

# Electron Transport in Manganite Bicrystal Junctions

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**Abstract**—The transport and magnetic properties of junctions created in  $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$  thin films epitaxially grown on substrates with a bicrystal boundary have been investigated. In tilted neodymium gallate bicrystal substrates, the  $\text{NdGaO}_3(110)$  planes are inclined at angles of  $12^\circ$  and  $38^\circ$ . The temperature dependences of the electrical resistance, magnetoresistance, and differential conductance of the junctions at different voltages have been measured and analyzed. It has been found that the magnetoresistance and electrical resistance of the junction significantly increase with an increase in the misorientation angle, even though the misorientation of the easy magnetization axes remains nearly unchanged. The ratio of the spin-dependent and spin-independent contributions to the conductance of the bicrystal junction increases by almost an order of magnitude with an increase in the misorientation angle from  $12^\circ$  to  $38^\circ$ . The magnetoresistance of the junction increases with decreasing temperature, which is most likely associated with an increase of the magnetic polarization of the electrons. It has been shown that, at low (liquid-helium) temperatures, the conductance depends on the voltage  $V$  according to the law  $V^{1/2}$ , which indicates the dominant contribution from the electron–electron interaction to the electrical resistance of the junction. An increase in the temperature leads to a decrease in this contribution and an increase in the contribution proportional to  $V^{3/2}$ , which is characteristic of the mechanism involving inelastic spin scattering by surface antiferromagnetic magnons.

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## 1. INTRODUCTION

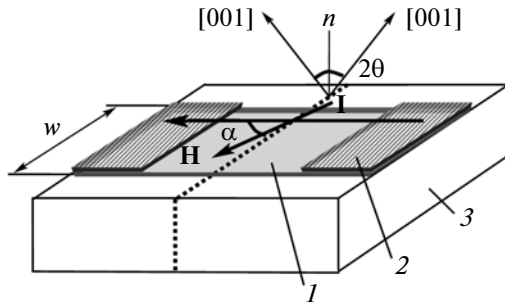
Manganites are perovskite materials based on manganese oxide  $\text{MnO}_3$ , representatives of the class of transition metal oxides. Apart from the colossal magnetoresistance (CMR), which was discovered relatively recently (in 1994) and is associated with the high-field behavior of manganites, there is a large low-field (tunneling) magnetoresistance at grain boundaries [1]. This effect has been observed in both polycrystalline and artificial bicrystal boundaries that are formed during the growth of films on bicrystal substrates or in multilayer structures [2]. Considerable interest in the tunneling magnetoresistance stems primarily from the possibility of using this phenomenon in various devices, such as magnetoresistive read heads, magnetic sensors, memory devices, etc.

Electron transport in manganite bicrystal junctions is usually considered as the charge and spin transfer through a barrier connecting two ferromagnets. The type of electron transport is determined by the structure, composition, and size of the barrier region. The majority of transport mechanisms are based on tunneling of spin-polarized carriers through a barrier. A more detailed analysis shows that there is a need to take into account nonmagnetic [1, 3] and magnetic [4–6] impurities in the barrier and the presence of a

boundary region with a modified magnetic parameters in the system [5, 7]. Moreover, there exists a model accounting for the hopping mechanism of electron transfer through the barrier region [5, 8].

In contrast to the majority of published works by authors who studied bicrystal boundaries of magnetic materials deposited on substrates with a misorientation of crystallographic axes by the rotation about the normal to the substrate plane (misoriented bicrystal junctions), we used tilted bicrystal junctions. The magnetic-field dependence of the conductance of misoriented bicrystal junctions made of  $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$  epitaxial thin films deposited on  $\text{SrTiO}_3$  substrate was discussed in [5, 6]. It was noted that the strain of manganite films near the bicrystal boundary strongly affects the magnetoresistance of the boundary. The inelastic tunneling involving magnons in magnetic tunnel junctions was investigated theoretically in [9, 10].

