Shubnikov de Haas quantum oscillation of the surface states in the metallic Bismuth Telluride sheets*

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Abstract. Metallic Bi2Te3 crystalline sheets with the room-temperature resistivity of above 10 mΩ cm were prepared and their magnetoresistive transport was measured in a field of up to 9 T. The Shubnikov de Haas oscillations were identified from the secondly-derived magnetoresistance curves. While changing the angle between the field and normal axis of the sheets, we find that the oscillation periods present a cosine dependence on the angle. This indicates a two-dimensional transport due to the surface state. The work reveals a resolvable surface contribution to the overall conduction even in a metallic topological insulator.

1 Introduction

Three-dimensional topological insulators (TIs) has been acquired great attention in recent years because of its novel electronic structure, which features a band insulator in its bulk and a metallic gapless surface state (SS) [1–6]. Such an electronic configuration has been verified by mapping the electronic structure using angle-resolved photoemission spectroscopy (ARPES) [7–10]. Spin helicity has also been demonstrated for the Dirac-type SS [11], which suppresses the impurity-induced back-scattering and leads to a dissipationless carrier transport. Based on such topological SS, the Majorana fermion, dissipationless spintronic devices and even quantum spin hall effect are investigated. Bi2Te3 and its related thermoelectric family are such TIs. Due to the strong spin-orbit interactions, the electronic bands from Bi and Te (or Se) atoms cross and construct a topology-nontrivial junction near the Γ point in the k space [1]. The interfacial evolution between a TI material and a normal insulator (e.g. vacuum) requires closing the band gap, therefore the upper conduction band and the bottom valance band kiss through an electronic gap. This results in a band insulator and a metallic SS [12]. Upon a strong magnetic field (B), Landau levels are formed within the Dirac cone. The change of B manipulates the occupation of the landau levels and leads to the SDH oscillation of the overall conduction. It is periodic over 1/B. The unique character of the SS’s SDH oscillation is its 2D behavior, where the 1/B period of the SDH oscillation solely depends on the normal component of the TI sheets. Most data on the topological SS conduction is mostly observed in bulk-insulating samples [18,29,30].

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The SDH oscillations can also be found in some well-crystalline metallic samples, but they mostly exhibit three-dimensional behavior. It is proposed that the 2D transport from the topological SS is screened out and the surface carriers might be scattered by the bulk carriers in the metallic samples [18,30]. Here, we report such evidence of resolvable SS transport in metallic TI samples, where the metallic Bi$_2$Te$_3$ crystals are prepared and the SS transport is identified by the two-dimensional SDH oscillations [31,32].

2 Experimental

Single crystals of Bi$_2$Te$_3$ were prepared by melting high-purity Bi and Tellurium powders (5N, all from Alfa Aesar) and annealing in sealed quartz ampoules. Five batches of crystals were prepared with varying Bi/Te ratios and heating conditions to optimize the carrier concentrations and mobility. The samples were then obtained by mechanical exfoliation with a blade, which dimensions are normally tens of millimeters. The thickness of the samples is as small as a few micrometers. Such flakes are crystalline sheets. As shown in the inset of Figure 1c, six-probe contacts were made by depositing the room temperature-cured silver paste on the 1 mm $\times$ 1 mm regions. The measurements of electronic transport between 10 and 300 K were then conducted using a commercial cryostat with a Keithley 4200 semiconductor analyzer. A typical measurement current is 0.1 mA. The magnetoresistance (MR) measurements at 5 K were performed in a Quantum Design Physical Property Measurement System. The Hall response was measured at the field of 1 T.

