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Transport properties of Cu-doped bismuth selenide single crystals at high magnetic fields up to 60 Tesla: Shubnikov–de Haas oscillations and π-Berry phase

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ABSTRACT

We report Shubnikov-de Haas (SdH) and Hall oscillations in Cu-doped high quality bismuth selenide single crystals. To increase the accuracy of Berry phase determination by means of the of the SdH oscillations phase analysis we present a study of n-type samples with bulk carrier density $n \sim 10^{19} - 10^{20}\ \mathrm{cm}^{-3}$ at high magnetic field up to 60 Tesla. In particular, Landau level fan diagram starting from the value of the Landau index $N = 4$ was plotted. Thus, from our data we found π-Berry phase that directly indicates the Dirac nature of the carriers in three-dimensional topological insulator (3D TI) based on Cu-doped bismuth selenide. We argued that in our samples the magnetotransport is determined by a general group of carriers that exhibit quasi-two-dimensional (2D) behaviour and are characterized by topological π-Berry phase. Along with the main contribution to the conductivity the presence of a small group of bulk carriers was registered. For 3D-pocket Berry phase was identified as zero, which is a characteristic of trivial metallic states.

1. Introduction

The discovery of a class of three-dimensional topological insulators (3D TIs) have generated a lot of excitement in condensed matter physics [1,2]. Due to the strong spin-orbit interaction on the surface of 3D TI there is a special state of two-dimensional (2D) topological metal originated from the spin-polarized Dirac fermions with linear energy dispersion. In momentum space the surface electron modes are arranged in a band consists of odd number of Dirac cone with a singular (Dirac) point. In addition to fundamental interest in new topologically non-trivial states and quasiparticles, unusual properties of 3D TIs induct them as primary candidates to eventually realize a new generation of spintronic devices and quantum computing.

The theory predicts that in TIs when electron makes circular motion around the Dirac point in the momentum space the wavefunction acquires the geometrical π-Berry phase ϕ_B (for example, [3]). Therefore, the π-Berry phase provides a key evidence for the presence of Dirac of the nature of carriers in 3D TIs. An experimental value of the Berry phase can be obtained directly from magnetotransport measurements by analyzing the phase of the Shubnikov-de Haas (SdH) oscillations. In SdH effect, when the Landau levels (LLs) are formed at strong magnetic fields, the Onsager semiclassical quantization condition should be realized [4]. Therefore the oscillating part of the longitudinal conductivity $\Delta\sigma_{xx}$ in 2D system can be expressed as:

$$\Delta\sigma_{xx} \sim \cos 2\pi\left[\frac{B_F}{B_N} + \frac{1}{2} - \beta\right], \qquad (1)$$

where B_F – frequency of the SdH oscillations; B_N – magnetic field position of $\Delta\sigma_{xx}$ extreme, corresponding to Nth LL; $\beta = \phi_B/2\pi$ – phase parameter of the SdH oscillations (Berry phase). According to the [3] in a topologically trivial band with a parabolic dispersion, ϕ_B is zero and therefore $\beta = 0$; whereas, in a Dirac electronic system, $\beta = 1/2$ due to a topologically non-trivial π-Berry phase.

Usually, the phase parameter of the SdH oscillations β can be extracted from so-called Landau level fan diagram, which is a plot of inverse "index" magnetic field $1/B_N$ as a function of LL number N (N is determined as the nearest integer to F/B_N relation). Then one can find β from the cutoff on the abscissa axis by extrapolating $1/B_N$ to 0. To exclude mistakes in identifying β there are several rules for plotting the fan diagrams. First, LL number N always corresponds to the magnetic field B_N in position of $\Delta\sigma_{xx}$ minima (the case when the Fermi energy lies between neighboring Nth and $(N+1)$ th LLs,); whereas, index $N + 1/2$

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is attributable to position of $\Delta\sigma_{xx}$ maxima (the case when the Fermi level is at center of *N*th LL). Second, for semiconductors with an anisotropic dispersion law it is necessary to calculate the conductivity in accordance with the formula $\sigma_{xx} = \rho_{xx}/(\rho_{xx}^2 + \rho_{xy}^2)$ (where ρ_{xx}, ρ_{xy} – components of the resistivity tensor), especially in case when $\rho_{xx} \sim \rho_{xy}$. It is also possible to extract phase parameter β from the Hall conductivity $\Delta\sigma_{xy}$ obtained from the relation $\sigma_{xy} = -\rho_{xy}/(\rho_{xx}^2 + \rho_{xy}^2)$ [5]. However, it is necessary to take into account the phase shift $\pi/4$ between Hall and SdH oscillations.

