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Quantum Effects of Non-Ballistic Transport in Films Based on Compound PbSnAgTe

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Based on the theory of weak localization, taking into account the mechanism of spin-orbit scattering, the patterns of change in the magnetic conductivity of films PbSnAgTe are considered. The dependences of the magnetoresistance of PbSnAgTe films in magnetic field perpendicular to the surface of the film are studied.

It is shown that for polycrystalline films on mica-muscovite substrates, the time of spin-orbital interaction depends on the composition and may change the sign of the magnetoresistance.

Keywords: quantum effects, weak localization, thin films, lead telluride, kinetic phenomena.

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Introduction

Lead telluride is a well-known material for sources and detectors of infrared radiation in the optical spectrum [1, 2]. Semiconductor materials such as PbTe are also suitable for studying non-ballistic effects. Due to the high dielectric permittivity $\varepsilon = 1350$ at 4.2 K in PbTe and the low effective mass $m^* = 0.024m_0$, this leads to effective screening from ionized impurities and defects. The result is a very high mobility of bulk samples, which may exceed 10^2 m^2/Vs at low temperatures [2]. Thanks to these properties, PbTe can be used in a new field - spintronics. One possible application of PbTe is to use it as a spin filter [3], due to the high value of the Lande factor.

The negative magnetoresistance, which is often observed in semiconductors, is explained by the theory of weak localization and spin-orbital interaction [4]. These effects are purely quantum effects as a result of the interference of an electron with itself and the interaction of an electron with its spin in a magnetic field. In this case, quantum corrections to magnetoresistance are used.

For films of undoped PbTe negative values of magnetoresistance were observed in [5]. However, the vast majority of studies show that the magnetoresistance phenomenon for PbTe is described by classical laws. The dependence of the magnetoresistance on the temperature for one-dimensional material in the form of nanowires was investigated in [6]. But even for 1d structure there is no manifestation of the effect of non-ballistic transport. The introduction of impurities, i.e. the modification of the electronic subsystem, and obtaining polycrystalline films, can affect on the processes of charge transport. The magnet transport measurements for p-type films of $Pb_{1-x}Eu_xTe$ for different composition and temperatures are presented in work [7]. It turns out, that the introduction of a paramagnetic impurity influences on the interaction of carriers of current and leads to the appearance of weak localization and strong spin-orbit interaction due to the Zeeman effect.

In this paper, the experimental dependences of the magnetoresistance of PbSnAgTe films from the composition in a magnetic field perpendicular to the surface of the film are obtained. Their explanations are analyzed based on the theory of quantum corrections to conductivity associated with weak localization and spin-orbit interaction. It should be noted that doping by Sn in the investigated compositions significantly changes the electrical properties, as well as the type of conductivity. The compositions have different types of conductivity: $Pb_{14}Sn_4Ag_2Te_{20}$ and $Pb_{16}Sn_2Ag_2Te_{20}$ – p-type, $Pb_{18}Ag_2Te_{20}$ – n-type. This is confirmed by the Hall measurements of the carrier mobility μ (Table 1).

I. Experimental details

The samples were grown by depositing of the vapor phase of the presynthesized material $Pb_{14}Sn_4Ag_2Te_{20}$,

$Pb_{16}Sn_2Ag_2Te_{20}$ and $Pb_{18}Ag_2Te_{20}$ materials in a vacuum on fresh chips (0001) of mica-muscovite substrate. The temperature of the evaporator was T_e = 870 K and the temperature substrates T_s = 470 K. The thicknesses of the films are set by the deposition time within (1-3) min and are measured by microinterferometer MII-4 using digital image processing methods.

Measurement of electrical parameters of the films was at temperatures from 77 K to 300 K and at constant magnetic fields on the automated device. It provides a process for measuring electrical parameters and initial registration and processing of data. Measured sample had four Hall contacts and two current contacts. As the ohmic contacts was used a silver film. The current through the sample was ≈ 1 mA. The magnetic field was directed perpendicular to the film surface. The induction of magnetic field was 0 – 1.2 Tesla. For measurement of the temperature were used platinum thermoresitors. Measurement error is not more than 3%.

Theoretical calculation for describing the character of the dependence of the magnetoresistance in a perpendicular magnetic field is carried out using the means of the mathematical package Maple 18.

