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Hall effect and quantum oscillations of magnetoresistivity in the topological insulator Bi_2Se_3. The role of bulk and surface carriers

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At temperatures of 5 and 10 K and in magnetic fields up to 9 T, the field dependences of the magnetoresistivity and Hall resistivity of a topological insulator Bi_2Se_3 single crystal were measured. It was shown, that the Hall effect is due to bulk carriers, while the Shubnikov–de Haas oscillations are associated with two-dimensional carriers.

Keywords: Bi_2Se_3 single crystal, crystal growth, Shubnikov–de Haas oscillations, Berry phase, Fermi surface.

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1. Introduction

Topological insulators are assigned to a new class of quantum materials whose production and comprehensive study is of high fundamental and applied interest. Electronic structure of such materials is quite unusual: the inside of a topological insulator is a semiconductor, while its surface is a two-dimensional metal. Such metallic surface states are spin-polarized and protected against scattering. Topological insulators are very promising for thermoelectronic applications [1–4], spintronic devices and quantum computers [5–7] as well as in micro- and nanoelectronics [8,9].

Bi_2Se_3 is classified a topological insulator. On the one hand, its electronic structure is well studied both experimentally and theoretically [10–12]. On the other hand, the existing literature on electronic properties of bismuth selenide not always accurately agree and correlate together: in particular, this is applicable to such properties as current carrier concentration and mobility [13–17]. This may be associated both with different quality of the given crystals and the difference in methods that are used to get information on particular electronic parameters.

This study reports the results of experimental examinations of the Hall resistivity and Shubnikov–de Haas oscillations in the magnetoresistivity of Bi_2Se_3 single-crystal followed by calculation of the charge carrier concentrations using the Hall effect and Shubnikov–de Haas effect data.

2. Experiment

Topological insulator Bi_2Se_3 single-crystals were grown by the Bridgman–Stockbarger method. Bi, Se components were taken in stoichiometric ratio 2 : 3, then these components were mixed and placed in a quartz tube. The tube was evacuated up to 10^{-4} atm and sealed tight. Then the tube was heated up to 850°C and held during 30 h. At the second stage, the prepared components were ground and placed in a quartz tube with elongated sharp tip coated with a graphite on the inside. The tube was evacuated to a residual pressure of $\sim 10^{-4}$ atm, sealed and placed in a furnace with a large temperature gradient of approx. 50 deg/cm. Then the tube was heated up to a temperature about 750° C until the input components were fully molten. The tube was held for 2 h, then slowly placed in a cold zone of the furnace at a rate of ~ 3 mm/h. Single-crystals grown during this process had a cylindrical shape with sharp tip, $\sim 5{-}7$ mm in diameter and $\sim 10{-}20$ mm in length. Resistance ratio of the synthesized crystals RRR $(\rho_{300\,K}/\rho_{5\,K}) \approx 4.8$, which is comparable with the known literature data [18,19]. For magnetotransport measurements, a sample was prepared by chipping the whole single-crystal, the sample dimensions were equal to $0.35 \times 1.3 \times 3.22$ mm.

Figure 1 shows the fragment of diffraction pattern recorded on the Bi_2Se_3 surface and the detail shows single-crystal photo. It can be seen that all peaks may have $(00l)$ index, whence it follows that the single-crystal surface coincides with $(00l)$ type plane.

Magnetoresistance ρ_{xx} and Hall resistivity ρ_{xy} were measured using a 4-point scheme at the Collaborative Access Center of the Institute of Metal Physics, Ural Branch, Russian Academy of Sciences, on PPMS-9 physical measurement system at 5 and 10 K and magnetic fields up to 9 T.

3. Results and discussion

Figure 2 shows field dependences of the Hall resistivity ρ_{xy} of Bi_2Se_3 single-crystal at 5 and 10 K. It is shown that ρ_{xy} has a negative value and depends linearly on magnetic