In most cases, the use of tilted bicrystal junctions makes it possible to significantly improve the microstructure of the boundary and to decrease the concentration of dislocations in the boundary plane as compared to misoriented bicrystal junctions [11–14]. Usually, the magnetoresistance is defined as  $\text{MR} = (R_{\text{max}} - R_0)/R_0$ , where  $R_{\text{max}}$  is the maximum resistance



**Fig. 1.** Schematic drawing of the central part of the sample with an LSMO film bridge crossing the bicrystal boundary (bridge width  $w = 8 \mu\text{m}$ ): (1) LSMO bridge crossing the bicrystal boundary, (2) Au/LSMO bilayer used for the four-point resistance measurement, and (3) NGO bicrystal substrate. The bicrystal boundary in the substrate is shown by the dotted line. The angle  $\alpha$  determines the direction of the applied magnetic field  $\mathbf{H}$  with respect to the direction of current flow  $\mathbf{I}$ . The misorientation of the crystallographic directions of two parts of the film is determined by the angle  $2\theta$ .

of the junction, which is observed in low magnetic fields and corresponds to the opposite orientations of the magnetizations of the two film parts forming the junction, and  $R_0$  is the resistance of the junction at  $H = 0$ . Using tilted bicrystal junctions, we managed to obtain the magnetoresistance higher than 150% for the  $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$  junctions with a misorientation angle of  $24^\circ$  [12] and approximately 6% for the  $\text{La}_{0.67}\text{Sr}_{0.33}\text{MnO}_3$  (LSMO) bicrystal junctions with a misorientation angle of  $38^\circ$  [13, 14]. The purpose of this work was to investigate the magnetoresistance and conductance of LSMO bicrystal junctions at low temperatures and to determine the mechanism of spin transport.

## 2. SAMPLE PREPARATION AND EXPERIMENTAL TECHNIQUE

The growth of LSMO films with a thickness of 120 nm on  $\text{NdGaO}_3$  (NGO) bicrystal substrates with a symmetric rotation of the  $\text{NGO}(110)$  planes around the  $\text{NGO}[1\bar{1}0]$  direction through angles  $2\theta = 12^\circ$  and  $38^\circ$  was performed by laser ablation in an oxygen atmosphere at a pressure  $P = 0.2$  mbar and a substrate temperature  $T = 750^\circ\text{C}$ , followed by cooling in oxygen at a pressure of 1 bar [14, 15]. For the manganite films grown on the tilted NGO substrates, the epitaxial relationships are as follows:  $\text{LSMO}(001) \parallel \text{NGO}(110)$  and  $\text{LSMO}[100] \parallel \text{NGO}[1\bar{1}0]$ . The pseudocubic lattice constant for LSMO is  $a_L = 0.388$  nm, whereas the lattice constants for the  $\text{NGO}(110)$  substrate (orthorhombic unit cell with  $a = 0.5426$  nm,  $b = 0.5502$  nm, and  $c = 0.7706$  nm) along the  $[001]$  and  $[1\bar{1}0]$  directions are  $a_N = 0.3853$  nm and  $b_N = 0.3863$  nm, respectively [15, 16]. In the case of the epitaxial growth, the

crystal structure of the bicrystal substrate is repeated in the manganite film. As a result, the bicrystal boundary is formed in the film. The strain of the film due to the compression in the substrate plane gives rise to a uniaxial magnetic anisotropy, with the easy axis directed, according to [16], along the  $\text{NGO}[1\bar{1}0]$  direction.

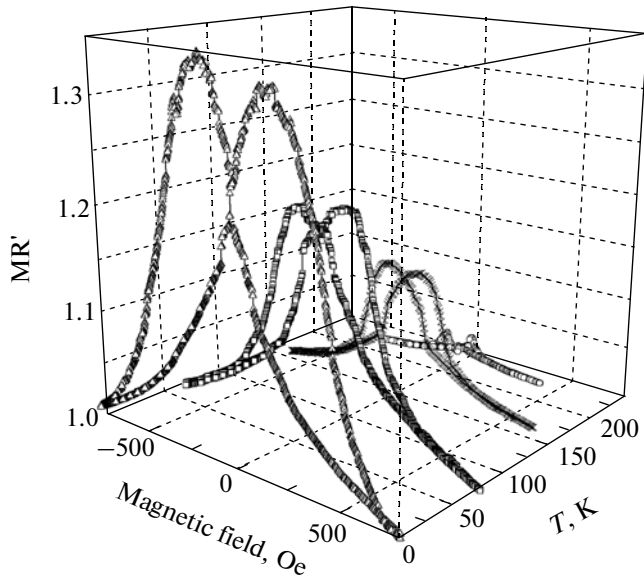
The bridges crossing the bicrystal boundary were formed by ion-beam etching using a photoresist mask and had a width of  $8 \mu\text{m}$  (Fig. 1). All the electrophysical measurements were carried out by the four-point method using platinum or gold contact pads. A direct current (dc) flowed through the bridge in the film plane perpendicular to the boundary, and the direction of the external magnetic field was varied in the film plane and was specified by the angle  $\alpha$  measured from the direction of current flow (Fig. 1).

