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Maximum magnetic field dependence of longitudinal magnetoresistance in thin bismuth films

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The longitudinal magnetoresistance in bismuth films was studied as a function of thickness and deformation in the film plane. The presence of a maximum turning into a negative magnetoresistance was found on the magnetofield dependences of the longitudinal magnetoresistance. It was shown that the difference of the sizes of the crystallites of the films makes a much smaller contribution to the displacement of the maximum by the magnetofield dependence than the effect of deformation in the film plane because of the difference of the coefficient of thermal expansion of bismuth and the substrate material. The different nature of the maximum formation for films with a thickness greater than and less than 100 nm was shown.

Keywords: Thin films, bismuth, longitudinal magnetic resistance, quantum limit in magnetic field, chiral anomaly, Weyl semimetal, weak electron localization effect.

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1. Introduction

Narrow-band semiconductors and semimetals are good model materials for the experimental study of quantum effects. This is attributable to the low values of the Fermi energy and the low effective mass of the electrons, which leads to large values of the de Broglie wavelength of the electrons. The possibility of growing highly perfect crystals and films makes it possible to achieve high mobility of charge carriers.

Bismuth plays an important role in solid state physics, because many effects were first discovered in it (the Shubnikov-de Haas, de Haas–van Alphen effects, cyclotron resonance in metals). Active studies of bismuth are currently under way because of the growing interest in coherent phenomena in the transport properties of materials [1–8], as well as increased attention to Dirac materials and topological insulators [9–19].

The non-monotonic nature of the magnetofield dependences of magnetoresistance in a longitudinal magnetic field (the direction of current parallel to the magnetic field) has not received a generally accepted interpretation despite significant interest in the transport properties of bismuth thin films [20].

Non-monotonic changes are observed in the magnetic field dependences of the longitudinal magnetoresistance in thin films, as well as in massive bismuth crystals. A decrease of resistance in a strong magnetic field occurs in a massive bismuth crystal under the impact of an increase of the concentration of charge carriers in the ultraquantum region of magnetic fields [21,22]. The change of electrical resistance in a parallel magnetic field at low temperatures in thin bismuth films is also characterized by an increase of a weak field and a decrease of a strong field. A maximum is formed in the intermediate field area [23]. Moreover, unlike a massive single crystal in thin films, there are several possible causes for the occurrence of the maximum depending on the longitudinal magnetic resistance and the magnitude of the magnetic field.

First, the maximum of longitudinal magnetoresistance in thin films can occur when the classical dimensional effect in galvanomagnetic properties is realized. The drop of resistance with the increase of field is attributable to the fact that an increasing number of electrons avoid surface scattering as the average values of the radius of the electron orbits decrease, which is accompanied by a decrease of its contribution to the resistance of the sample. The maximum position is determined by the condition [3]:

$$B_{\max} \approx \frac{P_F}{e}\,(lt)^{-1/2}, \tag{1}$$

where t — film thickness, l — free path length of charge carriers, P_F — Fermi momentum of charge carriers. If P_F and l are constant, the value of $B_{\max}$ should increase with the decrease of the film thickness.

The same reason as in a massive single crystal of bismuth, associated with an increase of the concentration of charge carriers in the ultraquantum region of magnetic fields may be the second reason for the non-monotonic dependence of the magnetoresistance in case of the realization of the longitudinal effect in the case of thin films [21,22]. An increase of the concentration of holes and electrons in the ultraquantum region is demonstrated in calculations performed in Ref. [24] for the case of the direction of the magnetic field along the binary axis of the crystal ($H \parallel C_2$).

The minimum value of the magnetic field for reaching the quantum limit is determined by the orientation of the magnetic field relative to the crystallographic axes and does not depend on the free path of the charge carriers. The transition to the ultra-quantum region of magnetic fields takes place in case at different field values for samples with different concentrations of charge carriers. The value of $B_{\max}$ increases with the increase of the charge carrier concentration.

The effect of weak localization manifested in weakly disordered metals and is caused by the quantum mechanical interference of electrons during their scattering may by the third reason. This effect leads to a decrease of the mobility of charge carriers and, as a result, to an increase of the resistivity of the material. The effect of weak localization is destroyed in the magnetic field, therefore, a negative magnetoresistance, i. e. a decrease of resistivity in the magnetic field is one of the characteristic features of weak localization [25,26].

