Magnetoelectric effect in bismuth - neodymium ferrite - garnet films

A N Masyugin, S S Aplesnin, Y Y Loginov and O N Bandurina

Reshetnev Siberian State University of Science and Technology, 31 Krasnoyarsky Rabochy Av., Krasnoyarsk, 660037, Russian Federation

E-mail: apl@iph.krasn.ru

Abstract. The prehistory of the dielectric properties of bismuth – neodymium ferrite – garnet films on a Gd₃Ga₅O₁₂ (111) substrate cooled in an electric and magnetic field were established. The dynamic characteristics of the electric polarization are studied at large times when the electric field is turned on and off. The influence of a magnetic field on the residual polarization was found. The dependence of the electric polarization on the electric field in a magnetic field is determined. Experimental data are explained in the Debye model.

1. Introduction

Multiferroics are characterized by a strong interrelation of the magnetic and electrical subsystems [1-6], which in single-phase ferromagnetic materials is realized due to the spin-lattice and electron-lattice interactions. Studies of single-phase materials are important for a deeper understanding of electromagnetic phenomena in solids and are of interest for the creation of a new generation of solid-state electronics devices.

Yttrium ferrite garnet has cubic symmetry with an inversion center [7] and at low temperatures (below 130 K) a structural transition with triclinic lattice distortion is detected [8, 9]. The electric polarization may be caused by deformation of the structure, which leads to breaking of the center inversion as a result of epitaxial film stress on the substrate or cationic substitution on dodecahedral nodes; surface electron states, magnetic domain structure. In films (BiLu)₃(FeGa)₅O₁₂ thickness of 10 μm, grown by liquid phase epitaxy on a substrate Gd₃Ga₅O₁₂ with a substrate orientation (210) is found the electric polarization of the domain walls, which can be switched by the external magnetic field [10]. Electric polarization is absent on films with substrate (111) orientation. These effects can be explained due to inhomogeneous magnetoelectric interaction and changes the magnetic anisotropy of the electric field [11]. The last factor can be neglected using external magnetic field an order of magnitude greater than the saturation field.

2 Magnetostriiction

The purpose of the research is to establish the mechanism of the magnetoelectric interaction in a strong magnetic field in bismuth - neodymium ferrites - garnets films deposited on a garnet substrate. The Nd₀.₅Bi₂.₅Fe₅O₁₂ film (450 nm) was studied on a single-crystal substrate Gd₃Ga₅O₁₂ (GGG) in the (111) direction. The films were obtained by the method of epitaxial deposition [7].

The films have a maximum of magnetostriction and electrostriction at a temperature of 200 K. Expansion goes into compression of the film in a magnetic field at cooling. The experimental data are explained in terms of model of ferroelectric domains and the magnetoelectric interaction. The
maximum of magnetostriction is associated with the formation of a dipole glass and domain pinning [12].

3. Capacitance
Cooling the films in a magnetic field \( H = 12 \text{ kOe} \) and in an electric field \( E = 400 \text{ V/cm} \) from room temperature to \( T = 80 \text{ K} \) leads to a decrease in capacitance within one percent. After cooling and further heating to \( T = 300 \text{ K} \) film capacity decreases by (3-4) % (figure 1).

![Figure 1. Frequency dependence of capacitance of Nd\(_{1}\)Bi\(_{2}\)Fe\(_{5}\)O\(_{12}\) (450nm) / Nd\(_{2}\)Bi\(_{1}\)Fe\(_{4}\)Ga\(_{1}\)O\(_{12}\) (90nm) film on glass (a): 1 - initial state at \( T = 320 \text{ K} \), 2 - after cooling in a field \( E = 400 \text{ V} / \text{cm} \) and heated to \( T = 320 \text{ K} \), 3 - after cooling in a field \( E = 400 \text{ V} / \text{cm} \) at \( T = 80 \text{ K} \), 4 - after cooling in a magnetic field \( H = 12 \text{ kOe} \) at \( T = 80 \text{ K} \); and dielectric loss tangent (b): 1 - initial state at \( T = 320 \text{ K} \), 2 - after cooling in a magnetic field \( H = 12 \text{ kOe} \) at \( T = 80 \text{ K} \). Capacitance and dielectric losses calculated in the Debye model (1) (solid lines).](image)