The directions of the easy axis of the magnetic anisotropy in the LSMO films were determined using the techniques based on the resonance absorption of electromagnetic radiation by the ferromagnetic films. We measured the angular dependences of the ferromagnetic resonance spectra in the X-band and the angular dependences of the high-frequency absorption spectra in a parallel orientation. In this case, the samples were rotated through  $360^\circ$  around the normal to the substrate surface, while the dc magnetic field and the magnetic component of the electromagnetic field were always perpendicular to each other and remained in the film plane [14, 15].

## 3. MAGNETORESISTANCE

Figure 2 shows a family of magnetoresistance curves for the LSMO bicrystal junction at four temperatures. It can be seen from this figure that the low-field contribution changes the shape of the magnetoresistance curve depending on the temperature, and the maximum magnetoresistance is achieved at low (liquid-helium) temperatures. As the external magnetic field increases, the magnetoresistance decreases. Such a high-field dependence of the magnetoresistance is characteristic of manganites and is explained by the manifestation of the CMR effect in these materials [2]. The CMR effect exerts a noticeable influence on the magnetoresistance of the bicrystal junction in high magnetic fields (for our samples, this influence is observed beginning with magnetic fields of the order of 1–2 kOe). In low magnetic fields (of the order of several hundred oersteds), the observed magnetic field hysteresis is characteristic of ferromagnets.

The magnetoresistance is usually normalized using the electrical resistance of the junction  $R_{H=0}$  at  $H = 0$ . However, the electrical resistance at  $H = 0$  varies depending on the prehistory of the measurements, which introduces an uncertainty into the value of magnetoresistance (see inset to Fig. 3). In this work, as a measure of the magnetoresistance, we used  $\text{MR}' =$

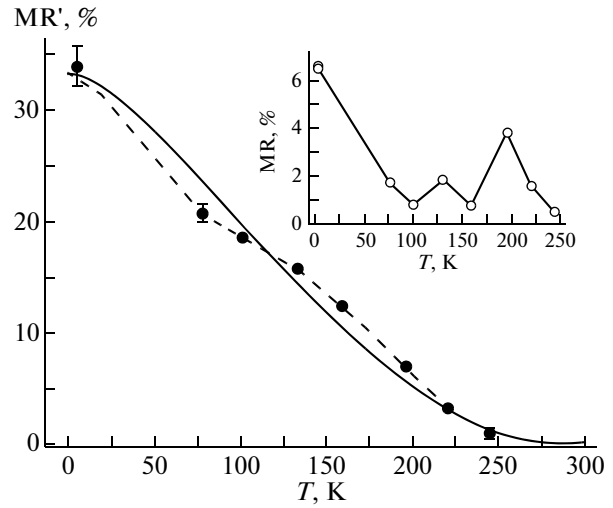


**Fig. 2.** Dependences of the magnetoresistance normalized to the electrical resistance in the magnetic field  $H = 750$  Oe for the LSMO bicrystal junction with a misorientation angle  $2\theta = 38^\circ$  at four temperatures.

$(R_{\max} - R_H)/R_H$ , where  $R_H$  is the electrical resistance of the junction in a finite magnetic field. In our case, we chose the magnetic field  $H = 0.75$  kOe, which exceeds the anisotropy field, i.e., the field in which the hysteresis effects disappear. Figure 3 shows the dependence  $MR'(T)$ , which, in contrast to the dependence  $MR(T)$  (see inset to Fig. 3), increases monotonically with decreasing temperature. In the chosen definition of the magnetoresistance, there is an error due to the presence of colossal magnetoresistance in manganite films. However, it is known that, at low temperatures and in low magnetic fields of the order of several hundred oersteds, the CMR contribution from the films forming the junction is relatively small [2].