Weak localization of electrons leads to a different decrease of conductivity for the three-dimensional and two-dimensional cases. For the three-dimensional case:

$$\Delta\sigma_{3d} = G_0 L_\varphi^{-1}, \tag{2}$$

where $G_0 = \frac{e^2}{2\pi^2\hbar} = 1.23 \cdot 10^{-4}\,\Omega^{-1}$; L_φ — the length of the electron phase failure. The two-dimensional case is realized if the film thickness is less than L_φ and the electrons are elastically reflected from the film surface. The decrease of the conductivity of the thin film due to weak localization of electrons is determined by the expression:

$$\Delta\sigma_{2d} = G_0 \ln\left(\frac{\tau_\varphi}{\tau}\right). \tag{3}$$

where τ_φ is the relaxation time of the electron wave function phase, τ is the relaxation time of the electron pulse.

The effect of weak localization decreases when the magnetic field is applied. The correction to the conductivity of a thin film is determined by the following expression in the case of a parallel magnetic field:

$$\Delta\sigma_{2d}^{B_{\text{II}}} = G_0\left[\frac{3}{2}\ln\left(\frac{t^2e^2B^2D}{2\hbar^2}\,\tau_\varphi^* + 1\right) - \frac{1}{2}\ln\left(\frac{t^2e^2B^2D}{2\hbar^2}\,\tau_\varphi + 1\right)\right], \tag{4}$$

where B is a magnetic field induction, D is a diffusion coefficient, $\frac{1}{\tau_\varphi^*} = \frac{1}{\tau_\varphi} + \frac{4}{3}\frac{1}{\tau_{SO}}$, where τ_{SO} is a spin relaxation time due to spin-orbit interaction [27]. In the case when $\tau_{SO} \gg \tau_\varphi$ quantum corrections of weak localization lead to negative magnetoresistance.

The contribution of a chiral anomaly in the Weyl semimetals may be the fourth reason [28.29]. See [30] to learn more about the theory of topological semimetals. The conduction band and the valence band coincide in energy at some points of the Brillouin zone in Dirac and Weyl semimetals. T- or P-symmetry is broken in Weyl semimetals, unlike Dirac semimetals, which leads to the removal of degeneracy from Dirac points, and they become separated in k-space into pairs of Weyl points with different chirality. Therefore, Weyl semimetals are searched for among crystals with a non-centrally symmetric crystal structure, or among magnetic materials (with broken T-symmetry).

The chiral anomaly in Weyl semimetals is caused by the application of parallel electric and magnetic fields. The abnormal contribution to electrical conductivity [29]:

$$\sigma = N_W\,\frac{e^2\nu\tau}{4\pi\hbar L_B^2}, \tag{5}$$

where $L_B = \sqrt{\hbar/eB}$ — magnetic length, ν — Fermi velocity of electrons, N_W — number of Weyl points. It can be seen from the expression (5) that the chiral anomaly in Weyl semimetals can make a linear contribution to the negative longitudinal magnetoresistance.

The energy spectrum of the bismuth crystal in the L point of the Brullien zone has a gap of the order of 10 MeV [31,32]. Numerous studies have demonstrated that bismuth films grow on various (crystalline and amorphous) substrates in such a way that the trigonal axis of the crystallites is perpendicular to the plane of the substrate [33–39]. A bismuth film located on a substrate with a TEC different from bismuth is subject to stretching or compression deformation in a plane in accordance with this crystallographic orientation, which is generally equivalent to uniaxial deformation along the trigonal axis of the crystal [40,41]. The amount of deformation was estimated by X-ray diffraction analysis in Ref. [42]. This deformation, in the case of stretching in the trigonal plane, leads to a change of the bismuth band structure with a decrease and subsequent closure of the gap in the L point of the Brillouin zone, according to ab initio modeling [43]. High pressure and, consequently, mechanical deformations can also disturb the P symmetry. An example of such a result, obtained by ab initio calculation for a solid solution of bismuth-antimony, is given in Ref. [44]. Thus, changes of the band structure of bismuth in the thin-film state can lead to the occurrence of the Weyl semimetal phase with a certain amount of planar deformation created by the difference in the TEC of the substrate and film materials.

An attempt is made in this paper to determine the main causes leading to non-monotonicity and negative magnetic resistance depending on the magnetic field at low temperatures in bismuth films. The magnetofield dependences of the magnetoresistance of bismuth films formed on substrates with different thermal expansion coefficients (TEC), as well as films of various thicknesses at temperatures of 10 K in a magnetic field up to 8 T were studied for this purpose. The maximum of the longitudinal magnetoresistance based on the magnetofield dependence for films with a thickness of more than 100 nm is associated in the discussion with the achievement of the ultraquantum region of magnetic fields. Moreover, possible reasons for the formation of a maximum on magnetofield dependences for thinner films are discussed, the main of which are the effects of an increase of the concentration of charge carriers with an increase of the magnetic field in the ultraquantum region of magnetic fields, the effect of a chiral anomaly in Weyl semimetals, and the effect of weak localization.

