Magnetoresistance of rare-earth-substituted garnet ferrite films

A N Masyugin¹, S S Aplesnin¹,², Y Y Loginov¹ and A I Stognij³

¹Reshetnev Siberian State University of Science and Technology, 31 Krasnoyarskii rabochii prospekt, Krasnoyarsk, 660037, Russia
²Kirensky Institute of Physics SB RAS, 50/38 Akademgorodok St., Krasnoyarsk, 660036, Russia
³SSPA “Scientific-Practical Materials Research Centre of NAS of Belarus”, 19 P. Brovki St., Minsk, 220072, Belarus

E-mail: aplesnin@sibsau.ru

Abstract. Thin epitaxial films of garnet ferrite were studied using impedance spectroscopy. The influence of the magnetic field on the transport characteristics was investigated on the basis of the current-voltage characteristic characteristics (I-V) measured without a field and in a magnetic field H = 12 kOe. An increase in resistance in bismuth ferrite garnet was detected in a magnetic field at a temperature above room temperature.

1. Introduction
The colossal magnetoresistance in inhomogeneous semiconductors with phase separation [1-4], the coexistence of crystallographic phases [5, 6] and polarons is being actively studied due to the possible use of materials in spintronics [7]. In recent years, interest has increased in the study of electrical properties in a magnetic field for paramagnetic materials, which can be used to create random access memory [8-10]. The exchange coupling between localized and delocalized electrons leads to anomalies in the temperature dependence of electrical resistance and formation of magnetoresistance, which is pronounced especially strong in the compounds with the 4f rare-earth elements [11]. In semiconductors, the spin–orbit interaction at the interface can attain high values and, according to the theoretical calculations [12–13], leads to a significant increase in the magnetoresistance at the heating in the paramagnetic state. In magnetic films, a large potential gradient at the interface can induce a spin-orbit interaction.

A linear magnetoelectric effect [14–15] and the coexistence of current channels with ferroelectric regions were found in bismuth ferrite garnet films, which makes it possible to control the current with a magnetic and electric field.

Purpose of the study is to establish the influence of a magnetic field and its mechanism on the transport characteristics of garnet ferrite in results of the replacement of yttrium with bismuth and cerium ions.

2. Experimental results and discussion
Iron garnets are good insulators with high Curie temperature. In the deposition of films with a thickness of 100-200 nm, the film – substrate interface plays a significant role, where the formation of charged defects and the polycrystalline structure of the films are possible. In this case, the formation of magnetic polarons at the interface is possible. Spin-phonon interaction can change the magnetic structure [16-19].
Garnet ferrites with cerium exhibit charge fluctuations of $\text{Ce}^{+3} - \text{Fe}^{+3} = \text{Ce}^{+4} - \text{Fe}^{+2}$ with a gap of 1.3 eV. At the interface and at the polycrystalline boundaries, fluctuations in the crystal field will lead to a decrease in the gap. Oxygen vacancies can also lead to ferrous iron with the formation of electron-hole states. Perhaps two transport channels along the anionic and cationic systems. This can be established by the method of impedance and IR spectroscopy, from the current-voltage characteristic and electrical resistance in magnetic fields up to 12 kOe.

We studied epitaxial films $\text{Nd}_1\text{Bi}_2\text{Fe}_5\text{O}_{12} (450\text{nm})/\text{Nd}_2\text{Bi}_1\text{Fe}_4\text{Ga}_1\text{O}_{12} (90\text{nm})$ on a glass substrate and $\text{Nd}_0.5\text{Bi}_2.5\text{Fe}_5\text{O}_{12} (450\text{nm})$ on a GGG single crystal substrate grown in the (111) crystallographic direction. The films have a ferrimagnetic structure with a Curie temperature $T_c = 450 \text{K}$. In the vicinity of $T_c$, the conductivity and impedance exhibit a kink characteristic of the polaron type of current carriers. This indicates the exchange interaction of the spins of mobile electrons with localized spins.

The influence of the magnetic field, the transport characteristics were studied on the basis of the I-V measured without a field and in a magnetic field $H = 12 \text{kOe}$ applied perpendicular to the film and along the film. Figure 1 shows the I-V of films on a glass and GGG substrate. The I-V hysteresis is associated with the presence of regions with electric polarization and is shifted along the axis of the electric field by 20–30 V for the film on GGG. When heated above 280 K, the hysteresis width decreases several times and the displacement field disappears. In a magnetic field, the resistance decreases below room temperature and increases above 280 K. The magnetoresistance $\Delta R = (R(H)-R(0))/R(0)$ exhibits anisotropy relative to direction of the magnetic field to the normal of the film. This is due to the orientation of the magnetization of the film, which reveals a spin-reorientation transition at the room temperature. The I-V of the film on the glass (figure 1 b) has a rectangular shape and the current does not depend on voltage up to 320 K. In this film, electric polarization is induced by an external electric field and a pyrocurrent $I_p = dP/dt$ arises, the value of which exceeds the external current. At the heating the resistance of the film decreases and the external current exceeds the $I_p$ current. In a magnetic field, the hysteresis loop practically does not change. An increase in the resistance in a magnetic field $(R(H)-R(0))/R(0) \sim 0.01$ was found on alternating current up to 300 K. The dependence of $\Delta R (H)$ on the field is satisfactorily described by the power-law function $\Delta R (H) \sim H^2$ (figure 2a).