II. Theoretical part

The motion of an electron in a disordered electronic system isn't ballistic, but diffuse, when the electron collides many times with impurities and other defects of the crystalline lattice. Such a quantum effect as a weak localization for a system of non-interacting electrons is due to the wave properties of these quantum particles. And there is an interference of electronic waves when electrons are scattering. One of the manifestations of the effect is the appearance of a negative magnetoresistance, that is, the conductivity of the system increases with the increase of the magnetic field induction. Therefore, quantum corrections for conductivity, associated with the above mentioned effect, are used. The correction for conductivity due to weak localization is determined by the equation [4]:

$$ds = -\frac{2e^2D}{p\mathbf{h}}t\,C(r,r',w)\,, \qquad (1)$$

where *C(r, r', ω)* –cooperon, which determines the amplitude of scattering on the impurity, D is the diffusion coefficient, e is the electron charge, k_B is Boltzmann's constant, μ is the mobility of the carriers, is the time of elastic scattering. The function *C(r, r', ω)* satisfies the equation [4]:

$$\left[-iw+\frac{D}{\mathbf{h}^2}\left(-i\nabla-\frac{2e}{\mathbf{h}}\mathbf{A}\right)^2+t_j^{-1}\right]C(\mathbf{r},\mathbf{r}',w)=\frac{d(\mathbf{r}-\mathbf{r}')}{t}, \qquad (2)$$

where ω – the frequency of the external field, A – is the vector potential of the magnetic field.

For films of arbitrary thickness in the magnetic field perpendicular to the surface, the boundary conditions are defined as

$$\left.\left(\Delta_z-\frac{2ie}{\mathbf{h}}A_z\right)\right|_{z=0}C=0\,,\qquad \left.\left(\Delta_z-\frac{2ie}{\mathbf{h}}A_z\right)\right|_{z=d}C=0\,.$$

The solution of the differential equation (1) within the diffusion approximation for ω = 0 will be [8] the expression:

$$C(r,r')=\mathbf{h}\sum_n\frac{y_n(\mathbf{r})y_n^*(\mathbf{r}')}{4eBD\left(n+\frac{1}{2}\right)+\mathbf{h}t_j^{-1}+\mathbf{h}D\left(\frac{pm}{d}\right)^2}, \qquad (3)$$

$\psi_n(\mathbf{r})$ – normalized wave functions of a particle with charge 2e in a magnetic field, n, m are quantum numbers, B is the magnetic field induction, d is the thickness of the film, τ_φ is the phase relaxation time of the wave function.

Substituting (3) into (1) for the magnetoconductivity $\Delta\sigma = \delta\sigma(B) - \delta\sigma(0)$ and taking into account that $d\rightarrow\infty$ we obtain:

$$\Delta s\,(B)=\frac{e^2}{2p^2\mathbf{h}}f_2\left(\frac{4eDB}{\mathbf{h}}t_j\right) \qquad (4)$$

where $f_2(x)=\ln x+\Psi\left(\frac{1}{x}+\frac{1}{2}\right)$, $\Psi(x)$ – Digamma function, or logarithmic derivative of the gamma function.

Spin-orbital interaction strongly affects the magnetoresistance of the system, since it leads to relaxation of the spin. In this case, the magnetoprotection sign may even change. The Hamiltonian of electrons in the conduction band for cubic crystals has the form [4]

$$H=\frac{p^2}{2m^*}+\boldsymbol{\sigma}\left(d\cdot p_x\left(p_y^2-p_z^2\right)\right). \qquad (5)$$

σ – Pauli matrix, p is the quasimomentum of an electron.

As a result, the cooperon will be the next:

$$C(r,r')=\frac{3}{2}\mathbf{h}\sum_n\frac{1}{4eBD\left(n+\frac{1}{2}\right)+\mathbf{h}t_j^{-1}+2\mathbf{h}t_{so}^{-1}}-\frac{1}{2}\mathbf{h}\sum_n\frac{1}{4eBD\left(n+\frac{1}{2}\right)+\mathbf{h}t_j^{-1}}, \qquad (6)$$

τ_{so} – spin relaxation time.

Then the dependence of the contribution of the quantum correction caused by the spin-orbit interaction, as a function of the magnetic field, directed perpendicular to the plane of the film, has the form:

$$\Delta s\,(B)=\frac{e^2}{2p^2\mathbf{h}}\left[\frac{3}{2}f_2\left(\frac{4eDB}{\mathbf{h}}t_j^*\right)-\frac{1}{2}f_2\left(\frac{4eDB}{\mathbf{h}}t_j\right)\right], \qquad (7)$$

τ_φ^*– modified time, taking into account spin-orbital interaction, $\left(t_j^*\right)^{-1}=t_j^{-1}+\frac{4}{3}t_{so}^{-1}$.