2. Experimental methodology

The method of thermal evaporation in vacuum was used to obtain bismuth films. The purity of the initial bismuth was 99.999%. The following materials were used for the substrates: mica (muscovite) of grade SOV, Si, CaF_2, NaCl. The use of substrates with different TEC makes it possible to obtain different concentrations of charge carriers in films due to film deformation in accordance with Ref. [18,45]. The TEC value for the used substrates and the estimated ratio of the charge carrier concentration in the films on these substrates (n_{film}/n_{bulk}) are listed in the table. The substrates were coated with AD polyimide-based varnish for creating a block structure similar in size to the crystallites. The coating thickness did not exceed 500 nm. In addition, films on a mica substrate were also examined without coating.

Two methods of thermal sputtering in vacuum were used to obtain films of different thicknesses. The method of resistive heating of the material in vacuum to 10^{-3} Pa was used in the case of films with a thickness of more than 150 nm. The following parameters were used for film sputtering: deposition rate — 5 nm/s, substrate temperature during deposition — 413 K, annealing was carried out at a temperature of 513 K for 60 min. Bismuth films with a thickness of 150 nm or less were obtained in a system with an electron beam evaporator with the following sputtering parameters: vacuum level — up to 10^{-6} Pa, deposition rate — 0.1 nm/s, substrate temperature during deposition 413 K, annealing was carried out at a temperature of 473 K for 60 min.

The thickness of films with a thickness of more than 150 nm was checked by optical interferometry using MII-4 Linnik microinterferometer. The thickness of films with a thickness of 150 nm or less was checked by atomic force microscopy (AFM) in accordance with the methodology described in Ref. [46]. The error of determination of the film thickness of the methods used did not exceed 10%.

The crystal structure of the films was examined using X-ray diffractometer „DRON-7" from Burevestnik and a scanning probe microscope Solver-P47 Pro from NT-MDT. The relationship between the surface and volume structure of bismuth films is described in Ref. [38,39,47], which was used to determine the volume structure of films with a thickness of less than 100 nm.

The resistance was measured at a temperature of 10 K in the range of magnetic fields from 0 to 8 T using a four-probe method at direct current. The measurement error of the relative magnetoresistance was no more than 5%.

Relative concentration of charge carriers in bismuth films n_{film}/n_{bulk} at 77 K on substrates with different TEC (α) [45]

Material of substrate	Si	Mica	Bi(111)	CaF_2	NaCl
α, $10^{-6}\,K^{-1}$	2.54	7.5	11	18	39
n_{film}/n_{bulk}	0.45	0.8	1	1.6	2.9

The scientific equipment of the Shared-Access Interdisciplinary Resource Center „State of art physical and chemical methods for the formation and study of materials for the needs of industry, science and education" (Herzen University) was used in the paper.

3. Results and discussion

An analysis of the X-ray diffraction results showed that the predominant direction of the crystallographic axis C_3 of the films is perpendicular to the film plane. The indexing of diffraction maxima from bismuth films has shown that they belong to the trigonal plane of the bismuth single crystal. The displacement of the position of the maxima on various substrates corresponds to the concept of stretching of the trigonal plane of bismuth films on substrates with a TEC less than that of the film material, and compression on substrates with a TEC of the substrate material exceeding the TEC of the film material.

In addition, the surface structure of bismuth films was studied by the AFM method. Figure 1 shows AFM images of bismuth films formed on various substrates. The growth patterns have the same shape and orientation within the same crystallite. The boundaries of the crystallites are distinguished by thin solid lines due to the natural oxidation of the film. The structure of the obtained films differs primarily in the size of the crystallites. Bismuth films formed on single-crystal mica substrates had a large-block structure with crystallite sizes an order of magnitude larger than the film thickness (Figure 1, *a*). In contrast, bismuth films formed on polyimide-coated substrates had a small-block structure. The sizes of the crystallites were of the order of the thickness of these films (Figure 1, *b*).

