Shubnikov de Haas Oscillations in Bi Wires Doped with Acceptor Impurities

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***Abstract* — Shubnikov de Haas (ShdH) oscillations in single-crystal wires of Bi doped with acceptor impurities of Sn up to 0.3at% with diameters of 100 nm to 5 µm were investigated. The wires obtained by the liquid phase casting were single-crystals of the cylindrical form in glass insulation with the standard $(10\underline{1}1)$ orientation along the wire axis. All investigated samples exhibited ShdH oscillations of charge carriers in L and T points of the Brillouin zone in a temperature range $2.1 < T < 4.2$ K. The period of oscillations is independent of diameter. It is shown, that with an increase in the degree of doping Bi with an acceptor impurity, the Fermi surface of T - holes increases by more than an order of magnitude; L – holes, by 3 times. The cyclotron mass, Dingle temperature and the position of the Fermi level ε_F of T holes during doping were calculated. The results were compared with similar data obtained on bulk samples.**

***Index Terms* — wires in glass insulation, Bi-Sn, Shubnikov de Haas effect, Fermi surface**

I. INTRODUCTION

Bismuth material has unique electronic properties because of a small effective mass of carriers, high anisotropy of the Fermi surface, low concentration of carriers and large mean free path. Single-crystal samples of Bi exhibit strong magnetoresistance effect [1, 2] and clearly defined ShdH oscillations of magnetoresistance.

Small effective mass of the carriers in Bi leads to a large Fermi wavelength λ_F = 50 – 60 nm [3]. The mean free path length l_e in Bi at 4.2 K is more than 1 mm, which is several orders of magnitude greater than in metals [4]. Owing to this, Bi is the most suitable material for the study of quantum and classical size effects.

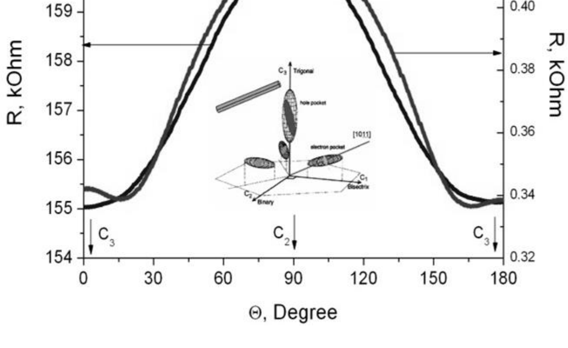
The low value of intrinsic carrier concentration in Bi makes it possible to change it over a wide range through weak doping; this, together with a low density of states in relevant areas, leads to significant changes in the Fermi energy. From a theoretical point of view, this provides an additional opportunity to study the band structure by scanning its Fermi level as well as to implement and investigate the different types of electronic topological transitions. From practical point of view, the doping allows optimizing the thermoelectric parameters of the material.

The specificity of manufacturing of single-crystal wires in a glass envelope i.e. high pulling rates and sharp supercooling in the crystallization zone give the possibility slightly extend the solubility region of the acceptor (Sn, Pb) and donor (Te, Se) impurities in Bi. The introduction of the impurities reduces the relaxation time, which leads a decrease in the amplitude of the ShdH oscillations. Since the amplitude of ShdH oscillations increases with increasing magnetic field and achieves its maximum value near the field of the quantum limit, which is 20-22 T in the case of the T-zone in the wires of pure bismuth, the most promising method for studying the T-zone of conductivity during doping is research in high magnetic fields.

In this study the ShdH oscillations in single-crystal wires of Bi strong by doped with acceptor impurities (Sn) were investigated.

II. SAMPLES AND EXPERIMENTAL METHODS

The wires of pure Bi and bismuth doped with Sn in glass insulation were obtained by liquid phase casting using an improved Ulitovsky-Teylor method [5-7]. The row material used to obtain Sn-doped wires was single-crystal ingots an appropriate composition previously synthesized and homogenized by the method of horizontal zone recrystallization. The uniform distribution of impurities in the wires was achieved due to the extremely high temperature of drops (1100°C) in the melt during stretching of the wires and its intensive mixing by a high-frequency electromagnetic field of the inductor (880 GHz) for a few minutes [5]. Monocrystallinity and crystallographic orientation of the samples were controlled by means of X-ray diffraction (Oxford Excalibur) and angle diagrams of transverse magnetoresistance (fig.1).



