Coexistence of bulk and surface states probed by Shubnikov–de Haas oscillations in Bi2Se3 with high charge-carrier density

de Vries, Eric; Pezzini, S; Meijer, M.J.; Koirala, N.; Salehi, M.; Moon, J.; Oh, S.; Wiedmann, S.; Banerjee, Tamalika

Published in:
Physical Review. B: Condensed Matter and Materials Physics

DOI:
10.1103/PhysRevB.96.045433

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Document Version
Publisher’s PDF, also known as Version of record

Publication date:
2017

Link to publication in University of Groningen/UMCG research database

Citation for published version (APA):

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Topological insulators (TIs), hosting spin-momentum locked surface states, received considerable interest in the past decade potentially serving as a platform for exploring many surface states, received considerable interest in the past decades. However, it is increasingly becoming clear that these surface transport channels can be accompanied by a finite conducting bulk, as well as additional topologically trivial surface states. To investigate these parallel conduction transport channels, we studied Shubnikov–de Haas oscillations in Bi$_2$Se$_3$ thin films, in high magnetic fields up to 30 T so as to access channels with a lower mobility. We identify a clear Zeeman-split bulk contribution to the oscillations from a comparison between the charge-carrier densities extracted from the magnetoresistances and the oscillations. Furthermore, our analyses indicate the presence of a two-dimensional state and signatures of additional states the origin of which cannot be conclusively determined. Our findings underpin the necessity of theoretical studies on the origin of and the interplay between these parallel transport channels for a careful analysis of the material’s performance.

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Topological insulators are ideally represented as having an insulating bulk with topologically protected, spin-textured surface states. However, it is increasingly becoming clear that these surface transport channels can be accompanied by a finite conducting bulk, as well as additional topologically trivial surface states. To investigate these parallel conduction transport channels, we studied Shubnikov–de Haas oscillations in Bi$_2$Se$_3$ thin films, in high magnetic fields up to 30 T so as to access channels with a lower mobility. We identify a clear Zeeman-split bulk contribution to the oscillations from a comparison between the charge-carrier densities extracted from the magnetoresistances and the oscillations. Furthermore, our analyses indicate the presence of a two-dimensional state and signatures of additional states the origin of which cannot be conclusively determined. Our findings underpin the necessity of theoretical studies on the origin of and the interplay between these parallel transport channels for a careful analysis of the material’s performance.

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In this paper, we used thin films of n-type Bi$_2$Se$_3$ with thickness $t$ = 10, 20, 30, and 100 quintuple layers (QLs) grown by molecular-beam epitaxy (MBE) on Al$_2$O$_3$(0001) substrates in a custom-designed SVTA MOS–V–2 MBE system at a base pressure lower than 5 × 10$^{-10}$ Torr following the methods as described in previous work [28]. The quality of the obtained films was characterized through various techniques [16, 29–32]. The films were patterned into Hall bars by using a combination of photolithography and Ar plasma dry etching. Contact pads consisting of Ti/Si/Au(70 nm) were made by combining photolithography with electron-beam evaporation. The resulting Hall bars [inset Fig. 1(a)] have dimensions of 2400 × 100 μm$^2$ where the resistance is measured over a probing length between 1400 and 2000 μm. The magnetotransport measurements have been performed in a cryostat with an out-of-plane rotation stage placed in a 30-T Bitter-type magnet in a four-probe geometry using the ac modulation technique at

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FIG. 1. (a) Out-of-plane magnetic field dependence of the longitudinal sheet resistance $R_{xx}$ and (b) the Hall resistance $R_{xy}$ for $t = 10$ QL at $T = 1.4$ K. The data (black) can be fitted with the two channel model (red) in good agreement. Oscillations in $R_{xx}$ are clearly visible beyond 15 T. Insets (a): Residual $\delta$ vs magnetic field and Hall bar geometry with TI channel in blue and contact pads in yellow. Inset (b): Residual $\delta$ vs magnetic field. (c) The second derivative of the resistance with respect to the magnetic field $-d^2 R_{xx}/dB^2$ plotted vs $1/B$. A clear oscillatory pattern is present with multiple oscillations. Beyond 15.5 T (0.065 T $^{-1}$) the oscillatory pattern can be reconstructed from oscillations with $f_x = 0.122 \pm 0.003$ kT, $f_{xy} = 0.236 \pm 0.003$ kT, and $f_y = 0.291 \pm 0.001$ kT. (d) Inset: Resulting FFT spectrum of the full oscillatory pattern. (d) Magnetic field evolution of the (smoothed) FFT spectrum analyzed for different FFT ranges starting from 11 T towards higher fields with steps of 2 T, as schematically depicted in (c). The FFT amplitude $A_{\text{FFT}}$ is plotted vs frequency $f$ where the curves are offset by 0.07 for clarity.