Among the investigated 3D TIs bismuth selenide Bi_2Se_3 with layered crystal structure is of great interest. Firstly, Bi_2Se_3 with relatively large energy gap (0.3 eV) and simple surface state spectra seems to be viable for room temperature applications [1]. Second, Bi_2Se_3 attracted particular attention, because the intercalating with different metals (for example, Cu, Sr or Nb) between quintuple layers in crystal structure induces superconductivity below $\simeq$ 4 K [6–8]. In practice, in stoichiometric Bi_2Se_3, due to the large quantity of Se-vacancies, the Fermi level crosses the conduction band of a degenerate semiconductor, hence, no insulating properties are observed in the bulk. In addition Cu doping increases bulk carrier density up to $\sim 10^{20}$ cm^{-3}. Thereby, resolving the conductance of the non-trivial surface states from the bulk contribution has been a great challenge for studying the transport and magnetotransport properties of this 3D TI. Also the question about Berry phase in Bi_2Se_3 compound is still unclear. For example, in Ref. [9] it was reported about impossibility of identifying Berry phase from magnetotransport data obtained for Bi_2Se_3 micro flake, which is due to low accuracy in determining the position of the oscillation extremes. Arguably, until recently there was no unambiguous experimental data on the Berry phase in 3D TIs. Nevertheless, some attempts were made to explain the disagreement between the experimental and the theoretical values. For instance, it was shown that in real 3D TI the dispersion is not strictly linear, it contains a parabolic component, that can affect an experimental value of the Berry phase [10]. Another main focus was the influence of the Zeeman effect on phase of SdH oscillations at high magnetic fields. As a result, in several papers it was shown that LL fan diagram had a nonlinear behaviour (for example, [11]). On the other hand it was reported that the disagreement of the theoretical value of the SdH oscillations phase can be explained by the limitation of the semiclassical approach for large values of the LL indices [12].

In our recent work [13] we have investigated heavily doped $Bi_2Se_3Cu_x$ single crystals at magnetic field up to 20 T. Despite the high bulk carrier density (up to $n \sim 10^{20}$ cm^{-3}) we have confirmed the 2D nature of the oscillations. It has been suggested that SdH oscillations in our crystals can be related to several parallel conducting channels as in undoped Bi_2Se_3 [14]. We also observed the coexistence of 2D and 3D contributions to the conductivity, as a result, two types of oscillations with different frequencies were detected. Although obtained values $\phi_B = 2\pi\ (0.45 \div 0.7)$ were close to theoretical ones for Dirac fermions, observed quantum oscillations in fields up to 20 T had sufficiently large filling factor (from $N = 17$). Therefore, in this paper, by means of magnetotransport measurements at high magnetic fields up to 60 T we give a careful study of Berry phase ϕ_B behaviour in high-quality $Bi_2Se_3Cu_x$ single crystals. We also provide a detailed and accurate analysis of data from our recent work [13] to confirm the Dirac nature of carriers in our samples.

2. Materials and methods

High quality Cu-doped bismuth selenide ($Bi_2Se_3Cu_x$) single crystals were grown using modified Bridgman method. X-ray diffraction spectra (not shown) are consistent with previous reports of our single crystals [15,16]. For transport measurements, selected bulk single crystals with mirror-like surfaces with typical in-plane dimensions of (1.5 × 1) mm^2 were used. The thickness of the samples was in a range of 30 – 50 μm. The n-type samples exhibit metallic behaviour ($d\rho/dT > 0$) at temperatures down to ≃20 K with saturation at lower temperatures. Residual resistance ratio [$RRR \equiv R(300\ K)/R(4\ K)$] was 3 – 5 [16]. We extract bulk carrier density of our investigated samples from linear part of Hall resistivity curves at low temperatures, that was in the range of $10^{19} - 10^{20}$ cm^{-3}. The measurements up to 20 T were carried out at magnetic fields of the superconducting solenoid in Shared Facility Center for Studies of HTS and other Strongly Correlated Materials, Lebedev Physical Institute. High field experiments were performed in pulsed magnetic fields up to 60 T with the pulse duration of 150 ms. The facilities at the Dresden High Magnetic Field Laboratory were used. Magnetic field dependencies of longitudinal R_{xx} and Hall resistance R_{xy} were studied. In each measurement, the magnetic field was varied between "–" and "+" in order to subtract the contribution of R_{xy} to R_{xx} and vice versa. Experimental technique and crystal growth conditions are presented in [13] in detail.