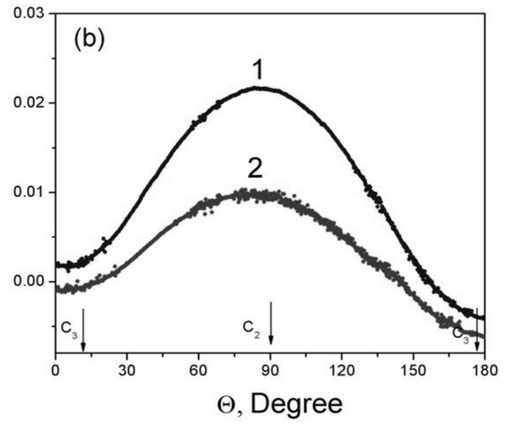


Fig. 1. Angle diagrams of transverse magnetoresistance R(Θ) of Sn doped wires of Bi in magnetic field B = 0.5 T at T = 4.2 K (a): 1. Bi-0.05at%Sn, d = 0,6 μm; 2. Bi-0.1at%Sn, d = 0,5 μm and at T = 300 K (b): 1. Bi-0.2at%Sn, d = 0.4 μm; 2. Bi-0.3at%Sn, d = 0.6 μm. Inset (a): schematic of the Fermi surface of Bi showing the hole pocket along the trigonal axis and three electron pockets in a plane perpendicular to the trigonal axis. Direction $10\underline{1}1$ is indicated.

As the wires of pure Bi, the Sn doped wires have the same crystallographic orientation ($10\underline{1}1$) along the wire axis. At this crystallographic orientation the wire axis is deflected by an angle of 19.5° in the bisector – trigonal plane (fig. 1, inset).

Figure 1 shows angle diagrams of transverse magnetoresistance of the Bi wires with different doping level which indicate the identity of the crystallographic orientation of all compositions of the studied wires. At Θ = 90° the magnetic field is directed along the axis of the second order C_2 ($B \parallel C_2$) (binary axis); at Θ = 0 the direction of the magnetic field coincides with the trigonal axis C_3 ($B \parallel C_3$).

The investigation of the angle diagrams of transverse magnetoresistance made it possible to study the ShdH oscillations in definite directions and get information on the maximum and minimum cross-section of the Fermi surface of the electrons and holes during doping. ShdH oscillations were studied in a temperature range of 2.1 – 4.2 K in magnetic fields up to 14 T both in the longitudinal and transverse magnetic field orientation ($B \parallel I$, $B \perp I$) at the International Laboratory of High Magnetic Field and Low Temperatures (Wroclaw, Poland).

III. RESULTS AND DISCUSSION

The ShdH effect in BiSn alloy wires with the concentration of the impurity holes up to 0.3at% was studied. In the longitudinal magnetic field ($B \parallel I$) ShdH oscillations of charge carriers at L and T points of the Brillouin zone were recorded for all investigated samples. Longitudinal orientation ($B \parallel I$) was quite convenient because the monotonic motion *R(B)* in this direction is weak. Figures 2 and 3 show ShdH oscillations of longitudinal magnetoresistance of the wires with different compositions on *R(B)* (fig. 2) and on *dR/dB(B)* (fig. 3).

It is noteworthy that the negative magnetoresistance effect is a absent in the wires containing more than 0.1at%Sn, that is with increasing impurity of Sn, the galvanomagnetic size effect is suppressed as a result of a decrease in the mean free path of carriers due to impurity scattering. In longitudinal configuration the magnetic field ($B \parallel I$) is oriented along the wire axis which corresponds to a direction close to the bisector axis. The deviation is 19.5° in the bisector – trigonal plane.