In low magnetic fields and at liquid-helium temperatures, where the polarization is close to 100% and CMR is relatively small because the temperature is far from the Curie point, the dominant contribution comes from the magnetoresistance of the boundary. In order to estimate the contribution from the conductance of the bicrystal junction to the total conductance of the sample, we used the approach proposed in [17, 18]. In this approach, we consider the tunneling conductance of spin-polarized carriers between two ferromagnetic media separated by a tunneling barrier. It is necessary to take into account that the magnetizations on opposite sides of the barrier are directed at different angles  $\beta_1$  and  $\beta_2$  with respect to the boundary. The analytical expression for the spin conductance  $G_{sp}$  in this situation has the following form [17, 18]:

$$G_{sp} = G_{sp}^0 [1 + P^2 \cos(\beta_1 - \beta_2)]. \quad (1)$$



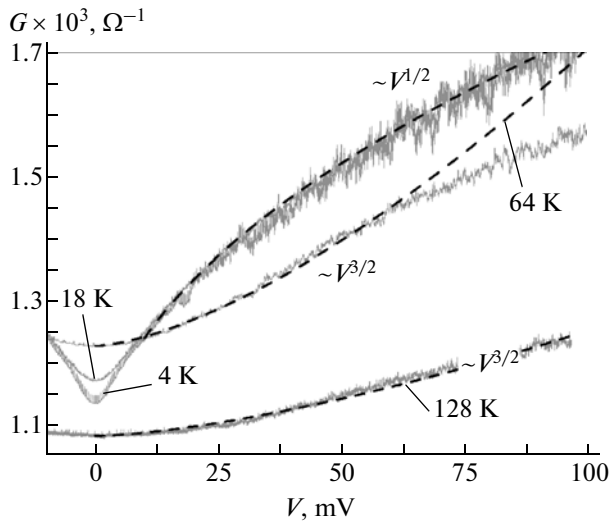
**Fig. 3.** Temperature dependence of the magnetoresistance  $MR'$  (points). The solid line is the calculated temperature dependence  $MR'(T)$ . The inset shows the dependence of the magnetoresistance  $MR$  on the temperature for the same junction.

Here,  $G_{sp}^0$  is the conductance of the polarized spins and  $P$  is the polarization of the spin. Taking into account, for completeness, the contribution to the conductance from the non-polarized carriers  $G_{ns}$ , we can write the expression for the electrical resistance of the tunneling barrier [10, 19]:

$$R = \frac{1}{G_{sp} + G_{ns}} = \frac{R_{sp}}{1 + P^2 \cos(\beta_1 - \beta_2) + g}. \quad (2)$$

Here,  $R_{sp} = 1/G_{sp}$  and  $g \equiv G_{ns}/G_{sp}$ .

Our measurements performed for tilted bicrystal junctions using the techniques based on the resonance absorption of electromagnetic radiation have demonstrated that the misorientation of the easy magnetization axes of the two film parts forming the junction is rather small (approximately  $1^\circ$ ). But, at the same time, the values of the anisotropy fields differ significantly. For a rough estimate, we will further assume that the autonomous (without a field) magnetizations  $M_1$  and  $M_2$  are directed parallel to each other. For sufficiently high values of the external magnetic field, the magnetizations are aligned parallel to each other and directed along the external field. According to our calculations for films with uniaxial anisotropy, the maximum magnetoresistance is observed in the vicinity of the anisotropy fields where the reorientation of the magnetizations of the two film parts of the junction takes place. It is at this point where the external magnetic field is approximately equal to the anisotropy field, the electrical resistance reaches the maximum value  $R_{\max}$ . It is well known that the polarization of



**Fig. 4.** Dependences of the conductance on the applied voltage  $G(V)$  for the LSMO bicrystal junction with a misorientation angle of  $38^\circ$  at different temperatures (experimental curves). Dashed lines show the fits of the experimental curves by power-law functions.

