

Spin-orbit interaction in thin bismuth films

Yu. F. Komnik, I. B. Berkutov, and V. V. Andrievskii*

B. N. Verkin Institute for Low-Temperature Physics and Engineering, Ukrainian National Academy of Sciences, 47 pr. Lenina, Khar'kov 61103, Ukraine
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The magnetic-field dependences of the resistance of thin (100–700 Å thick) bismuth films at low temperatures are analyzed using quantum corrections to the conductivity with weak electron localization. It is shown that the spin-orbit scattering time τ_{so} is much shorter than the phase relaxation time τ_φ of the electrons (the case of a strong spin-orbit interaction). It is found that τ_{so} tends to increase with the film thickness. This shows that the surface scattering of the electrons plays a dominant role in spin-orbit processes. Apparently, strong spin relaxation in the presence of surface scattering is due to the gradient of the internal crystal-field potential near the surface of the metal, resulting in lifting of the spin degeneracy and in the appearance of a spin gap (Rashba mechanism). © 2005 American Institute of Physics.
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1. INTRODUCTION

The behavior of the conductivity of a metal in a magnetic field yields information about the structure and parameters of the electronic energy spectrum.¹ For example, the asymptotic behavior of the conductivity in strong magnetic fields reflects the topological properties of the electronic spectrum, and the magnetoquantum effects (Shubnikov-de Haas oscillations) yield information about the characteristic parameters of the spectrum.² An understanding of quantum effects of an interference nature, weak electron localization,^{3–5} and the electron-electron interaction^{5–8} has opened up the possibility of obtaining information about the relaxation times and interaction parameters of electrons from the behavior of the magnetoresistance.

Thin bismuth films were the first object where the effects of weak localization and the interaction of electrons were observed.^{9–12} Subsequently, these effects were observed in many weakly disordered conductors of different dimensions.^{13,14} According to theory,⁵ the contribution of weak localization to the conductivity of a two-dimensional system as a function of a magnetic field B oriented perpendicular to the film plane has the form

$$\Delta\sigma_B = \sigma(B) - \sigma_0(B) = \frac{e^2}{2\pi^2\hbar} \times \left[\frac{3}{2} f_2 \left(\frac{4eBD}{\hbar} \tau_\varphi^* \right) - \frac{1}{2} f_2 \left(\frac{4eBD}{\hbar} \tau_\varphi \right) \right], \quad (1)$$

where $f_2(x) = \ln x + \Psi(1/x + 1/2)$; Ψ is the logarithmic derivative of the Γ function; D is the electron diffusion coefficient; τ_φ is the phase relaxation time; τ_φ^* is the modified time taking account of the spin-orbit interaction: $(\tau_\varphi^*)^{-1} = \tau_\varphi^{-1} + 4/3\tau_{so}^{-1}$; and, τ_{so} is the spin relaxation time due to the spin-orbit interaction.

For a weak spin-orbit interaction ($\tau_{so} \gg \tau_\varphi$) Eq. (1) describes a positive magnetoconductivity (negative magnetoresistance), which is typical for the manifestation of weak localization. For a strong spin-orbit interaction ($\tau_{so} < \tau_\varphi$) the

second term in Eq. (1) determines $\Delta\sigma_B$; this corresponds to the manifestation of an anomalous positive magnetoresistance. Then Eq. (1) becomes

$$\Delta\sigma_B = -\frac{1}{2} \frac{e^2}{2\pi^2\hbar} f_2 \left(\frac{4eBD}{\hbar} \tau_\varphi \right). \quad (2)$$

The case of a positive anomalous magnetoresistance in the weak localization effect has been termed “antilocalization.”

It is a positive magnetoresistance that has been found to be characteristic for thin bismuth films manifesting weak localization, whence it was concluded that a strong spin-orbit interaction is characteristic for this object. The reason for this remained unclear. After all, in a bulk bismuth crystal whose crystal lattice is described by a weakly distorted cube with a center of inversion there is no reason for a strong spin-orbit interaction to appear in the absence of a magnetic field.

In works concerning the effects due to weak localization in bismuth films the expression (2) was used to describe the behavior of the magnetoconductivity, so that information was obtained only about the phase relaxation time τ_φ . In Ref. 12 it is noted that the spin-orbit interaction time τ_{so} in bismuth films is much shorter than τ_φ and apparently varies with the film thickness. Modern computers make it possible to analyze the change in magnetoconductivity using Eq. (1) and to determine the behavior of τ_{so} as a function of the thickness and other parameters. This could be helpful for determining why the spin-orbit interaction in bismuth films is strong.