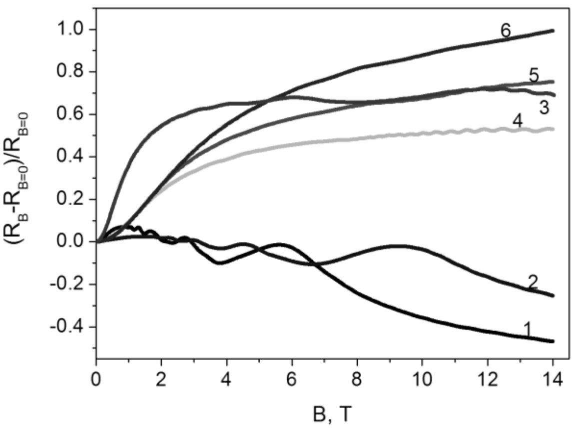


Fig. 2. Field dependences of longitudinal magnetoresistance *ΔR/R(B)* of the Bi-Sn wires with different concentration of Sn at T = 4.2 K: 1. Bi-0.05at%Sn, d = 0,55 μm; 2. Bi-0.07at%Sn, d = 0.3 μm; 3. Bi-0.1at%Sn, d = 0.8 μm; 4. Bi-0.15at%Sn, d = 0.4 μm; 5. Bi-0.2at%Sn, d = 0.2 μm; 6. Bi-0.3at%Sn, d = 1.7 μm.

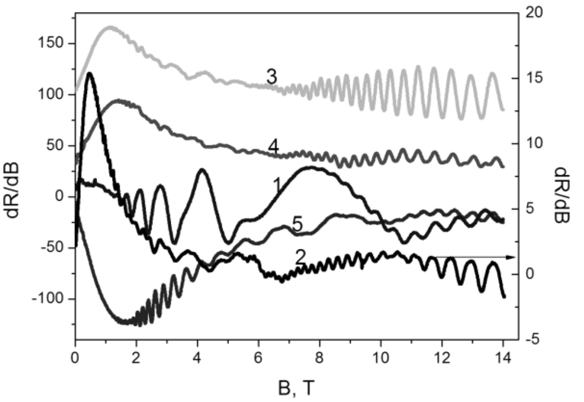


Fig. 3. Field dependences of derivative of longitudinal magnetoresistance *dR/dB(B)* of the Bi-Sn wires with different concentration of Sn at T = 4.2 K: 1. Bi-0.07at%Sn, d = 0.3 μm; 2. Bi-0.1at%Sn, d = 0.8 μm; 3. Bi-0.15at%Sn, d = 0.4 μm; 4. Bi-0.2at%Sn, d = 0.2 μm; 5. Bi-0.3at%Sn, d = 1.7 μm.

The ShdH oscillations are periodic in indirect magnetic field B^{-1}, with a period of $\Delta\left(\frac{1}{B}\right) = \frac{2\pi e}{hcS}$, which is inversely proportional to the extremal cross-section S of the Fermi surface in the plane normal to $\vec{B}$. In the longitudinal geometry with $B \parallel I$ in Bi wires, there are three different extreme cross-sections: one hole ellipsoids S_{Th}, two equivalent electron ellipsoids with a larger size $S_{L_{2,3}}$ and one electron ellipsoid of a smaller size S_{L_1}. In the pure Bi wires, the period of the SdH oscillations from the cross section of the Fermi surface T - holes perpendicular to magnetic field *B* $\Delta_T\left({}^{1}\!/\!_{B}\right)$ = 0,57*10^{-5}Oe^{-1}, $\Delta e_{2,3}\left({}^{1}\!/\!_{B}\right)$ = (3±0,2)*10^{-5}Oe^{-1}, $\Delta e_1\left({}^{1}\!/\!_{B}\right)$ = (7,5±0,5)*10^{-5}Oe^{-1}. We observed ShdH oscillations in all single-crystal Sn doped wires of Bi.

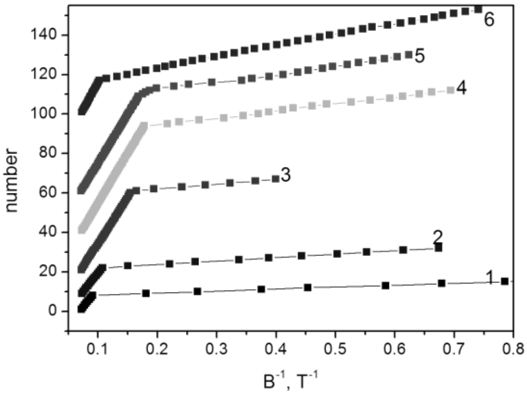


Fig. 4. Dependences of the quantum number n of ShdH oscillations vs. indirect magnetic field B^{-1} of the Bi-Sn wires with different concentration of Sn at T = 4.2 K: 1. Bi-0.07at%Sn, d = 0.3 µm; 2. Bi-0.1at%Sn, d = 0.8 µm; 3. Bi-0.15at%Sn, d =0.4 µm; 4. Bi-0.2at%Sn, d = 0.2 µm; 5. Bi-0.3at%Sn, d = 1.7 µm.