The dependence of the dielectric constant on frequency at low temperatures is satisfactorily described in the Debye model with a relaxation frequency of 5 MHz. The dielectric relaxation in the low frequency region \( \omega <300 \text{ Hz} \), is caused by electron jumps in the film defects \( \text{Im}(\varepsilon) = \sigma/\omega \) [13–15]. At temperatures above room temperature, the diffusion contribution of the domain boundaries is added to the dielectric susceptibility in the form:

\[
\text{Re}(\varepsilon) = \varepsilon_0 + \chi_0/(1+(\omega \tau)^2) - \text{vlg}(\omega), \quad \text{Im}(\varepsilon) = \chi_0 \omega \tau/(1+(\omega \tau)^2) + \sigma/\omega
\]  

(1)

where \( \tau \) is the relaxation time, \( \sigma \) is the conductivity, \( \nu \) is the relaxation rate, \( \chi_0 \) is the dielectric susceptibility in a constant field.

4. Remanent polarization
The dynamic characteristics of the electric polarization at large times is determined by measuring the charge when a rectangular voltage pulse is switched on with amplitude \( E = 400 \text{ V} / \text{cm} \) with frequency \( \omega = 0.01, 0.003 \text{ Hz} \) (figure 2). The residual polarization found after turning off the field disappears at a temperature \( T = 300 \text{ K} \) (figure 3). The electric polarization depends on the direction of the magnetic field applied perpendicularly and parallel to the film. Coefficient magnetoelectric interaction is the second-rank tensor.
Figure 2. Dependences of the charge induced by periodic pulses of the electric field 400 V / cm from time, at 360 K (a), 300 K (b), 80 K (c), 200 K (d): 1, 6 - without a magnetic field; 2, 4, 5 - in a magnetic field of 12 kOe perpendicular to the film; 3 - in a magnetic field of 12 kOe along the film.

Figure 3. Residual charge after the impulse of the electric field of 400 V/cm: 1 - in a magnetic field of 12 kOe along the film; 2, 3 - in a magnetic field of 12 kOe perpendicular to the film; 4 - without magnetic field.
The electric polarization was found from the relation $P = \int j \, dt$ when measuring the current in an external quasi-periodic field with a frequency $w = 0.01 \, \text{Hz}$ with different orientations of the magnetic field [16-18]. Electric polarization versus the external field is shown in figure 4. The shift of polarization is caused by negatively charged defects in the vicinity of the interface. Hysteresis is observed in the region of large electric fields and is associated with the formation of an electric field at the interface of the substrate–film. When heated, the relative polarization bias is halved from $T = 80 \, \text{K}$ to $T = 300 \, \text{K}$. In the general case, the polarization is determined by the field of charged defects, the interface field ($P_0$) $P = P_0 + \chi \alpha E$. The dielectric susceptibility has a maximum at $T = 280 \, \text{K}$.

**Figure 4.** Electric polarization from an external electric field at 80K (a), 160K (b), 120K (c), 280K: 1 - without a magnetic field, 2 - in a magnetic field directed along the film, 3 - in a magnetic field directed perpendicular to the film.

5. Conclusion
A decrease in the capacity of the film after cooling from room temperature to $T = 80 \, \text{K}$ in an electric field is found. The frequency dependence of the film capacity is described in the Debye model, the relaxation time is found. The critical temperature of the disappearance of the residual polarization and the anisotropy of the electric polarization in a magnetic field are found. The temperature of the maximum dielectric constant is established.

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