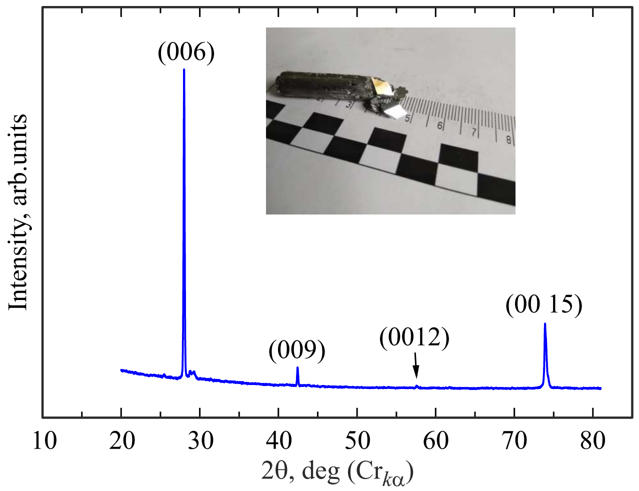

Figure 1. The fragment of natural chip diffraction pattern of Bi_2Se_3 single-crystal. The detail shows the photo of Bi_2Se_3 single-crystal and its chip.

field. This means that electrons are the main type of current carriers. In accordance with the one-band model, their concentration may be estimated as follows

$$n_{\mathrm{Hall}} = 1/(eR_{\mathrm{H}}), \tag{1}$$

where $R_{\mathrm{H}} = \rho_{xy}/B$ is the Hall coefficient, e is the electron charge. The calculated values of n_{Hall} are listed in the table which shows that the carrier concentration decreases slightly with temperature and this agrees with the data in [12,18,20].

Figure 3 shows the field dependences of the magnetoresistivity of Bi_2Se_3 at 5 and 10 K. At both temperatures, magnetoresistivity smoothly increases as the magnetic field grows, and the temperature rise results in the growth of ρ_{xx}.

The Shubnikov–de Haas oscillation analysis is another method to determine electronic structure and, in particular, to estimate current carrier concentration. Figure 3 shows that magnetoresistivity oscillations are observed in fields higher than 7 T. According to the Onsager quantization rule, the extreme cross-section area of the Fermi surface A_{F} is related to the oscillation frequency F as follows

$$F = A_{\mathrm{F}}\hbar/(2\pi e), \tag{2}$$

where $\hbar$ is Planck's constant. Figures 4, *a* and *b* show oscillating parts of the magnetoresistivity calculated by subtracting the monotone polynomial parts from the dependences in Figure 3. To define the oscillation frequency the fast Fourier transform was plotted and is shown in Figure 4, *c* and *d*. Note that at both temperatures single frequencies as listed in the table correspond to these oscillations. Using F and A_{F}, the wave vector on the Fermi surface k_{F} and the bulk electron concentration $n^{\mathrm{3D}}_{\mathrm{SdH}}$ may be calculated as follows

$$k_{\mathrm{F}} = \sqrt{A_{\mathrm{F}}/\pi}, \tag{3}$$

$$n^{\mathrm{3D}}_{\mathrm{SdH}} = \frac{1}{3}\,k_{\mathrm{F}}^3/\pi^2. \tag{4}$$

The calculated electronic structure parameters are listed in the table.

It shown that the concentrations calculated from the Hall effects and Shubnikov–de Haas oscillations differ by more than twice. Such difference may be in various types of current carriers that contribute to the Hall effect and Shubnikov–de Haas effect. Thus, topological insulators are known to have on their surface massless Dirac fermions with linear dispersion law. The Lifshitz–Onsager quantization rule is the most widely known method of determining