Figure 2 shows the results of measurement of the resistivity for bismuth films with a thickness of 300 nm in a longitudinal magnetic field from 0 to 8 T and the longitudinal magnetic resistance in the specified range of magnetic fields. It can be seen from the presented dependences for films on pure mica and mica coated with varnish that the sizes of the film crystallites have a lesser effect on the maximum position than the use of a different substrate, i. e. the effect of deformation. The position of the maximum of the longitudinal magnetoresistance for bismuth films on mica is in the region of 2 T, both with and without a polyimide sublayer. The deformation caused by the difference of the TEC of the film and the substrate leads to a more significant change of the position of the

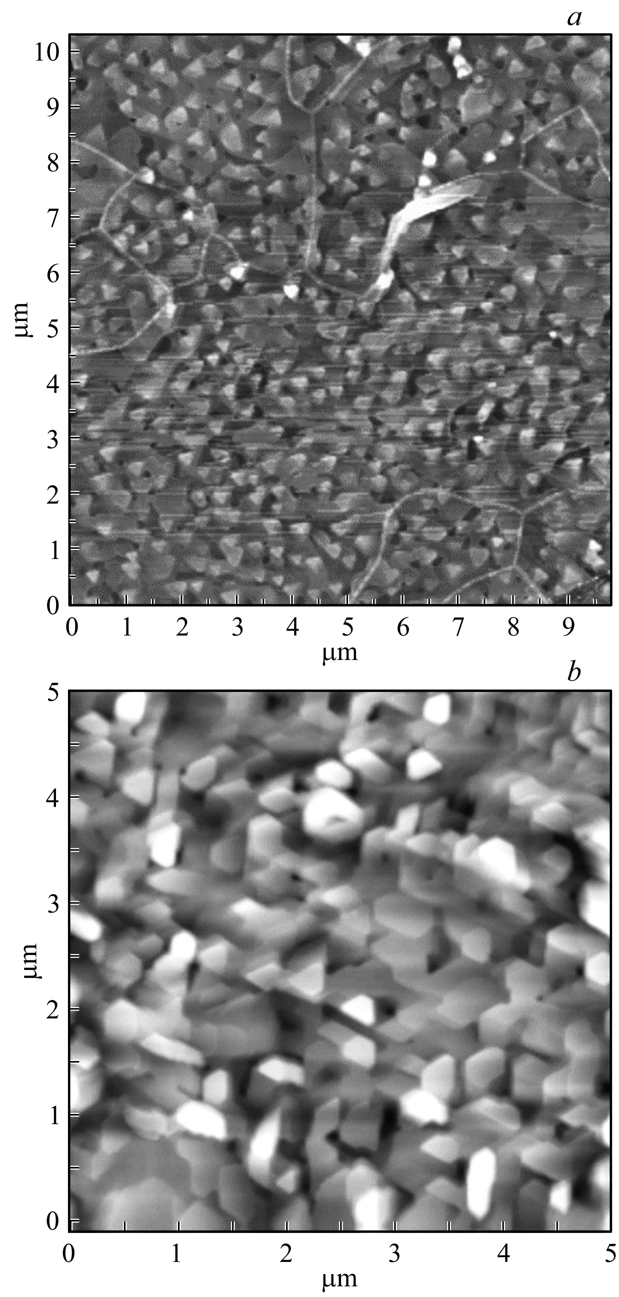

Figure 1. AFM images of bismuth films (thickness 300 nm): *a* — formed on a mica surface; *b* — formed on a polyimide-coated substrate.

maximum based on the magnetofield dependence in this case. A slight shift of the maximum position for a film on a mica substrate coated with polyimide relative to the maximum on pure mica can be explained by the effect of compression deformation created by the buffer layer. The TEC of polyimide $(40-48)\cdot 10^{-6}\,\mathrm{K}^{-1}$, depending on the composition and method of imidization) is greater than the TEC of mica and bismuth, which, despite the smallness of the layer, leads to some compression deformation.

As noted in the introduction, an increase of the concentration of charge carriers in the ultraquantum region of magnetic fields may be one of the reasons for the presence of a maximum on the dependence of the longitudinal magnetoresistance in bismuth in both films and single crystals. The magnitude of $B_{\max}$ of the magnetic field corresponding to the maximum magnetic resistance increases with the increase of compression strain (NaCl and CaF substrates$_2$) and decreases with the increase of tensile deformation (mica and silicon substrates). Based on data on changes in the concentration of charge carriers in bismuth films on various substrates (see the table), it can be concluded that the value $B_{\max}$ increases with the increase of the concentration of charge carriers and decreases with the decrease of the concentration, which is a characteristic feature of reaching the ultraquantum region of magnetic fields.