![Figure 1.](image_url)

**Figure 1.** (a) - I-V of films on a GGG substrate at 160K $H = 0$ (1), $H = 12\text{kOe}$ perpendicular to the film (2), along the film (3), at 280K $H = 0$ (4), $H = 12\text{kOe}$ perpendicular to the film (5) along the film (6). (b) I-V of films on a glass substrate at 380K $H = 0$ (1), 120K $H = 0$ (2), $H = 12\text{kOe}$ directed at angles 0 ° (3), 90 ° (4), 180 ° (5), 270 ° (6), 360 ° (7) relative to the normal to the film.

The resistance in the frequency range 100-500 Hz increases in the magnetic field by 5-8% at $T = 400 \text{K}$ and the magnetoresistance $\Delta R = (R (H) - R (0)) / R (0)$ is 2-3% at $T = 350 \text{K}$ and 450 K, and disappears at room temperature. Magnetoimpedance does not exceed 0.3%.
At these temperatures, the activation energy decreases from 0.27 eV to 0.18 eV. DC resistance increases smoothly with heating and reaches a maximum at \( T = 300 \text{ K} \) with a further decrease. In a magnetic field, the change in current does not exceed one percent.

Domain and crystal boundaries can be determined from impedance. In the analysis of the experimental results, the equivalent circuit approximation was used. The impedance hodograph is described by one semicircle, corresponding to the R, C circuit, where the capacitance is due to crystalline boundaries (figure 2b). In the high frequency region, a linear contribution to the impedance is added. This element models the impedance of a linear diffusion process occurring in a homogeneous layer with a finite thickness. The impedance in this process depends on the resistance, particle diffusion coefficient, and the thickness of the Nernst diffusion layer. At high frequencies, the experimental data are well approximated by a linear Warburg impedance diagram [20, 21]:

\[
Z_W(j \omega) = k_f (j \omega D_0)^{-1/2} = k_f (\omega D_0)^{-1/2} (1 - j)
\]  

where \( k \) is the coefficient, \( D \) is diffusion. The diffusion contribution to the impedance is observed at room temperature and above. Diffusion decreases with increasing frequency. The impedance hodograph shown in figure 2b is described in the model of equivalent circuits by one semicircle. The relaxation time \( \tau = RC \) (R-resistance, C-capacitance) drops sharply when heated from \( \tau = 10 \text{ s} \) at \( T = 300 \text{ K} \) to \( \tau = 0.1 \text{ s} \) at \( T = 340 \text{ K} \). An increase in the relaxation time with decreasing temperature indicates the formation of a charge glass state at room temperature. Broken Bi-O bonds on the interface form a dipole moment and regions with local electric polarization. This polarization is shielded by free charge carriers formed by impurities and structural defects.

\[ Z_W(j \omega) = k_f (j \omega D_0)^{-1/2} = k_f (\omega D_0)^{-1/2} (1 - j) \]  

Figure 2. (a) – Dependence of the active resistance of a film on a glass substrate on the magnitude of the magnetic field at temperatures of 160K (1), 240K (2), 380K (3), (b) – impedance hodographs at temperatures of 340K \( H = 0 \) (1), \( H = 12 \text{ kOe} \) (2), 300K \( H = 0 \) (3).

The participation of electrons of a alone bismuth pair in hybridization is confirmed by the presence of a magnetic field on Bi nuclei. A change in the length of the cation-anion bond leads to a displacement of oxygen and to a change in the exchange interaction between oxygen ions and to uniaxial anisotropy. An external magnetic field can switch the direction of magnetic anisotropy and the electric polarization of the Bi-O bond. In a spin-reorientation transition, the mobility of current carriers in an external magnetic field changes as a result of s-d interaction. This mechanism explains the change in the sign of magnetoresistance in temperature. The scattering of current carriers by magnetic inhomogeneities, the magnetic moments of which can be changed by a magnetic field, was previously considered on sulfides [22-24].

CeFeO$_2$ films were obtained on a GGG single crystal substrate grown in the (111) crystallographic direction by thick 200 nm. Two transitions were found from the optical absorption
spectra associated with the transition of electrons between the $^6\text{A}_1 \rightarrow ^4\text{T}_2$ levels in the tetrahedron at 2.3 eV and with the charge transfer $2p \rightarrow 3d$ through oxygen $\text{Fe}_{\text{tet}} – \text{O} – \text{Fe}_{\text{oct}}$ at 2.6 eV. «Tails» in the range 1.7–2.3 eV are due to fluctuations in the crystal field in tetrahedra at the interface and grain boundaries. The energy of charged defects are determined from the IR spectra. The IR spectrum contains two absorption lines in the frequency range $\omega_1 \sim 5520 \text{ cm}^{-1}$ and $\omega_2 = 6800 \text{ cm}^{-1}$. One line at $\omega_1$ disappears at 300 K, the other at 400 K.