2. EXPERIMENTAL AND COMPUTATIONAL PROCEDURES

The experimental magnetic-field dependences of the resistance measured at liquid-helium and higher temperatures for four series of thin-film bismuth samples, in each of which three or four thicknesses were represented, were analyzed. The samples were obtained by condensing a molecular bismuth beam in a high vacuum (5×10^{-6} mm Hg) on a substrate (glass, mica) at room temperature. The films possessed texture: the C_3 axis was oriented in a direction normal to the film plane. The film thickness range was 100–700 Å; the

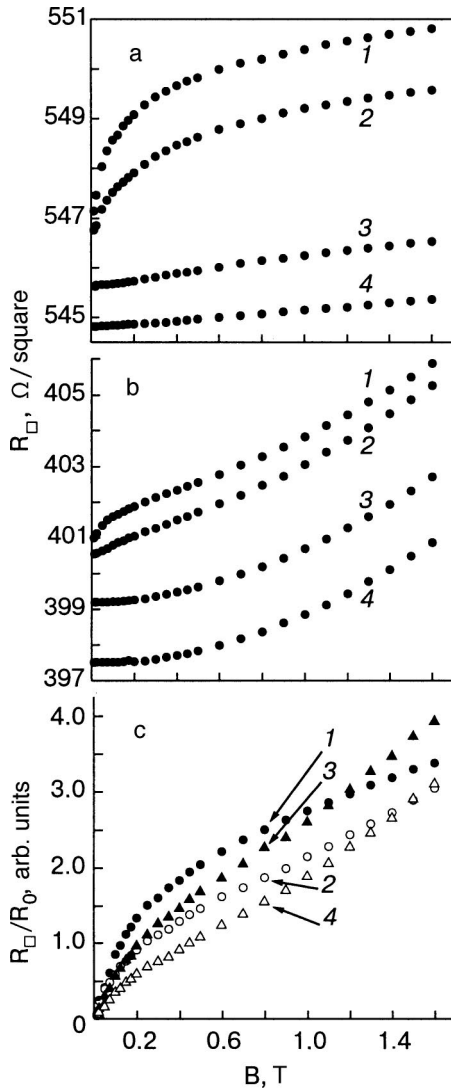


FIG. 1. Magnetic field variation of the resistance of samples in series 3 with thicknesses 380 Å (a) and 570 Å (b) at temperatures T , K: 2 (1), 4.2 (2), 14 (3), and 20 (4). Magnetic field variation of the resistance of samples in series 2 with thicknesses (in Å): 180 (1), 280 (2), 400 (3), and 450 (4) at temperature 4.2 K (c).

effects due to weak electron localization are not manifested clearly for larger thicknesses. The sample series differed somewhat by the conditions under which the films were condensed (substrate temperature and condensation rate) and, correspondingly, by their transport characteristics. The traces of the magnetoresistance curves were obtained in magnetic fields up to 1.6 T at temperatures 1.5–4.5 K and also at 14–20 and 77 K (examples of magnetoresistance traces are displayed in Fig. 1).

For comparing with theory the magnetoresistance traces were converted into the magnetic-field variation of the conductivity of a two-dimensional system using the relation $\Delta\sigma = -(\Delta R/R) \cdot (1/R_\square)$, where R_\square is the resistance of a square section of film, which is an analog of the resistivity in the two-dimensional case. This transformation is valid because the ratio $\Delta R/R$ is small and because measurements have shown that for a perpendicular orientation of the magnetic field the Hall component of the magnetoresistance $R_{\square,xy} \sim 10^{-2} R_{\square,xx}$.