The ellipsoid with the smallest extreme cross-section $S_{L_{e1}}$ does not appreciably contribute to the ShdH oscillations because its lowest Landau level reaches the Fermy level in a very low magnetic field. The two electron ellipsoids with larger extreme cross-section $S_{L_{e2,3}}$ indicate the occurrence of ShdH oscillations in the low field region and shift to higher fields with Sn concentrations (fig. 3).

The T-hole ellipsoid in this direction has a smaller period from ShdH oscillations in a strong magnetic field. As shown in fig. 4 conditional quantum number of ShdH oscillations n, which is used to identify ShdH peaks linearly depends on inverse magnetic field B^{-1}. All curves contain two periods from T-holes $\Delta_1\left({}^{1}\!/\!_{B}\right)$ and $L_{2,3}$ - holes $\Delta_2\left({}^{1}\!/\!_{B}\right)$.

Most pronounced ShdH oscillations on *R(B)* from the holes S_{Th} are observed in the wires with a concentration of Sn 0.1at% and 0.15at%, where the monotonic course in a strong magnetic field is absent (curves 3, 4 in fig. 2). The analysis of the periods of ShdH oscillations has shown that with increasing Sn both periods from T – hole ellipsoid and $L_{2,3}$ – holes ellipsoids decrease, which is representative of an increase in the cross-section S_{Th} and $S_{L_{2,3}}$ during doping Bi wires with acceptor impurities of Sn (fig. 4). However, in the case of doping 0.05-0.3at%Sn the cross-section of T – holes increases by no more than in 1.5 times, while the section of easy holes in L - $S_{L_{2,3}}$ increases in 6 times.

The ShdH oscillations are due to Landau quantization of the cyclotron orbits of the carriers [8]. In an externe magnetic field *B* applied along the Z direction, the electron and hole orbits are quantized into Landau levels with energies of $E=\left(\nu+{}^{1}\!/\!_{2}\right)\hbar\omega_c+{}^{\hbar^2\kappa_z^2}\!/\!_{2m*}$, where m* is the carrier effective mass, $\omega_c={}^{eB}\!/\!_{m^*c}$ is the cyclotron frequency, υ is an integer and $\hbar\kappa_z$ is the component of the momentum parallel to *B*. With increasing magnetic field, the occupancy of each Landau level and the separation between adjacent Landau levels $\hbar\omega_c$ become larger. As a result, the Landau levels below the Fermi level are sequentially driven across the Fermi level. The crossing of the Landau levels is accompanied by abrupt changes in the density of states, which gives rise to oscillations in the resistivity. The ShdH oscillations are most prominent at low temperatures where the Landau levels are sharply defined. At higher temperatures, where $\kappa_B T\ ?\ \hbar\omega_c\approx{}^{\hbar eB}\!/\!_{m^*c}$ the oscillations are masked by thermal oscillations between the Landau levels. The temperature dependence of the amplitude of ShdH oscillations is approximately $:\ \exp\left(-\frac{\kappa_B T}{\hbar\omega_c}\right)$.

From the dependences of the amplitude of ShdH oscillations of the resistance *R(B)* (at 4.2 and 2.1 K) the effective masses of holes in T and electrons were calculated. It was found that m_c^h of holes increased in 2 times in comparison with pure Bi, which indicates the nonparabolicity of spectrum in T point of the Brillouin zone and is consistent with the data obtained for bulk samples of bismuth doped with acceptor impurities [9]. Dingle temperature $T_D={}^{\hbar}\!/\!_{\kappa_B\tau}$, where $\tau=\frac{l}{\upsilon_F}={}^{l\cdot m^*}\!/\!_{\hbar\kappa_F}$ is the carrier relaxation time, was calculated from the field dependences of the amplitude of ShdH oscillations and differed for L and T holes. In strongly doped wires, the Dingle temperature is 6 – 7 K, which is 6 - 7 times more than in the pure wires of Bi.

However, ShdH oscillations are clearly visible against the background of a monotonic dependence R(B) in all crystallographic directions.

This result attests to a high quality of Bi doped wires in glass capillary prepared by liquid phase casting with the length of several dozen meters and paves the way for further quantum transport studies in pure and doped 1-D wires.

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