For this reason it is of interest to study the effect of film thickness on the value of $B_{\max}$, since an increase of the charge carrier concentration is observed with a decrease of the bismuth film thickness according to many studies [48–51]. This effect is most studied in bismuth films on mica. The increase of concentration becomes especially strong for thicknesses less than 50–100 nm.

Figure 3 shows the results of a study of the longitudinal magnetoresistance of bismuth films with a thickness of 20 to 1000 nm on a mica substrate. It can be seen from the figure that the position of the maximum magnetic resistance practically does not depend on the thickness in films with a thickness of more than 100 nm, however, the maximum magnetic resistance abruptly shifts to the region of weaker magnetic fields for smaller thicknesses. The character changes at a thickness of 113 nm according to the results of extrapolation of experimental results (Figure 3, *b*). This nature of the thickness dependence of the maximum magnetoresistance for films with a thickness of less than 100 nm indicates that the achievement of the ultraquantum region of magnetic fields cannot be the reason for its formation in them, since the concentration of charge carriers increases in bismuth films with a decrease of thickness [48–51]. The classical dimensional effect also cannot be the reason for the observed dependence, since $B_{\max}$ should increase with the decrease of the film thickness if it is realized in accordance with the expression (1).

It should be especially noted that when a magnetic field is applied, a maximum of magnetic resistance is not formed for a film with a thickness of 45 nm, and a negative value of magnetic resistance is observed in the entire range of the studied fields. The dependence of the longitudinal magnetoresistance on the magnitude of the magnetic field is close to linear in this case. Linear negative magnetoresistance is also observed for a film with a thickness of 20 nm, but the effect is weakly expressed in it, since the magnitude of the magnetoresistance itself is small due to significant scattering of charge carriers by the film thickness.

It is obvious that the maximum on the magnetofield dependences for films with a thickness of 45 nm and films with a thickness of more than 100 nm is attributable to various mechanisms. Moreover, the shape of the magnetofield

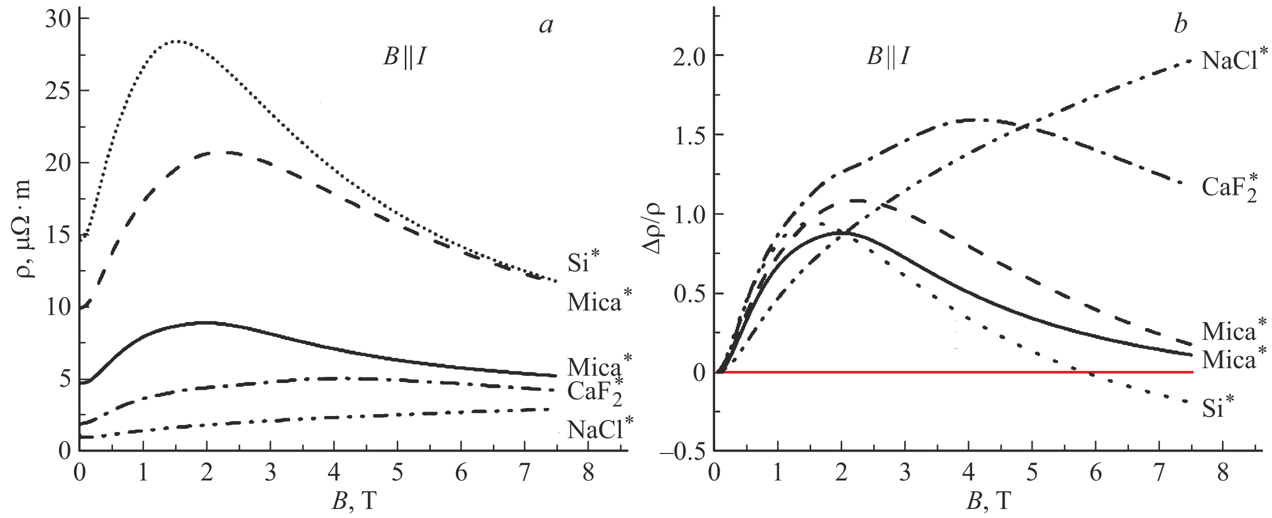

Figure 2. Dependences of the resistivity (*a*) and magnetoresistance (*b*) of bismuth films (300 nm) on substrates with different TEC on the magnitude of the longitudinal magnetic field. The substrates coated with a buffer layer of polyimide are marked by the asterisk sign.