To determine the quantum correction introduced into the

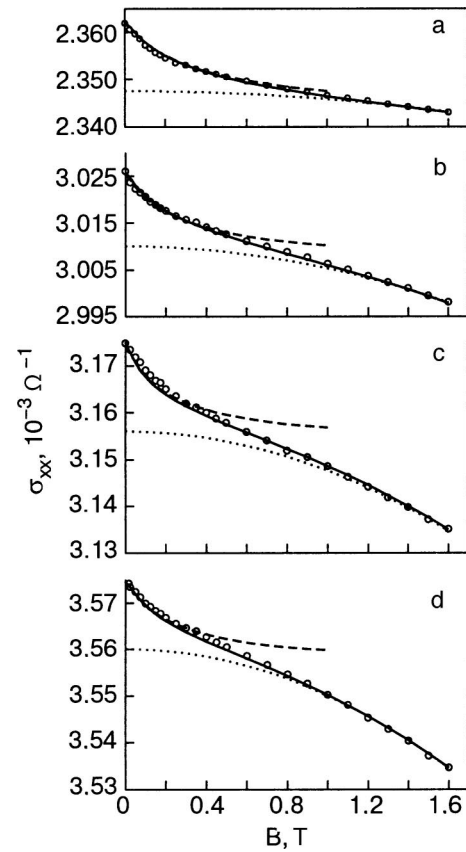


FIG. 2. Description of the experimental dependences (○) of the conductivity of bismuth films on the magnetic field by Eq. (1) for weak localization taking account of the Drude contribution to the magnetoconductivity (—); separation of the Drude contribution (···) from the magnetic field variation of the conductivity; magnetic field variation of the quantum correction due to weak localization (with respect to the conductivity at $B=0$) (---). Examples are demonstrated for samples in series 2 at temperature $T=4.2$ K with different thicknesses (in Å): 180 (a), 280 (b), 400 (c), and 450 (d).

conductivity by weak electron localization it is important to take account of the contribution of the classical (Drude) magnetic-field variation of the resistance of the form $\mu^2 B^2$ (μ is the mobility) to the magnetoresistance, which was not done in previous works on weak localization in bismuth films. We used a technique proposed in Ref. 15, specifically, in the range of logarithmic saturation of the function (1) the mobility values for which the function $\sigma_B = \sigma_0^D / (1 + \mu^2 B^2)$ is identical to the experimental dependence $\sigma(B)$ were determined.¹⁾ In Fig. 2 the dotted line demonstrates the procedure for finding the Drude magnetoresistance of one series of samples.

Next, the value of the “two-dimensional” concentration n_2 follows from the expression $\sigma_0 = (R_\square^0)^{-1} = ne\mu$. The relation $n_3 = n_2 L^{-1}$, where L is the film thickness, should be used for comparing with the data on the “three-dimensional” concentration obtained in Refs. 16–18. It should be noted that bismuth films exhibit a quantum-size effect,^{19–21} resulting in resistance oscillations as a function of the film thickness. These oscillations are clearly seen for thicknesses $L > 1000$ Å. Resistance oscillations can be neglected in the thickness range studied in the present work.

After the Drude contribution was removed from the magnetic-field dependences obtained for the conductivity (see dashed curves in Fig. 2), the dependence calculated us-

ing Eq. (1) and the desired values of the parameters τ_φ and τ_{so} were matched with the experimentally determined dependence reflecting the contribution of quantum corrections. The diffusion coefficient D was determined using Einstein's formula $D = G/2e\nu$, where G is the specific volume conductivity and ν is the electron density of states at the Fermi surface (for bismuth films $\nu \approx 10^{20} \text{ eV}^{-1} \text{ cm}^{-3}$ taking account of the change in the charge carrier concentration and the Fermi energy with decreasing film thickness¹⁷). The cyclotron mass of the electrons and holes in bismuth with the magnetic field oriented along the C_3 axis is $m^* = 0.063m_0$, where m_0 is the free-electron mass. It is this value of the effective mass of electrons and holes that was used to determine the elastic time τ from the conductivity. The time τ was found to be of the order 10^{-15} s .

The matching procedure was quite successful in all cases. This is demonstrated in Fig. 2 (solid curves). Aside from weak electron localization, the contribution of other corrections due to the electron-electron interaction is negligible. Our estimates show that for reasonable values of the interaction constant λ_B^D and known values of the Landé factor g (for $B \parallel C_3$ $g = 1.06$ for electrons and $g = 4.26$ for holes) for bismuth the quantum corrections to the magnetic-field dependence of the conductivity in the diffusion channel are much smaller than for weak localization (in contrast to their temperature dependence where the interaction corrections are greater than the correction due to weak localization).²² Similarly, including in the matching procedure the quantum correction due to interaction in the Cooper channel simply degrades appreciably the agreement between the experimental curves and Eq. (1) describing weak electron localization.

3. COMPUTATIONAL RESULTS

Our calculations of the contribution of weak localization to the magnetic-field variation of the conductivity of bismuth films differ from previous calculations^{9,10} by the following important features: in the first place the expression (1) was used in the calculations, making it possible to determine τ_{so} , and in the second place the contribution of the Drude variation of the magnetoconductivity was singled out and taken into account.