An ac current bias of 1 $\mu$A. In this paper, mainly the results on the sample with $t = 10$ QL will be discussed and comparisons will be made to the samples with larger thickness. Most of the results of the thicker samples can be found in Ref. [33].

The typical out-of-plane magnetic field dependence of the longitudinal sheet resistance $R_{xx}$ measured for the sample with $t = 10$ QL is shown in Fig. 1(a) where $R_{xx}$ tends to saturate at high magnetic fields. From this data and those for larger thickness as well as from the fitting (see below), we observe that the order of saturation is determined by the low mobility channel and the parabolic response at low fields is governed by the high mobility channel. Furthermore, the presence of at least two channels is clear from the nonlinear Hall resistance $R_{xy}$ [Fig. 1(b)]. As also shown earlier [30], we observe a slight upturn with a change in $R_{xx}$ $\sim$0.2% for samples with $t = 10$–30 QL below 10 K, indicative of the presence of defect states [10,12,34]. From the out-of-plane field dependence of the longitudinal and transverse resistance $R_{xx}(B)$ and $R_{xy}(B)$, we can extract the sheet carrier density $n_i$ and mobility $\mu_i$ for only two channels, which we expect to be due to the bulk and surface state(s). For that, we use a semiclassical Drude model where contributions from two parallel channels are summed in the conductivity tensor $\sigma$, which relates to the resistivity as $\rho = 1/\sigma$.

### TABLE I. Overview of the extracted charge-carrier densities $n_i/t$, $n_2$ and mobilities $\mu_1$, $\mu_2$ from the magnetoresistance measurements using the two-carrier Drude model.

<table>
<thead>
<tr>
<th>$t$ (QL)</th>
<th>$n_1/t$</th>
<th>$\times 10^9$ cm$^{-3}$</th>
<th>$n_2$</th>
<th>$\times 10^9$ cm$^{-3}$</th>
<th>$\mu_1$</th>
<th>$\times 10^3$ (V s)$^{-1}$</th>
<th>$\mu_2$</th>
<th>$\times 10^3$ (V s)$^{-1}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>10 ± 1</td>
<td>1.8 ± 0.1</td>
<td>3.4 ± 0.2</td>
<td>2060 ± 50</td>
<td>660 ± 50</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>20 ± 1</td>
<td>0.67 ± 0.03</td>
<td>2.4 ± 0.2</td>
<td>1190 ± 50</td>
<td>290 ± 50</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>30 ± 1</td>
<td>0.49 ± 0.02</td>
<td>2.7 ± 0.1</td>
<td>1250 ± 50</td>
<td>220 ± 50</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>100 ± 5</td>
<td>0.33 ± 0.02</td>
<td>4.2 ± 0.2</td>
<td>3800 ± 100</td>
<td>500 ± 100</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

As found from our analysis, simultaneous fitting of the $R_{xx}$ and $R_{xy}$ is required since $R_{xx}$ has a strong effect on the mobility and therefore will change the values found for $n_i$ from $R_{xy}$. An example of the simultaneous fit to the magnetoresistance curves $R_{xx}$ and $R_{xy}$ for the sample with $t = 10$ QL at 1.4 K is displayed in Fig. 1(a). A good agreement with the two-carrier model is obtained with a residual $\delta = (R_{\text{data}} - R_{\text{fit}})/R_{\text{data}}$ between 1 and 5% for both $R_{xx}$ and $R_{xy}$, but it is important to note that this analysis is limited to two channels and does not rule out the presence of more channels. Nevertheless, the good agreement between data and fit suggests that any additional state would have a similar mobility, which would add to an effective charge-carrier density in Eq. (1).