3 Results

In Figure 1a, we show a typical temperature profile of the furnace temperature during the preparation procedure. The temperature was rapidly increased to 1000 °C and stirred for a few hours. Keeping the stirring, the temperature was decreased to 500 °C to allow the crystalline nucleation in 5 days. A 5 day’s annealing from 500 °C to 420 °C finalized the preparation before dropping down to room temperature. The X-ray powder diffraction demonstrates its genuine crystal-line condition of No. 166 space group as referred to the PDF card 820358. The resultant crystals can be easily cleaved to the sheets with the dimensions of a centimeter as shown in Figure 1b. We carried out transport measurements to give the resistivity and Hall constants in our lab. A typical room-temperature resistivity distribution in a crystalline crump is shown in Figure 1b. We can see a few sheets near the wall of the glass sample are of some low resistivity of a few mΩ cm, while the highest resistivity of 32.3 mΩ cm (sample-I) appears in the center of the crump. The samples selected in this study
are located between them with a medium resistivity of around 10 mΩ cm (sample-M). The typical temperature dependence of the longitudinal electrical resistivity and carrier density is shown in Figure 1c. Sample-I is of negative temperature dependence. This shows that we can obtain the samples with an open bandgap (sample-I) for the study of ideal 3DTIs. Its low-temperature resistivity reaches 0.1 Ω cm, which is a satisfactory value in current TI research. Considering the reported values of the mobility and carrier density of the SS, one might expect a good SS contribution in such samples [29]. The resistivity of the sample-M presents positive temperature dependence with a metallic behavior and falls to around 1 mΩ cm. The SS contribution therefore becomes minor. We can also see the carrier density of the samples in Figure 1c, which is around $10^{18}$ cm$^{-3}$ and similar to the reported values. The mobility of the bulk carriers can also be calculated from the Hall measurement, which is around 2500 cm$^2$/Vs. We here successfully resolve the two-dimensional SDH oscillations from the SS in such samples as shown below.

Figure 2a shows a typical MR curve of sample-M. The resistivity increases by 8 times while increasing the field $B$ from 0 to 9 T. Interestingly to note, the MR measured with $B$||c axis displays a nearly linear increase versus $B$. Such linearity that the previous MRs of metallic Bi$_2$Te$_3$ did not show has been highlighted as a signature of the anomalous SS transport [18]. It has also been attributed to the competition and compensation between electron and hole bulk bands dominated in Bi$_2$Te$_3$, which is forming an interesting topic on its own and being still a matter of debate. No signature of quantum SDH oscillations can be obviously seen. However, when we check the secondly differentiated MR curve in Figure 2b, the SDH oscillations appears. A tiny oscillation is seen in the low-field range. Such an oscillation seems to exhibit a period of 0.25 T even if we tilt the samples. It is recently related to the Sondheimer oscillation [31]. Another stronger oscillation rises from 2 T. Its amplitude and beat keep growing while increasing the magnetic field, as seen in the typical SDH oscillations. The Landau levels can be formed at around 2 T, by which we expect a mobility of 5000 cm$^2$/Vs of the conducting electrons.

Figure 3a shows the angle-dependent SDH analysis, which demonstrates the 2D behavior of the SDH oscillations. We plot the secondly derived MR values versus $1/B$ and then find uniform periods. When the field is normal to the sheet ($\theta = 0$), the period is 0.054 T$^{-1}$. It decreases to nearly 0.03 T$^{-1}$ when the sample is tilted by 45°($\theta = 45°$) and continuously decreases during the sample tilting as shown in Figure 3a. The periodicity can be confirmed by the Fourier-transformation as shown in the inset of Figure 3b. A dominant period appears in the Fourier-transformed MR curves, in which the dominant frequency increases while $\theta$ increases to 45°. The clear 2D signature is demonstrated by multiplying $B$ with cos $\theta$ as shown in Figure 3b. The dominant frequency of the SDH oscillation increases with the angle initially, while all the oscillation frequencies coincide with each other within the measurement uncertainties after we transformed $B$ to the normal component of the magnetic field. This is clear evidence on the 2D transport due to the topological SS, as demonstrated by Qu et al. [18]. Reasonably, the contribution from the metallic bulk becomes dominant while $\theta$ grows large, over 45°. Please see the SDH oscillations and its FFT curve at 67.5° in the inset of Figure 3b, which shows two peaks. The weaker one shown by the blue arrows is attributed to the SS. The other peak belongs to the bulk, which persists within the measurement uncertainties and is shown by the red dashed lines. We have to note that the amplitudes in either the secondly derived MR values or the FFT curves are not related to the intensity of the conductance contributions. This allows only ratios of amplitudes to be analyzed. The thickness of the sample is a few micrometers, while its length/width is a few millimeters. The SDH oscillation from SS over the bulk state is predominant when $\theta$ is below 45°. However, it becomes much smaller than the BS contribution (around 18 T in all the FFT curves) when it arrives at a larger $\theta$. If we carefully check the FFT curves at low $\theta$, we will find the tiny feature from the bulk state. Such coexistence of the SDH oscillations from both a 2D SS and the bulk channel is common in the samples of TIs.