The size of the system is an important parameter when the processes of non-ballistic transport are considered. It is determined by the ratio between the smallest geometric film size and the length of diffusion during the time of the phase relaxation of the wave function $L_j=\sqrt{t_jD}$. If $L_\varphi>>d$, then the electronic system is considered to be two-dimensional (2D), and if $L_\varphi<<d$ – three-dimensional (3D) in the theory of weak localization. In order to analyze the magnetoresistance behavior of the studied films, it is necessary to take into account the fact that they are thick or quasi-two-dimensional. Therefore, in the extreme case of a thick

film $d>L_\varphi$, the correction for conductivity will be calculated taking into account the size of the film, like it was done in [8]:

$$\Delta\sigma_{Q2D}(\mathrm{B})=\frac{e^2}{2\pi^2\hbar}\cdot\frac{d}{l_B}\cdot\left[\frac{3}{2}f_2\left(\frac{4eDB}{\hbar}\tau_\varphi^*\right)-\frac{1}{2}f_2\left(\frac{4eDB}{\hbar}\tau_\varphi\right)\right],\ (8)$$

where $l_B=\sqrt{\hbar/eB}$ – magnetic length. It is often the parameter that characterizes the behavior and dimension of the electronic system in relation to the theory of weak localization with $l_B \ll L_\varphi$. If we change the magnetic field, we can change the dimension of the system.

In the case of weak spin-orbital interaction ($\tau_{so} \gg \tau_\varphi$) from formula (7), the magnetic conductivity will be positive, respectively, quantum corrections determine the negative magnetoresistance. This is one of the manifestations of weak localization. When there is the strong spin-orbital interaction ($\tau_{so} \ll \tau_\varphi$) an anomalous positive magnetoresistance with a logarithmic saturation in strong fields arises. The case of anomalous positive magnetoresistance in the theory of weak localization is called antilocalization. This phenomenon is observed in the presence of a spin-orbital interaction in the system, when the electron spin can turn over (change its direction) during the elastic scattering of the electron on the impurity or on the surface. Then in formula (7) the second term will be determinative and it changes to the next

$$\Delta\sigma(\mathrm{B})=-\frac{1}{2}\frac{e^2}{2\pi^2\hbar}f_2\left(\frac{4eDB}{\hbar}\tau_\varphi\right), \qquad (9)$$

If the time of the spin-orbital interaction is compared with the time of relaxation of the phase of the wave function of the electron $\tau_{so} \le \tau_\varphi$ the magnetoresistance curve in the positive region passes through the maximum and then becomes negative. Thus, the appearance of the curves of the dependence of conductivity or resistance on magnetic fild can qualitatively estimate the relation between τ_φ and τ_{so} and get information about the presence of spin-orbital interaction.

III. Results and discussion

In fig. 1 are shown the experimental dependences of the relative change in the resistivity $((\rho(B)-\rho(0))/\rho(0))$ on the induction of a magnetic field, which are oriented perpendicular to the plane of the sample, for films based on PbSnAgTe (LATT) compounds at a temperature T = 77 K. Taking into account the form of curves in Fig. 1. it can be said that the classical dependence of the magnetoresistance $\sim\mu B^2$ is executed only for a film of PbTe. For LATT compounds an anomalous dependence of the magnetoresistance is observed. For $Pb_{18}Ag_2Te_{20}$ films, the magnetoresistance is negative, for the film $Pb_{16}Sn_2Ag_2Te_{20}$ – it has a maximum in the positive region, and for $Pb_{14}Sn_4Ag_2Te_{20}$ - positive with a logarithmic dependence on the magnetic field. So, for the description of the electronic system we will use the effects of localization and anti-localizations. In addition, the magnetoresistance of films based on PbSnAgTe compounds is greater than for PbTe. Consequently, PbSnAgTe films have greater sensitivity to magnetic field changes and can be used as magnetic field sensors.

It should be noted that the films of the lead telluride also exhibit quantum-dimensional effects [9], which leads to the oscillation of the electrical parameters of the films from the thickness. Such oscillations appear at a thickness of d < 300 nm. In the investigated thickness range, these effects can not be taken into account. In addition to weak localization and spin-orbital interaction, the contribution of other corrections related to the effects of inter-electron interaction was not taken into account. Since this contribution is insignificant at the studied temperatures.