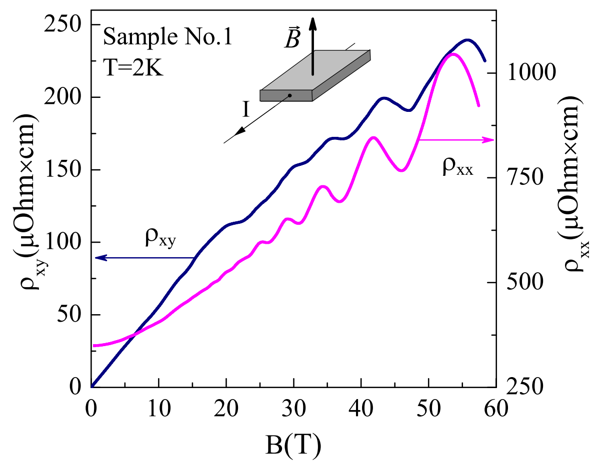

Fig. 1. Longitudinal resistivity as a function of magnetic field and Hall effect of the Cu-doped Bi_2Se_3 sample No.1. Signals $\rho_{xx}(B)$ and $\rho_{xy}(B)$ are plotted against magnetic field up to 60 T at $T = 2$ K. The slope of the $\rho_{xy}(B)$ curve is used to determine the Hall carrier density 1.1×10^{20} cm^{-3}.

3. Results and discussion

Fig. 1 shows field dependencies of longitudinal resistivity $\rho_{xx}(B)$ and Hall resistivity $\rho_{xy}(B)$ of a $Bi_2Se_3Cu_x$ sample No.1. As a result of the LLs formation at applied perpendicular magnetic fields up to 60 T and temperature $T = 2$ K, SdH and Hall oscillations were observed. From linear low-field part of the $\rho_{xy}(B)$ dependence we extract Hall carrier density $n_{Hall} = -1.1 \times 10^{20}$ cm^{-3}. The configuration of magnetic field *B* and electrical current *I* is depicted in the upper inset to Fig. 1.

Before we dive into an analysis of the phase parameter of SdH oscillations, we obtain longitudinal magnetoconductivity values σ_{xx} from $\rho_{xx}(B)$ and $\rho_{xy}(B)$ dependencies according to the formula $\sigma_{xx} = \rho_{xx}/(\rho_{xx}^2 + \rho_{xy}^2)$. For better resolving of SdH oscillations we plot an oscillatory component $\Delta\sigma_{xx}$ after subtraction of the smooth background, deduced by fitting a 4th-order polynomial, against inverse perpendicular magnetic field $1/B$ (Fig. 2a). Fourier analysis of the $\Delta\sigma_{xx}$ dependence revealed two frequencies, which indicates the existence of two groups of carriers. Moreover, oscillations with frequency $B_F^1 = 190$ T were not observed at applied parallel magnetic field, which points to their possible 2D behaviour. Oscillations of the second type with $B_F^2 = 28$ T are uniquely related to quantization of the bulk ellipsoidal Fermi surface, since they are also registered in parallel magnetic field. According to Lifshitz-Onsager relation [4] for spin-filtering states 2D carrier density n_{2D} can be determined as $n_{2D} = 2eB_F/h = 9.5 \times 10^{12}$ cm^{-2}, where e – electron charge and h – Planck constant. Thus, in this sample we observe the presence of 2D-layered transport due to the formation of quasi-2D conducting channels in bulk crystal as in our previous study [13].