The computational results agree with the basic behavior of characteristics such as the concentration and mobility, determined in Refs. 16–18, where the galvanomagnetic properties of bismuth films were studied.

All calculations were performed in the SI system. The values found for the mobility lie in the range $0.01\text{--}0.07 \text{ m}^2\text{V}^{-1}\text{s}^{-1}$, which agrees with calculations of the average mobility using the system of equations for galvanomagnetic properties of bismuth films.¹⁸ The values of μ increase smoothly with film thickness in each series, but they are different for different series because of differences in the structural characteristics of the films. This is also observed for the thickness dependences of the diffusion coefficient D and the average mean-free path length l . In the more perfect series 1 and 2 the path lengths l are somewhat greater than the thickness L ; in the series 3 and 4 l is slightly less than the film thickness.

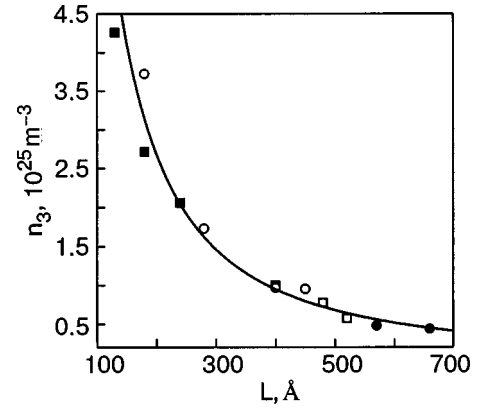


FIG. 3. Charge carrier concentration versus bismuth film thickness for different series: 1 (●), 2 (○), 3 (■), and 4 (□). Dependence of the form $n_3 = 7.6 \times 10^{28} \cdot L^{-1.5}$ (—).

In contrast to the transport characteristics the values of the “three-dimensional” charge carrier concentration fall on a single curve versus the film thickness (see Fig. 3). This dependence is essentially the same as that obtained in calculations using the system of equations for galvanomagnetic properties of films.^{16–18} The increase in carrier concentration with decreasing bismuth film thickness, first established in Ref. 16, was explained in Refs. 17 and 23 as being due to the nonuniform distribution of the potential, which has a bend near the surfaces, in the film. The nonuniform static potential in a semimetal results in an effective increase of band overlapping and increases the average carrier density.

The film-thickness and temperature dependences of the times τ_φ and τ_{so} are qualitatively different. The phase relaxation time τ_φ does not exhibit a clear dependence on L (Fig. 4a), and the spin-orbit relaxation time τ_{so} tends to increase somewhat with increasing thickness (Fig. 4b). As temperature increases, τ_{so} remains unchanged and τ_φ decreases (Fig. 5). This time is determined by inelastic scattering

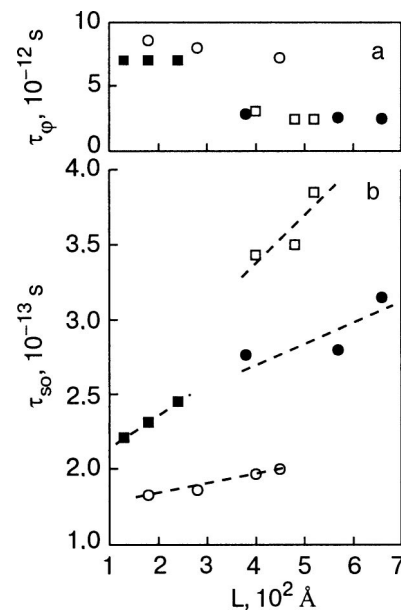


FIG. 4. Variation of the phase time τ_φ (a) and spin-orbit relaxation time τ_{so} (b) with increasing bismuth film thickness. The labeling of the series of samples is the same as in Fig. 3. The broken lines are drawn for clarity.