manganites at low temperatures is close to 100%. We assume that  $P = 1$ . Then, from expression (2), we find that the maximum resistance is determined by the equality  $R_{\max} = R_{\text{sp}}/(1 - P^2 + g)$ . For high magnetic fields, when the directions of the magnetizations  $M_1$  and  $M_2$  are parallel to each other and coincide with the direction of the magnetic field, we have  $R_H = R_{\text{sp}}/(1 + P^2 + g)$ . Taking into account the above-defined quantity  $\text{MR}'$ , we estimate the ratio of the conductances of polarized and non-polarized carriers as follows:

$$\text{MR}' = (R_{\max} - R_H)/R_H = 2P^2/(1 - P^2 + g). \quad (3)$$

Substituting the 100% polarization into expression (3), we obtain

$$\text{MR}' = 2/g. \quad (4)$$

Using the data presented in Fig. 3 and expression (4), we obtain  $g = G_{\text{ns}}/G_{\text{sp}} = 6.7$ . Therefore, the measured dc conductance of the bicrystal junction is predominantly determined by the transfer of non-polarized carriers. It is assumed that the temperature dependence of the polarization has the power-law form [10, 20, 21]

$$P(T) = P_0(1 - \varepsilon T^{3/2}). \quad (5)$$

Substituting expression (5) in place of  $P$  into formula (3) and taking into account the experimental value  $\text{MR}' = 6.5\%$ , we find  $\varepsilon = 2 \times 10^{-4} \text{ K}^{-3/2}$ . This value of  $\varepsilon$  is close in the order of magnitude to the available data obtained using photoemission spectroscopy for the free surface of LSMO films  $\varepsilon = 4 \times 10^{-4} \text{ K}^{-3/2}$  [20], but, at the same time, it is an order of magnitude different from  $\varepsilon = 4 \times 10^{-5} \text{ K}^{-3/2}$  for magnetic tunnel

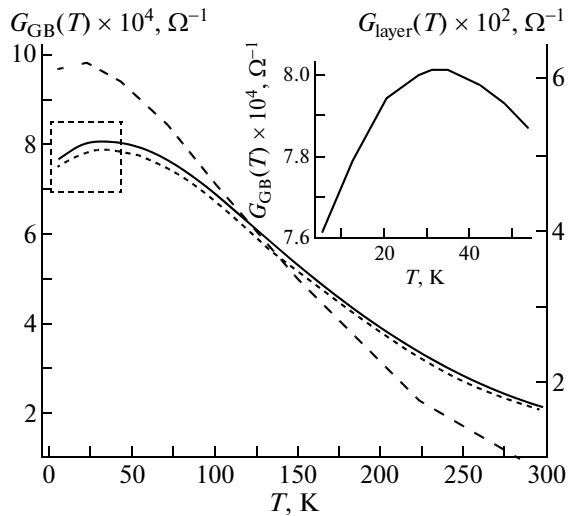
structures based on LSMO films with STO interlayers [20, 21]. For junctions with a small misorientation angle ( $2\theta = 12^\circ$ ), the magnetoresistance reaches a few fractions of a percent. In this case, the characteristic resistance of the junction  $RA$  is smaller (where  $A$  is the junction area), although the misorientation of the easy magnetization axes remains almost unchanged [14]. It should be noted that the magnetoresistance is significantly higher in  $\text{La}_{0.67}\text{Ca}_{0.33}\text{MnO}_3$  (LCMO) bicrystal junctions, where there is a transition layer with a lower Curie temperature in vicinity of the bicrystal boundary and the characteristic resistance of the boundary is greater than that for the LSMO bicrystal junctions [12].

#### 4. DEPENDENCE OF THE CONDUCTANCE OF THE JUNCTIONS ON THE APPLIED VOLTAGE

In order to determine the mechanism of charge carrier transfer, we measured a family of dependences of the conductance of the junctions on the voltage at different temperatures in the range from 4.2 to 300 K. Next, we choose a model that best describes the charge carrier transport in our experiment.

The bicrystal junction is considered as a tunnel junction with two metallic ferromagnetic electrodes and a rectangular potential barrier between them. The electron transport can be described by the mechanism of elastic tunneling through a rectangular barrier [22]. However, this model is applicable only for low temperatures and low voltages. In this model, when there occurs no spin transfer, there is no dependence of the electrical resistance on the magnetic field. A change in the dependence of the conductance on the voltage occurs as a result of the change in the shape of the barrier under a voltage at the junction and has the following form:  $G(V) = G_0 + G_2|V^2|$ , where the contribution satisfies the inequality  $G_0 \gg G_2|V^2|$ . Taking into account the fact that the dependence  $G(V)$  in our experiments (Fig. 4) in a wide range is not described by the law  $V^2$  and differs significantly from the linear function  $G = G_0$  and also that we observe strong dependences of the electrical resistance on the magnetic field even at several tens of oersteds, this mechanism of conduction will not be considered.