Figure 3. Real (a) and imaginary (b) parts of the Ce3Fe5O12 impedance at frequencies $\omega = 1 \text{ kHz}$, 10 kHz versus temperature.

The disappearance temperature of the second line coincides with the maximum temperature of 400 K of the imaginary part of the impedance (figure 3a), which is independent of frequency. The maximum dielectric constant is caused by charge fluctuations, which also lead to a local maximum of resistance on alternating current (figure 3a). The resistance in the frequency range 100-500 Hz increases in the magnetic field by 5-8% at $T = 400 \text{ K}$ and the magnetoresistance $\Delta R=(R(H)-R(0))/R(0)$ is 2-3% at $T = 350 \text{ K}$ and 450 K, and disappears at room temperature.

3. Conclusion
In bismuth ferrite, an increase in resistance in a magnetic field was detected at temperatures above room temperature. The maximum magnetoresistance of the order of several percent is achieved at a temperature of 450 K. The impedance hodograph is described by a single circuit and at high temperatures the diffusion contribution is added. Magnetic impedance not exceeding one percent was found.

References
[1] Zhu Y, Du K, Niu J, Lin L, W Wei W, Liu H, Lin H, Zhang K, Yang T, Kou Y, Shao J, Gao X, Xu X, Wu X, Dong S, Yin L and Shen J 2016 Nat. Commun 7 1250
[2] Tokura Y 2006 Reports Prog. Phys. 69 797
[3] Aplesnin S S, Ryabinkina L I, Romanova O B, Sokolov V V, Pichugin A Y, Galyas A I, Demidenko O F, Makovetski G I and Yanushkevich K I 2009 Phys. Solid State 51(4) 698
[4] Aplesnin S, Romanova O, Har’kov A, Balaev D, Gorev M, Vorotinov A, Sokolov V, and Pichugin A 2012 Phys. Status Solidi B. 249 812
[5] Bebenin N G, Zainullina R I and Ustinov V V 2018 UFN 188 801
[6] Udod L V, Aplesnin S S, Sitnikov M N and Molokeev M S 2014 Phys. Solid State 56(7) 1315
[7] Fert A 2008 UFN 178 1336
[8] Aplesnin S S and Sitnikov M N 2014 JETP Letters 100(2) 95-101
[9] Aplesnin S S, Romanova O B and Yanushkevich K I 2015 Phys. Status Solidi B 252(8) 1792-1798
[10] Aplesnin S S, Ryabinkina L I, Romanova O B, Velikanov D A, Balaev A D, Balaev D A, Yanushkevich K I, Galyas A I, Demidenko O F and Bandurina O N 2008 J. Exp. Theor. Phys. 106(4) 765-772

[11] Olson Reichhardt C J, Reichhardt C and Bishop A R 2004 Phys. Rev. Lett. 92 4

[12] He H T, Liu H C, Li B K, Guo X, Xu Z J, Xie M H and Wang J N 2013 Appl. Phys. Lett. 103 031606.

[13] Sánchez J C R, Vila L, Desfonds G, Gambarelli S, Attané J P, De Teresa J M, Magén C and Fert A 2013 Nat. Commun. 4 2944

[14] Aplesnin S S, Masyugin A N, Sitnicov M N, Rybina U I and Ishibashi T 2018 J. Magn. Magn. Mater. 464 44

[15] Popova E, Shengelaya A, Daraselia D, Japaridze D, Cherifi-Hertel S, Bocher L, Gloter A, Stéphan O, Dumont Y and Keller N 2017 Appl. Phys. Lett. 110 142404

[16] Aplesnin S S 1996 Phys. Solid State 38 1031

[17] Aplesnin S S 1998 J. Phys. Condens. Matter 10 10061

[18] Aplesnin S 2000 Phys. Rev. B. 61 6780

[19] Aplesnin S S 2003 J. Exp. Theor. Phys. 97 969

[20] Scherson D A and Newman J. J. 1980 Electrochem. Soc. 127 110

[21] Huang J 2018 Electrochim. Acta 281 170

[22] Petrakovskii G A, Loseva G V, Ryabinkina L I and Aplesnin S S 1995 J. Magn. Magn. Mater. 140–144 147

[23] Aplesnin S S, Romanova O. B., Gorev M V, Velikanov D A, Gamzatov A G and Aliev A M 2013 J. Phys. Condens. Matter 25

[24] Aplesnin S S, Ryabinkina L I, Romanova O B, Velikanov D A, Balaev A D, Balaev D A, Yanushkevich K I, Galyas A I, O. F. Demidenko O F and Bandurina O N 2008 J. Exp. Theor. Phys. 106 765