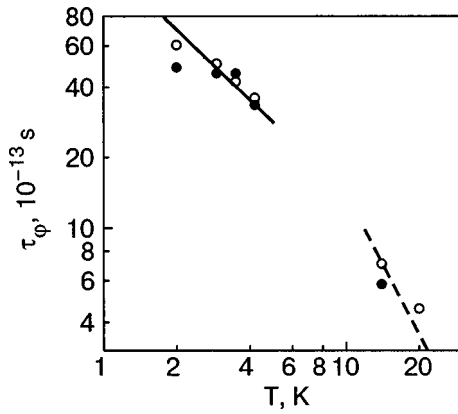


FIG. 5. Temperature variation of the phase relaxation time τ_ϕ of samples in series 3 with thickness, (in Å): 380 (○) and 570 (●); $\tau_\phi = 3.9 \times 10^{-13} \cdot T^{-1}$ (—), $\tau_\phi = 4 \times 10^{-12} \cdot T^{-2}$ (---).

processes—electron-electron and electron-phonon. At temperatures below 4 K electron-electron scattering predominates; for these processes the theory of the electron-electron interaction in disordered two-dimensional conductors predicts $\tau_{ee}^{-1} \propto T$.^{7,8} This dependence is satisfactorily realized for the groups of points in Fig. 5 below 4 K. As temperature increases, the electron-phonon scattering processes play an increasingly larger role. The dependence $\tau_\phi^{-1} \propto T^2$ is observed in bismuth films at temperatures above 4 K.²⁴ The reasons for the appearance of such a temperature dependence of τ_ϕ for bismuth films at $T > 4$ K and for beryllium,²⁵ niobium, and aluminum²⁶ films at $T > 10$ K are discussed in Ref. 24.

4. DISCUSSION

In this work the values of the time τ_{so} in bismuth films indicating a strong spin-orbit interaction were obtained. It was also found that τ_{so} tends to increase with film thickness.

The high spin-orbit scattering rate in thin bismuth films is due, in our opinion, to carrier interaction with the surface. We shall try to substantiate this below.

The idea that spin-orbit interaction plays a substantial role for electrons in small samples was first advanced in Refs. 27 and 28 in connection with the observation of a nonzero nuclear magnetic resonance frequency shift in superconducting films as $T \rightarrow 0$ (Knight shift). It was inferred that the direction of the spin of an electron scattered by the boundary can change, as a result of which the total spin of the system is no longer zero. In connection with this idea, Abrikosov and Gor'kov constructed a theory of spin-orbit interaction in the scattering of electrons by impurities.²⁹ The experimental data for the spin-orbit relaxation time determined from the Knight shift, the critical magnetic field of ultrathin films, experiments on tunneling of spin-polarized electrons into a superconductor, and spin resonance of conduction electrons in normal metals were analyzed in Ref. 30. In this work a check was made of whether or not the dependence of τ_{so}^{-1} on the atomic number Z of a metal satisfies the relation from Ref. 29:

$$\frac{\tau_{so}^{-1}}{\tau - 1} \approx (aZ)^4, \quad (3)$$

where τ is the elastic scattering time and $a = e^2/\hbar c = 1/137$ is the fine structure constant. The ratio $\tau_{so}^{-1}/\tau - 1 = \varepsilon$ characterizes the probability of a spin-orbit process in elastic scattering. In Ref. 30 it was assumed that since all data for τ_{so} refer to thin films, the main momentum scattering mechanism is surface scattering, so that the transit time $\tau^{sf} = L/v_F$ of an electron between two surfaces, where v_F is the Fermi velocity, was used as the time τ . It was found that the Z dependences of $\tau_{so}^{-1}/(\tau^{sf})^{-1}$ functionally agree with Eq. (3), but the experimental values are severalfold greater than the values calculated from this expression, ranging from small values ($\sim 10^{-5} - 10^{-7}$) for light metals (such as Li) up to very large values for heavy metals (for Sn ~ 0.1 , Pb ~ 0.5). Thus, for heavy metals a collision of an electron with the surface is very likely to be accompanied by a change in spin orientation. The strong spin-orbit interaction observed in bismuth films (the time τ_{so} turned out to be close in order of magnitude to the elastic scattering time τ) corresponds to the results of an analysis of the experimental data for τ_{so} in Ref. 30.

The ratio of the probabilities of spin-orbit scattering processes at the surface ε^{sf} and in the bulk ε^b of a film was estimated in Refs. 31 and 32. In Ref. 31 it was found for Mg films that ε^{sf} is one to two orders of magnitude greater than ε^b , depending on the type of substrate. In Ref. 32 it was also established for Au films that $\varepsilon^{sf} \gg \varepsilon^b$ (the values obtained were $\varepsilon^{sf} \approx 2 \times 10^{-2}$ and $\varepsilon^b \approx 4 \times 10^{-4}$).