4 Discussions

Detailed analysis of the transport environment was carried out for the samples to obtain the ratio of SS conduction.
Fig. 3. Two-dimensional SDH oscillations. (a) Angle-dependent second-derived MR curves of sample-M (high-field part). (b) Analysis of the frequency component in the FFT of the MR curve. The dominant frequency changes systematically with increasing angle. It is constant if the magnetic field is projected to the surface normal.

For the bulk transport of sample-M, their resistivity decrease from over 10 mΩ cm to around 1 mΩ cm while the temperature decreases from 300 K to 5 K. In the meantime, the mobility of the samples increases from 200 cm²/V s to around 3000 cm²/V s as determined by the Hall measurement as shown in Figure 1c. The dominant carriers are determined to be electrons and its volume density is around $10^{18}$ cm⁻³. The transport parameters of the SS can be extracted since the SDH oscillation exhibits the 2D behavior which is confirmed in the last paragraph. The dominant frequency of 18.5 T in Figure 3b leads to the areal carrier density of $4.6 \times 10^{11}$ cm⁻². Fitting the SDH oscillations according to the Dingle’s analysis [19], the effective mass of 0.09$m_e$ and the Dingle’s temperature of 3.2 ± 1.0 K are obtained. This leads to a mean free path of 45 nm, and the SS mobility¹ of 6825 ± 2100 cm²/V s. The error can be large due to very small amplitude of the SDH oscillations. Longitudinal zero-magnetic conductance $(G_{xx}(B)|B=0\tau)$, $G(0)$, can be acquired from 2 Dimensional Dirac Gas transportation theory, which is introduced in Qu’s article. According this theory, $G(0)$ has the following relations with wave vector number $(k_f)$ and elastic scattering-free-path($l$): $G(0) = (e^2/h)k_f l$, which also can be logically thought as surface state conductance $G_s = (e^2/h)k_f l$ if we confirm the existence of Surface State from sample-M. With $k_f l \sim 13.5$, we get $G_s = 5.184 \times 10^{-4}$ Ω⁻¹ while $R_{\text{bulk}} = 0.17$ Ω at 10 K. Thereby, the surface contribution to the overall conduction is then about 0.01 percent in sample-M.

As a discussion, the SS transport in the present samples falls between those in the fully insulating and highly metallic crystals. We compared our date to that of the Qu’s insulating sample, Q2, which mobility is $10600$ cm²/V s and the surface areal density is $7.2 \times 10^{11}$ cm⁻². The ratio of surface conduction is 0.3 percent [18]. Till now, all reported values for surface

¹ Band structures of Bi₂Te₃ have been extensively studied. A dispersion of $E = \hbar k + C_1 k^2$ has been manifested. We use a moderate value of effective mass of 0.09$m_e$ according to the recent measurements with the error of 10 percent.

5 Conclusions

We prepared a series of the TI crystals of Bi₂Te₃ by the melting-annealing approach. Some insulating samples reveal dominant SS contributions. We carried out the MR measurement and obtained the linear MR curves in some metallic Bi₂Te₃ crystalline sheets with the room-temperature resistivity of above 10 mΩ cm. The SDH oscillations were identified from the secondly-derived magnetoresistance curves and were attributed to the SS by field-tilting measurement. The work reveals a resolvable surface contribution to the overall conduction even in a metallic topological insulator.

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