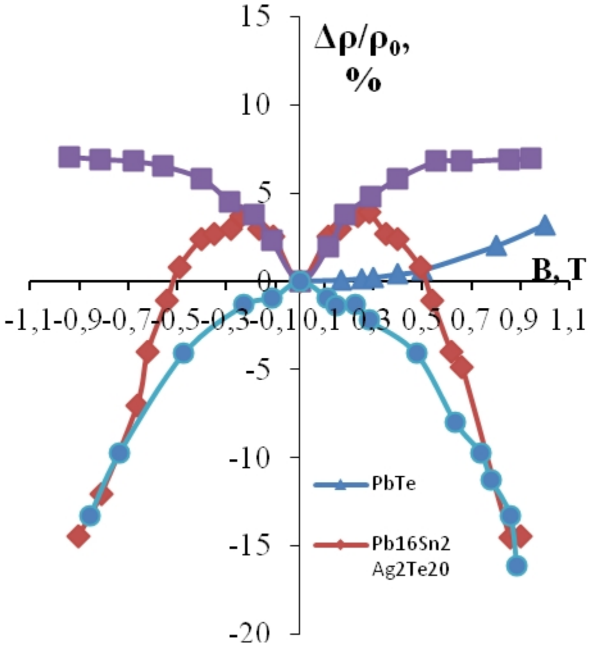


Fig. 1. Experimental dependences of the magnetoresistance in a perpendicular magnetic field at temperature T = 77 K for films of composition: ▲ – PbTe, ■ – $Pb_{14}Sn_4Ag_2Te_{20}$, ♦ – $Pb_{16}Sn_2Ag_2Te_{20}$, ● – $Pb_{18}Ag_2Te_{20}$ on fresh chips (0001) of mica-muscovite.

The concentration and mobility of current carriers in PbSnAgTe films were measured according to Holl measurements and are shown in Table 2. According to these data, the value of the product k_Fl was calculated, where k_F is quasiwave Fermi vector $k_F=(3\pi n)^{1/3}$, l – the free path of current carriers, n is the concentration of current carriers. To be able to apply the theory of weak localization to the electron gas it is necessary to check the Ioffe-Regel condition $k_Fl \gg 1$. As can be seen from Table 1 for all the samples under study, this condition is fulfilled, which allows us to apply the theory of non-ballistic transport for PbSnAgTe films.

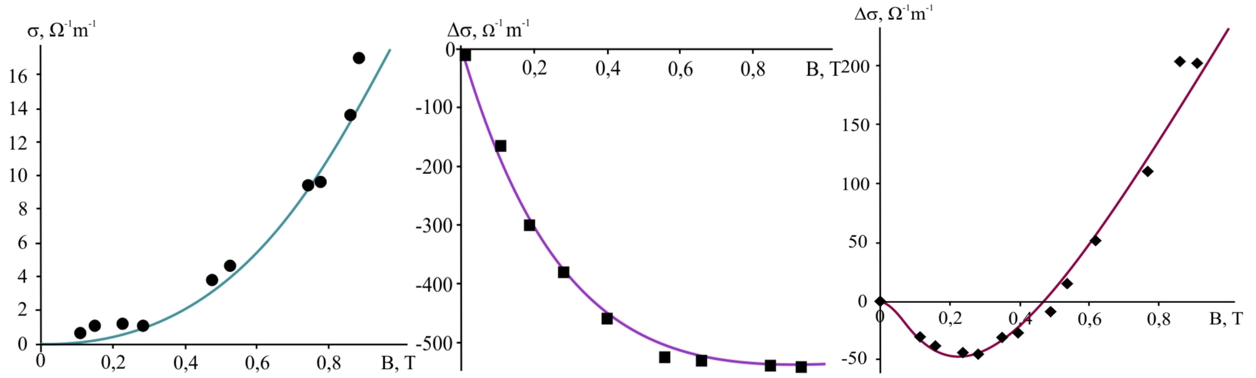
The results of the approximation of experimental dependences are given in Table 2 and in Fig. 2. The diffusion coefficient D was calculated by the formula $D=\frac{mk_BT}{e}$ [10].

For a film of a composition $Pb_{18}Ag_2Te_{20}$ at the themperature 77 K, the magnetic conductivity is positive

Table 1

Electrical properties for PbSnAgTe films on mica at a temperature of T = 77K

Composition	Concentration of current carriers n,p, cm^{-3}	Mobility μ, cm^2/Vs	Diffusion length $L\varphi$, nm	$k_f l$
$Pb_{18}Ag_2Te_{20}$	$n=6.25\cdot10^{16}$	88.5	12.3	12.2
$Pb_{16}Sn_2Ag_2Te_{20}$	$p=5.9\cdot10^{17}$	126.4	98.1	1135.2
$Pb_{14}Sn_4Ag_2Te_{20}$	$p=2.3\cdot10^{18}$	223.4	266.1	7428.6



a) b) c)