It is completely obvious that the tendency for τ_{so} to increase with film thickness (Fig. 4b) shows that the computed values of τ_{so} reflect primarily spin-orbit processes at a surface, since a decrease of the influence of the surface with increasing thickness where $\varepsilon^{sf} \gg \varepsilon^b$ should result in a weaker spin-orbit interaction in the film, i.e. an increase of τ_{so} . We shall illustrate this using the following qualitative relations. Let the conduction electrons in the film undergo elastic scattering in the bulk and at the surface of the film. Let the time τ_{so} found be represented by two components for spin-orbit processes in the bulk and at the surface:

$$\frac{1}{\tau_{so}} = \frac{1}{\tau_{so}^b} + \frac{1}{\tau_{so}^{sf}} = \frac{\varepsilon^b}{\tau^b} + \frac{\varepsilon^{sf}}{\tau^{sf}}, \quad (4)$$

where τ^b and τ^{sf} are the corresponding times of the elastic processes. Let $\tau^b \approx l/v_F$ and τ^{sf} equal to approximately L/v_F in the “pure” limit ($l > L$), and in the “dirty” limit ($l < L$) let τ^{sf} be determined by the electron diffusion time from one surface to the other $\tau^{sf} \approx L^2/D$, where the diffusion coefficient $D \approx v_F l$. Then it follows from the expression (4) that

for the “pure” limit

$$\frac{1}{\tau_{so}} = \frac{\varepsilon^b v_F}{l} + \frac{\varepsilon^{sf} v_F}{L}, \quad (5)$$

for the “dirty” limit

$$\frac{1}{\tau_{so}} = \frac{\varepsilon^b v_F}{l} + \frac{\varepsilon^{sf} v_F l}{L^2}. \quad (6)$$

It is obvious that for $\varepsilon^b \ll \varepsilon^{sf}$ the integral time τ_{so} found in both cases should increase with film thickness. An intermediate case ($l \approx L$) is realized in the objects studied. This makes it impossible to use the formulas presented above for the analysis.

We believe that the reason for the strong spin-orbit process accompanying surface reflection of electrons could be a mechanism associated with the bending of the potential near the surface of a conductor, which follows from the requirement that the wave function of charge carriers vanish at the boundary. The spin state of an electron changes in a region where a gradient $\nabla V(r)$ of the potential exists. The spin-orbit interaction Hamiltonian has the form³³

$$H_{so} = \frac{\hbar}{(2m_0c)^2} [\nabla V(r) \times \mathbf{p}] \hat{\sigma}, \quad (7)$$

where \mathbf{p} is the quasimomentum of an electron and $\hat{\sigma}$ is a Pauli matrix. The electron spin is oriented perpendicular to its momentum and to the gradient of the potential. For $k \neq 0$ (k is the wave number) the spin degeneracy is lifted and the states are split (the Rashba mechanism). The corresponding theory^{34,35} developed for a two-dimensional electron gas in heterojunctions in semiconductors predicts the appearance of two branches in the electron energy spectrum:

$$E^{\pm}(k) = \frac{\hbar^2 k^2}{2m} \pm \alpha k, \quad (8)$$

(α is the spin-splitting parameter) and the existence of a spin gap

$$\Delta_s = E^+ + E^- = 2\alpha k_F. \quad (9)$$

These ideas have been extended to inversion layers in semiconductors, i.e. regions of bending of the potential near a surface.

The existence of two branches in the spectrum of near-surface electrons with different spin polarization creates an additional channel for the spin-orbit process accompanying surface reflection of electrons. As a result the probability of a spin-orbit process accompanying scattering by the surface of a crystal is much higher than for elastic scattering of electrons by impurity atoms.

5. CONCLUSIONS

In summary, according to the ideas presented above, the answer to the question of why the manifestation of weak localization in thin bismuth films has the character of “antilocalization” is that the strong spin-orbit relaxation in thin bismuth films is associated primarily with surface scattering of electrons. It is supposed that electron scattering from a surface occurs under conditions where a gradient of the crystal potential exists near the surface, lifting the spin degeneracy in the electron spectrum near the surface and giving rise to intense spin relaxation accompanying surface scattering. As the film thickness increases and the contribution of surface scattering to the conductivity decreases, the spin-orbit interaction should become much weaker, as is qualitatively confirmed by the fact that the values found for the spin-orbit relaxation time τ_{so} tend to increase.

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*E-Mail: Andrievskii@ilt.kharkov.ua

¹⁾We have in mind the average mobility of two types of carriers, since calculations have shown^{16–18} that the electron and hole mobilities found for bismuth films from the system of equations for the conductivity, the magnetoresistance, and the Hall coefficient are close. The electron and hole concentrations remain equal to one another.

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