5.8 \times 10^{-12} \text{ m}^2/\text{N}, \tilde{d}_{31} = -3.88 \times 10^{-11} \text{ m/V}, q_{11} = 2.16 \times 10^{-10} \text{ m/A. The effective density of the composite is } \rho = 6327.2 \text{ kg/m}^3. \]

3. Conclusion
In this paper effective parameters of magnetostrictive-piezoelectric composite with the connectivity of 0-3 was found using a modified model of the cell in the form of a parallelepiped based on the equations of elastostatic, magnetostatics and electrostatics. The longitudinal mode of magnetoelectric effect taking into
account of gradient structure on the basis of the obtained effective parameters was considered. The use of three-component magnetostrictive-piezoelectric composite gradient structures, similar studied, allows to reduce mass-dimensional characteristics of the ME devices. This is possible due to the fact that there is no need to use a dc magnetic field for ME composite in ME devices.

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