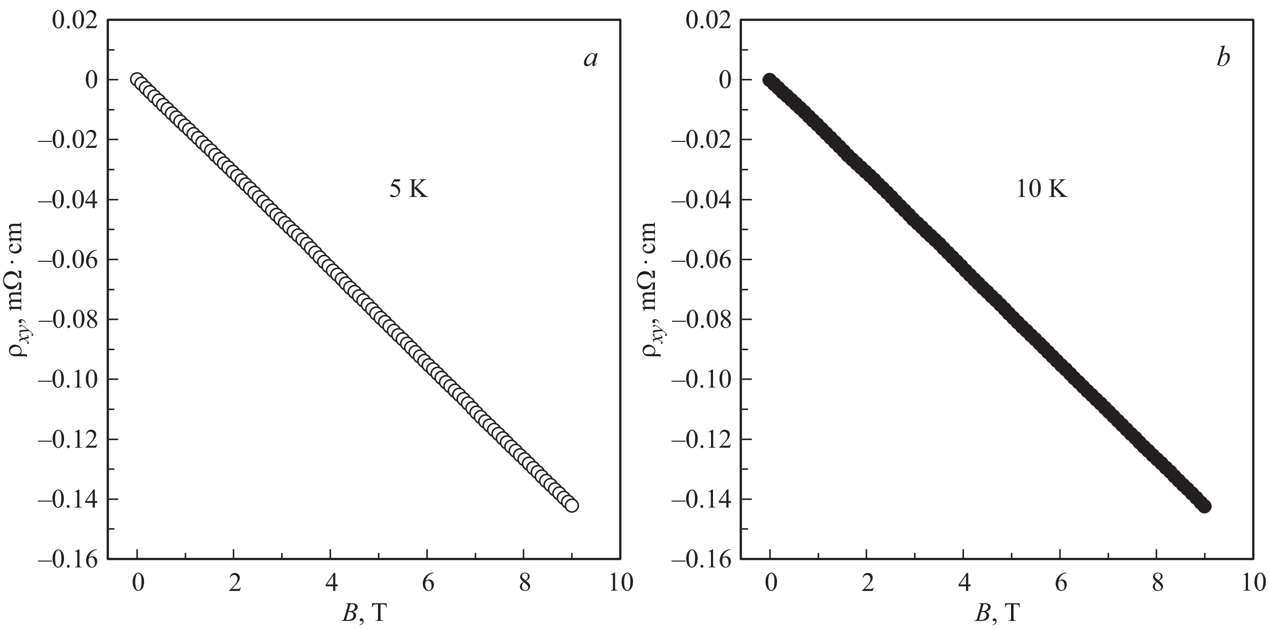

Figure 2. Field dependences of the Hall resistivity of Bi_2Se_3single-crystal at *a*) 5 and *b*) 10 K.

Electronic structure parameters of Bi_2Se_3 single-crystal at 5 and 10 K

Temperature	n_{Hall}, 10^{19} cm^{-3}	n_{SdH}^{3D}, 10^{19} cm^{-3}	F, T	A_F, Å^{-2}	k_F, Å^{-1}	n_{SdH}^{2D}, 10^{12} cm^{-2}	d, nm
5 K	3.98	1.97	230	0.02196	0.0836	5.56	1.4
10 K	3.96	1.91	225	0.02148	0.0827	5.44	1.37

Note. n_{Hall} is the current carrier concentration determined from the Hall effect, n_{SdH}^{3D} is the bulk carrier concentration determined from the Shubnikov–de Haas effect, F is the oscillation frequency, A_F is the extreme cross-section area of the Fermi surface, k_F is the wave vector on the Fermi surface, n_{SdH}^{2D} is the two-dimensional carrier concentration determined from the Shubnikov–de Haas oscillations, d is the two-dimensional layer thickness.