An overview of the extracted charge-carrier properties for all film thicknesses can be found in Table I; the data and fits to the magnetoresistance for the samples with larger thickness can be found in Ref. [33]. We listed $n_1/t$ because of its correspondence to the [three-dimensional (3D)] bulk channel (see discussion below), whereas $n_2$ is most likely linked to a 2D channel. The model describes the magnetoresistance behavior for $t$ up to 30 QL very well, but deviations from the model are observed for $t = 100$ QL, which will be discussed below. The correspondence between these extracted parameters and the information extracted from the SdH oscillations will be discussed in the remainder of this paper.

The possible presence of additional states can be analyzed by studying the SdH oscillations in $R_{xx}$, provided that the mobility of the channels is high enough [27]. For the sample with $t = 10$ QL, these oscillations can be observed from $\sim$10 T onwards, which indicates that transport channels are present with a mobility on the order of 1000 cm$^2$(V s)$^{-1}$. This is in agreement with estimates for $\mu_1$ as extracted from the magnetoresistance measurements (see Table I). To analyze the oscillations without the magnetoresistance background, the second derivative $-d^2 R_{xx}/dB^2$ is taken after interpolation and adjacent averaging of the data [33]. By plotting $-d^2 R_{xx}/dB^2$ versus $1/B$, we find an oscillatory pattern that shows additional oscillations from 15 T $\sim$0.067 T$^{-1}$, Fig. 1(d). We can follow the development of the oscillations by looking at the evolution of the fast Fourier-transform (FFT) spectrum when taking different ranges starting from 9 T $\sim$0.11 T$^{-1}$ towards
higher fields, where lower mobility channels start to contribute [Fig. 1(d)]. Below 15 T, as depicted by the black, red, and blue line in Fig. 1(d), one main frequency is observed indicated by α in the FFT spectrum as has been commonly reported in other works [11–13, 15–17, 19, 20].

Beyond 15 T, we find the clear presence of the harmonic 2α in the FFT spectrum which is due to the strong Zeeman splitting because of the large g factor in this material [35–38]. As shown in Fig. 2(a), the occurrence of Zeeman splitting is justified by an enhancement in oscillation amplitude when studying its dependence on the perpendicular to the in-plane component of the magnetic field, B⊥. Only the cyclotron energy is sensitive to this component, whereas the competing Zeeman energy is related to the total applied magnetic field. In addition to the high mobility channel linked to α, we find a lower mobility channel denoted by β. The appearance of the oscillation linked to β at higher magnetic field indicates that this channel has a mobility on the order of several times 100 cm²(V s)⁻¹ which is in agreement with the lower value μ₁ found from the earlier analysis of the magnetoresistance (Table I). Using the extracted three frequencies, we can reconstruct the oscillatory pattern at high fields as shown in Fig. 1(c) with deviations in the peak amplitudes of the pattern. Due to the good agreement between data and the reconstructed oscillatory pattern, we can conclude that in the used magnetic field range the magnetoresistance is dominated by these three frequencies. Nevertheless, additional channels with a lower mobility might be present but are beyond the resolution of our measurements.

To explore the dimensionality of the observed conduction channels, we can look at the angular dependence of the magnetic field orientation on the position of the frequency peaks. For 2D states we expect that their frequencies f scale with \( f \propto 1/\cos \theta \) where \( \theta \) is the angle between the surface normal and the direction of the applied magnetic field [inset Fig. 2(b)]. For bulk states it is commonly observed that \( f(\theta) \) initially follows the similar behavior but saturates between 30 and 60°, depending on the dimensions of the ellipsoidal pocket of these states [10, 18, 22, 39, 40]. However, few earlier works [11, 15, 41] report on a similar \( 1/\cos \theta \) dependence for the bulk states as well. Importantly, although we use thin films, the bulk will not show 2D behavior due to finite film thickness since the magnetic length \( l_B = \sqrt{n_e eB} \approx 8 \text{ nm} \) at fields of 10 T from which we start observing the oscillations. From these considerations for \( f(\theta) \) we can map out all observed peaks in the spectra at every angle \( \theta \) and check whether they fit into a 2D or 3D picture. In Fig. 2(b), the angular dependence of the observed peaks for \( r = 10 \text{ QL} \) is plotted from where we can trace the different channels α, 2α, and β up to an angle of 68°. Beyond this angle, the resolution of separate spectral peaks is limited, which is most probably linked to the low mobility of the channels and is manifested as a strong weakening of the oscillations at higher angles. Nevertheless, partially due to a higher mobility of the channel, we find a minor oscillation with \( f = (0.5 \pm 0.2) \text{ K} \) at \( \theta = 90° \) indicating that \( f_\alpha \) (and its harmonic) saturates and is due to the bulk channel with an elongated Fermi pocket. For the β peak, we find a \( 1/\cos \theta \) behavior which can be linked to the appearance of a 2D state.