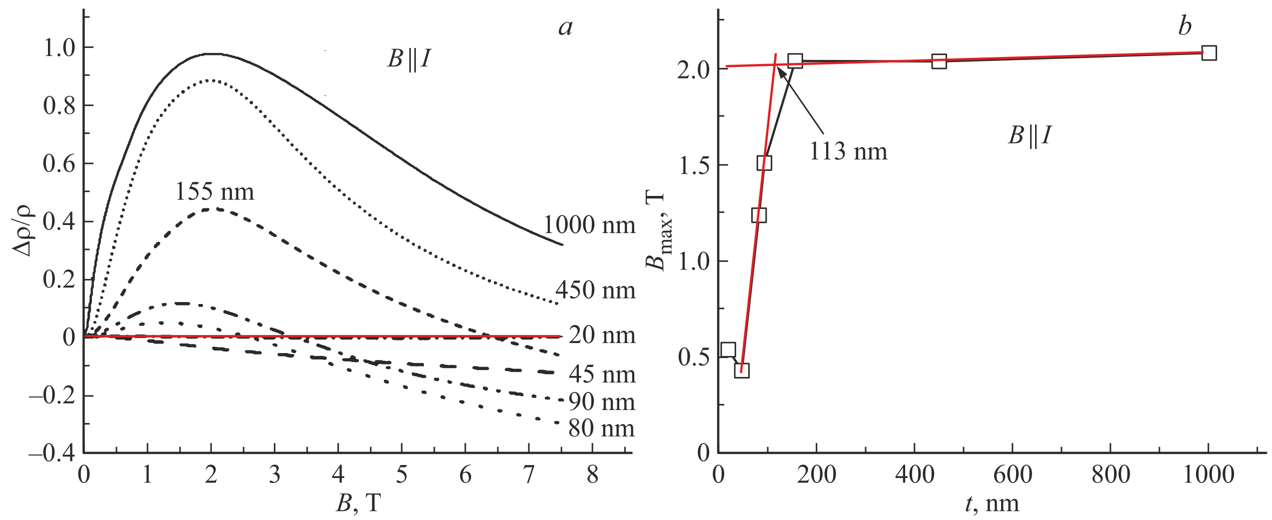

Figure 3. Position of $B_{\max}$ depending on the film thickness: *a* — magnetofield dependences of magnetoresistance for bismuth films with a thickness of 20 to 1000 nm on a mica substrate; *b* — dependence of the position of $B_{\max}$ on the thickness of the film (solid black line with squares — experimental results; solid red lines — extrapolations of two areas with different slope dependence).

dependences is determined by the combination of these two mechanisms in samples of intermediate thickness.

The observed linear dependence of the negative magnetoresistance on the magnitude of the magnetic field for a 45 nm thick bismuth film on mica may be a manifestation of a chiral anomaly in Weyl semimetals in accordance with the formula (5). However, such a dependence can also be observed in a certain range of magnetic fields for the effect of weak localization in accordance with expression (4). Therefore, further study is required to establish the nature of this phenomenon. One of the ways may be to study the longitudinal magnetoresistance in films with a thickness of less than 100 nm formed on substrates with higher and lower TEC than mica.

4. Conclusion

The results of a study of the longitudinal magnetoresistance of bismuth films of various thicknesses under conditions of various deformations in the film plane are presented in this paper. The deformation was created by using substrates with various TEC. An experiment was conducted using substrates coated with a thin buffer layer of polyimide to study bismuth films with identical structures obtained on different substrates.

The study of the magnetofield dependences of the longitudinal magnetoresistance of bismuth films with large-block and small-block structures formed on substrates with different temperature expansion, as well as films of various thicknesses at temperatures of 10 K in a magnetic field up to 8 T has shown that a maximum can be formed on the magnetofield dependences of the longitudinal magnetoresistance in the specified range of magnetic fields in thin bismuth films that passes into a negative magnetoresistance.

An analysis of the results showed a different nature of the formation of a maximum for films with a thickness greater than and less than 100 nm. This maximum in films with a thickness of more than 100 nm is formed due to the achievement of the ultra-quantum region of magnetic fields and the subsequent increase of the concentration of charge carriers. A negative magnetoresistance in a 45 nm thick bismuth film on a mica substrate may be a manifestation of a chiral anomaly in Weyl semimetals or a consequence of the weak localization effect. The shape of the magnetofield dependences in the intermediate thickness range is apparently determined by a combination of these two mechanisms.

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Conflict of interest

The authors state that they have no conflict of interest.

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