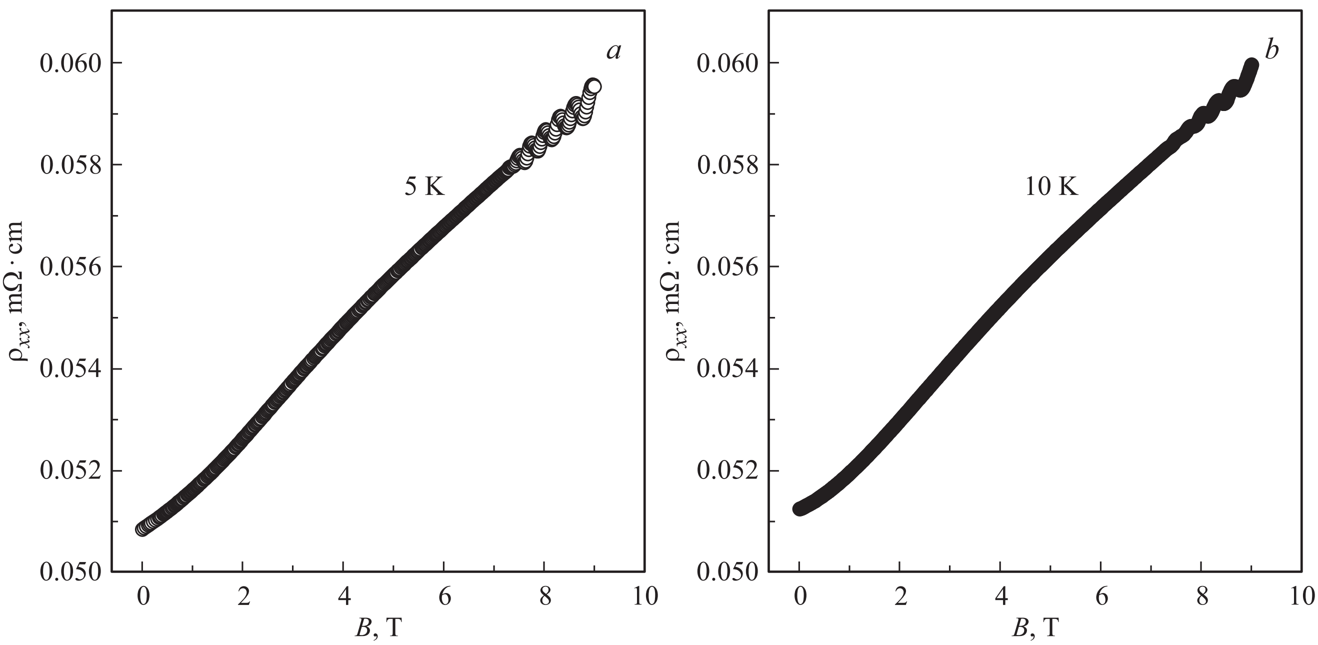

Figure 3. Field dependences of the magnetoresistivity of Bi_2Se_3 single-crystal at a) 5 and b) 10 K.

the Dirac fermions from oscillations:

$$n = F/B + \Delta, \quad (5)$$

where n is the integer index of the Landau level corresponding to the valley on the oscillating part of the magnetoresistivity, $n + 1/2$ is the semi-integral index corresponding to the peak, $\Delta = \beta + \delta$, β is the Berry phase factor corresponding to the Berry phase, δ is the phase factor depending on the Fermi surface geometry. When the system has Dirac fermions, then $\beta = 1/2$ [21]. From (5) it follows that the point of intersection of the straight line $n = f(1/B)$ with the y-axis gives the phase factor Δ. Figure 5 shows $n = f(1/B)$ curves and the corresponding values of Δ are equal to -0.26 and -0.47 for 5 and 10 K, respectively. Also note that frequencies derived from $n = f(1/B)$ were equal to 230.67 and 224.26 T, that matches well the values calculated using the fast Fourier transform (see the table).

It is well known that δ may be equal to: 1) zero: $\delta = 0$ for the two-dimensional Fermi surface, and 2) $\delta = \pm 1/8$ for the three-dimensional surface. Here, sign „+" is used for electrons and sign „–" is used for holes [22–24]. In our case, sign „+" is used for the three-dimensional Fermi surface because electrons are the main type of carriers as follows from the Hall effect. According our data, Δ is equal to -0.26 and -0.47 for 5 and 10 K, respectively. Assuming that $\delta = 0$, then $\Delta = \beta$. But if $\delta = \pm 1/8$, then $\beta = -0.385$ and -0.695 for 5 and 10 K, respectively. Hence, the nontrivial Berry phase takes place in both cases that is one of the arguments in favor of the fact that these oscillations are caused by two-dimensional states.

The difference of β on the ideal value $|\beta| = 1/2$ may be explained by nonideality of the Dirac dispersion law. Thus, theoretical study [25] shows that if the dispersion law has a quadratic contribution besides the linear contribution, then β differs from the ideal value. Such nonideal values of β were observed for Bi_2Se_3 and Bi_2Te_3, for example, in [15,17,26].

Other experimental studies [13,27,28] showed that in pure topological insulators Bi_2Se_3 and in Cu-doped Bi_2Se_3 with high carrier concentrations $n > 3 \cdot 10^{19}$ cm^{-3}, oscillations were observed only up to certain values of θ between the magnetic field direction and crystallographic axis c perpendicular to the sample plane and the dependences