Fig. 2. Dependences of the specific magnetic conductivity in a perpendicular magnetic field at a temperature T=77 K for the films of composition: (a), ● – $Pb_{18}Ag_2Te_{20}$,, (b), ♦ – $Pb_{16}Sn_2Ag_2Te_{20}$, (c),■ – $Pb_{14}Sn_4Ag_2Te_{20}$on fresh chips (0001) of mica-muscovite. Points – experiment, lines – calculation according to (8).

and the magnetosphere, respectively, is negative (Fig. 1, Fig. 2, a). The relation between the characteristic times is the following – $t_{so} >> t_j$. The transport of charge carriers is determined mainly by the effect of weak localization, and the effect of spin-orbital scattering is much smaller.

Table 2

Characteristic times: the relaxation time of the phase of the wave function τ_φ and the spin relaxation time τ_{so}, associated with the spin-orbital interaction for the PbSnAgTe films

Composition	Film thickness d, nm	τ_φ, s	τ_{so}, s
T=77K			
$Pb_{18}Ag_2Te_{20}$	540	$2.1\cdot10^{-12}$	$8.86\cdot10^{-11}$
$Pb_{16}Sn_2Ag_2Te_{20}$	810	$9.5\cdot10^{-11}$	$3.85\cdot10^{-11}$
$Pb_{14}Sn_4Ag_2Te_{20}$	540	$3.9\cdot10^{-10}$	$1.35\cdot10^{-11}$

For a film of composition $Pb_{16}Sn_2Ag_2Te_{20}$, the magnetic conductivity has a maximum in the negative region and then, with an increase in the magnetic field, reaches high positive values compared to the $Pb_{16}Sn_2Ag_2Te_{20}$ (Fig. 2, b). The time of spin-orbital interaction for this composition is in one order with the phase relaxation time, but $t_{so} \le t_j$. For a film $Pb_{14}Sn_4Ag_2Te_{20}$, the magnetic conductivity has high negative values with logarithmic saturation at a magnetic field B > 0.6 T (Fig. 2, c). That is, for this sample there is an antilocalisation phenomenon with strong spin-orbital interaction. The time of spin-orbital scattering is an order of magnitude less than the phase relaxation time.

As can be seen from Table 2, with increasing of content of the Sn, the time associated with the spin-orbital interaction τ_{so} decreases. This indicates that the magnitude of the spin-orbital interaction increases proportionately τ_{so}^{-1}. Since the thickness of the films is of the same order, in the systems under study the number of collisions of the electron with impurities increases. Similar results were obtained by the authors in [11] on the base of the study of the spectra of electron paramagnetic resonance in doped silicon. As the concentration of the doping impurity increases, spin-orbital interaction increases with scattering on the impurity. Spin-orbital interaction increases with the growth of the atomic number of the impurity. Time of phase relaxation of the wave function τ_φ, on the contrary, increases by increasing the content of the doping impurity. The introduction of Ag leads to the appearance of effects of weak localization, and the additional doping of Sn increases disordering in the system, which allows observing quantum-mechanical interference effects. The theory of electron scattering due to impurities in the superconductor was developed by Abrikosov and Gorkov [12], which shows that the spin rotation at scattering on the impurity can be due to spin-orbital interaction.

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Conclusions

1. Measurements of magnetoresistance for films based on PbSnAgTe compounds from composition are carried on.
2. The regularities of the change in magnetic conductivity are explained in the framework of the theory of weak localization, taking into account the mechanism of spin-orbit scattering.
3. It is shown that the appearance of the curves of the magnetic-field conductivity or resistance dependence allows us to determine the peculiarities of the transport of carriers in a magnetic field and to obtain information about the presence of spin-orbit interaction.
4. Observed quantum interference effects in PbSnAgTe films can be explained by spin-orbital interaction in scattering on impurities.

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О.Б. Костюк, М.А. Рувінський, Є.В. Івакін, М.Ю. Перегінчук

Квантові ефекти небалістичного транспорту в плівках на основі сполук PbSnAgTe

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На основі теорії слабкої локалізації з врахуванням механізму спін-орбітального розсіювання розглянуто закономірності зміни магнетопровідності плівок PbSnAgTe. Досліджено залежності магнетоопору плівок PbSnAgTe від складу в перпендикулярному до поверхні плівки магнітному полі.

Показано, що для полікристалічних плівок на підкладках зі слюди-мусковіт час спін-орбітальної взаємодії залежить від складу та може змінювати знак магнетоопору.

Ключові слова: розмірні ефекти, тонкі плівки, плюмбум телурид, термоелектричні властивості.