The development of the previously described model is a model that takes into account the presence of the boundary layer in the vicinity of the bicrystal junction. The properties of this layer can differ significantly from the properties of the electrodes due to additional scattering centers and the short mean free path. The boundary layers are most clearly observed in bicrystal junctions of LCMO films with a lower Curie temperature [12]. In the boundary layer, there can occur an increase of the electron-electron interaction, for example, due to the diffuse nature of the motion of electrons in a disordered dirty metal [23, 24]. In manganites, the strong electron-electron interaction is



**Fig. 5.** Dependence of the conductance on the temperature for a  $38^\circ$  bicrystal film (dotted line), an autonomous film (dashed line), and a bicrystal junction (solid line). The inset shows the conductance of the bicrystal junction at low temperatures.

responsible for the metallic conduction. The dependence of the conductance on the voltage has the form  $G(V, T) = G_0 + G_{1/2}|V^{1/2}|$ . The conductance  $G_0$  depends on the magnetic field  $H$ , and the quantity  $G_{1/2}|V^{1/2}|$  decreases rapidly with an increase in the temperature, which was actually observed in disordered metal oxides [24] at temperatures up to 10 K. Indeed, in our experiment at low temperatures ( $T \leq 18$  K), the dependence  $G(V)$  has a clearly pronounced contribution proportional to  $V^{1/2}$ . At higher temperatures, this contribution decreases and, in the field dependence of the conductance, is almost negligible (at  $T = 64$  K and above).

The hopping mechanism of conduction of the junction with an inhomogeneous barrier, which was considered by Glazman and Matveev [25], suggests the temperature dependence  $G(T) \sim T^{4/3}$ , which, in our case, is not observed. Furthermore, in the theory [25], there is no dependence of the conductance on the magnetic field.

The scattering of charge carriers by magnetic excitations with a nonlinear field dependence was considered in [9, 26]. The model of scattering of spin-polarized carriers [26] suggests the dependence  $G(V) = G_0 + G_2|V^2| + G_{3/2}|V^{3/2}|$  for the conductance of the magnetic junction. Here, the term  $G_2|V^2|$  is determined by bulk magnons, whereas the term  $G_{3/2}|V^{3/2}|$  is governed by surface antiferromagnetic magnons. The contribution  $G_{3/2}|V^{3/2}|$  becomes dominant for the dependence  $G(V)$  at temperatures above 50 K.

Therefore, as follows from the analysis of the dependence of the conductance of the bicrystal junction on the bias voltage, in our junctions there are two

mechanisms: (i) the electron–electron interaction at low temperatures, which suggests the presence of a boundary layer in the system, and (ii) the scattering of spin-polarized carriers by antiferromagnetic magnons in the boundary region at higher temperatures [27]. An increase in magnetoresistance with a decrease in the temperature is caused by the increase in the magnetic polarization and the weakening of the spin scattering mechanism.

The existence of two spin scattering mechanisms is confirmed by the temperature dependence of the conductance of the LSMO bicrystal junction and an autonomous film of the same composition (Fig. 5). It can be seen that a decrease in the temperature leads to a change in the temperature dependence of the conductance of the bicrystal junction in the range of 30 K. With an increase in the temperature, the conductance increases at temperatures below 30 K and decreases above this temperature.

## 5. CONCLUSIONS

The measurements of the angular dependence of the magnetic field corresponding to ferromagnetic resonance in bicrystal junctions have revealed that there are two ferromagnetically ordered spin subsystems in which the directions of the easy magnetization axes differ insignificantly (by approximately  $1^\circ$ ). The easy magnetization axes are directed along the bicrystal boundary and almost do not depend on the angle of crystallographic misorientation of the bicrystal substrate parts. The magnetoresistance (MR') increases with decreasing temperature; however, even at  $T = 4.2$  K, when the polarization of the LSMO films is close to 100%, MR' reaches only 30% for bicrystal junctions with a  $38^\circ$  misorientation of the planes. With a decrease in the misorientation angle, MR' decreases significantly and, at  $2\theta = 12^\circ$ , amounts to a few fractions of a percent. It has been shown that the low value of the magnetoresistance can be associated with both the scattering of spin-polarized carriers due to the strong electron–electron interaction in the disordered boundary layer at low temperatures and the scattering of spin-polarized carriers by antiferromagnetic magnons at high temperatures.

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