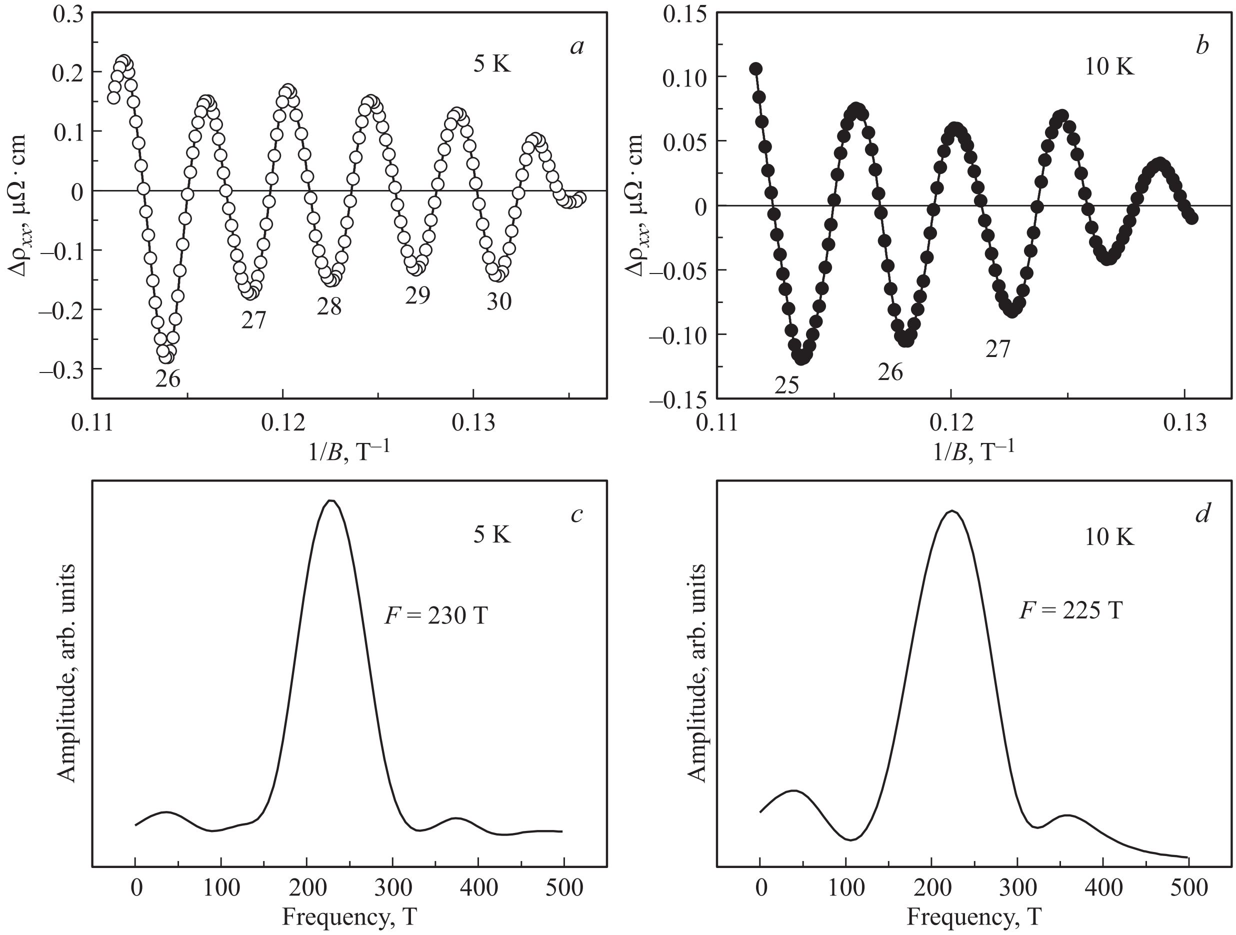

Figure 4. Oscillating parts of the magnetoresistivity of Bi_2Se_3single-crystal at *a*) 5 and *b*) 10 K. Fast Fourier transforms of the oscillating parts of magnetoresistivity at *c*) 5 and *d*) 10 K.

of oscillation frequencies were written as $F \propto 1/\cos\theta$. This indicated that these oscillations are caused by two-dimensional states, while for three-dimensional states, oscillations shall be observed up to 90°. In particular, in [13] this is associates with large number of two-dimensional channels in Bi_2Se_3. Besides the angular measurements of oscillation frequency, in [16] ,k_F for Bi_2Se_3 with $n = 5.6 \cdot 10^{19}\,cm^{-3}$ was measured using the angle-resolved photoelectron spectroscopy (ARPES) and, as emphasized by the authors, agrees well with k_F determined in their study [16] from oscillations, which is also a good argument (in addition to $F \propto 1/\cos\theta$) in favor of the fact that the oscillations are caused by two-dimensional state. It shall be also noted that the studies on observation over oscillations caused by two-dimensional states in topological insulators with high carrier concentrations ($n > 3 \cdot 10^{19}\,cm^{-3}$) show that the Fourier transform exhibits quite high frequency $F > 150$ T [13,16,27–29], while in studies on observation over oscillations in Bi_2Se_3 with carrier concentration $\sim 10^{17}–10^{19}\,cm^{-3}$ oscillations have much lower frequencies $F < 100$ T due to three-dimensional ellipsoidal Fermi surface [30–32].

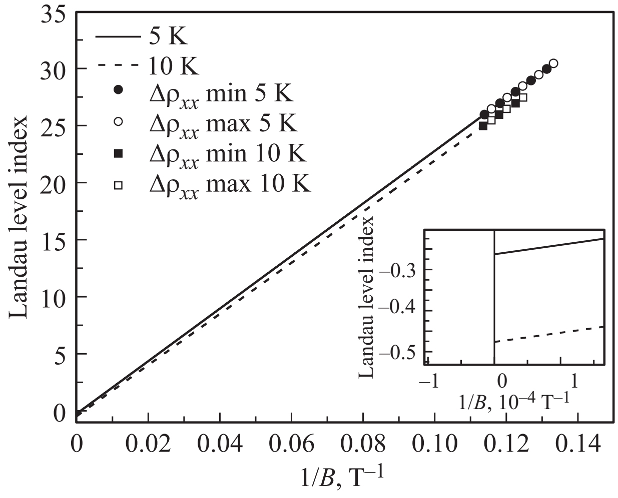

Figure 5. Dependences of the Landau level index on the reverse magnetic field $n = f(1/B)$ at 5 K (circles) and 10 K (boxes). The detail shows straight line segments near zero.

The data obtained herein shows that a nontrivial Berry phase was observed. The oscillation frequencies are quite high ($F > 200$ T), as well as current carrier concentrations are relatively high ($n > 3.9 \cdot 10^{19}\,cm^{-3}$). Note also that values of k_F agree well herein (table) with values of k_F for the Dirac states obtained using ARPES in [16] ($k_F = 0.084\,Å^{-1}$) and [11] ($k_F = 0.08\,Å^{-1}$). The foregoing suggests that the observed oscillations in the magnetoresistivity (Figure 2) are caused by the Dirac fermions with the 2D Fermi surface, while the main contribution to the Hall effect is made by the 3D carriers. This may explain the observed difference in concentrations determined from the Hall effect and Shubnikov–de Haas effect. Anyway, the 2D carrier concentration may be estimated as follows

$$n_{\mathrm{SdH}}^{\mathrm{2D}} = k_F^2/(4\pi). \qquad (6)$$

In addition, using expression

$$d = n_{\mathrm{SdH}}^{\mathrm{2D}}/n_{\mathrm{Hall}}, \qquad (7)$$

the 2D conducting layer thickness may be also estimated. The calculated values of $n_{\mathrm{SdH}}^{\mathrm{2D}}$ and d for 5 and 10 K are listed in the table. Note that the calculated values agree quite well with [13], where $n_{\mathrm{SdH}}^{\mathrm{2D}} = 7.8 \cdot 10^{12}\,\mathrm{cm}^{-2}$, $d = 1.7\,\mathrm{nm}$ and $n_{\mathrm{Hall}} = 4.7 \cdot 10^{19}\,\mathrm{cm}^{-3}$.

4. Conclusion

The study of the magnetoresistivity and Hall effect of topological insulator Bi_2Se_3 single-crystal with resistance ratio RRR ≈ 4.8 detected the Shubnikov–de Haas oscillations both at relatively low (5 K) and higher (10 K) temperatures. The current carrier concentrations and electronic structure parameters were estimated from the Hall effect (n_{Hall}) and Shubnikov–de Haas effect ($n_{\mathrm{SdH}}^{\mathrm{2D}}$) data. It is shown that the value of n_{Hall} is more than twice higher than $n_{\mathrm{SdH}}^{\mathrm{3D}}$. The analysis of the obtained data suggest that the observed difference is due to the fact that oscillations are related to 2D current carriers, while the main contribution to the Hall effect is made by 3D carriers.

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Conflict of interest

The authors declare that they have no conflict of interest.

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