Another way of clarifying the origin of the states is to extract the cyclotron mass from the temperature dependence of the oscillations which are observable up to ~50 K as shown in Fig. 2(c). Because of the presence of multiple oscillations it is difficult to extract the cyclotron mass from the FFT spectra. Inspired by recent work [42], we can extract the cyclotron mass by studying the temperature dependence of the peak amplitudes in the oscillations via the Lifshitz-Kosevich formalism. The result is shown in Fig. 2(d) where we can study the evolution of the cyclotron mass upon varying the magnetic field where different oscillations contribute. Comparing this result with the FFT spectrum evolution in Fig. 1(d) in which we observe a single channel up to 0.07 T⁻¹, we can conclude that the channel corresponding to the α peak has a cyclotron mass \( m_e = (0.15 \pm 0.01) m_e \) which is a typical value for the bulk conduction band [9]. Below 0.07 T⁻¹, we find a strong increase in the cyclotron mass up to ~0.28m_e after which it lowers to (0.20 ± 0.01)m_e and saturates. This higher value of \( m_e \) is probably due to the topological surface states [43], whereas a trivial 2DEG is supposed to have a similar mass as the bulk [23]. The interplay of the different oscillations could give rise to an increase in the cyclotron mass because channels with lower mobility (\( \alpha 1/m_e \)) start contributing, provided that the scattering times in the different channels are the same [42].

From the considerations above, we can match the charge-carrier densities extracted from the oscillations and from the magnetoresistance. From the FFT spectrum progression analysis, we can conclude that the α peak makes up the high mobility channel where the charge-carrier density \( n_\alpha = (1.26 \pm 0.06) \times 10^{11} \text{ cm}^{-2} \) when assuming bulk states \( n_{3D} = k_F^2 \bar{\nu}/(3\pi^2) \) with an ellipsoid pocket with ellipticity.
conceivable that this state is linked to the low mobility channel corresponds to the unaffected bulk layer. Furthermore, due to reduced scattering compared to that at any of the surfaces it is most likely that the high mobility channel of the Berry phase [33], but the extracted hints at a topological surface state. Furthermore, it is not clear whether this state resides at the top or bottom surface which partially accounts for the low mobility channel’s pronounced 2D state, either topologically trivial or nontrivial, effects under high magnetic field. Furthermore, we observe a beating pattern where oscillations of different frequencies partially cancel each other, yielding a loss of FFT amplitude. For the sample with \( t = 100 \) QL, we find a poor agreement between the charge-carrier densities from the magnetoresistance and the SdH oscillations where the bulk state (\( \alpha \) channel) can alone account for the total charge-carrier density \( n_1 + n_2 \). The fitting of the magnetoresistance data shows that the mobilities and charge-carrier densities could be different from the extracted values as stronger oscillations were expected for the mobilities extracted. The difference between the fit and data could originate from additional channels with a more distinct mobility suggesting that the two-channel model is too limited to describe the data properly. Lastly, from atomic force microscopy (AFM) images [33] we observe height variations across the film surface, which might influence fitting parameters such as the effective thickness of the transport channel and can cause changes to the bulk density \( n_1/t \) on the order of \( 2 \times 10^{17} \text{cm}^{-3} \). This is in the same range of charge-carrier densities found for the 2D states.

In conclusion, we find a good agreement between magnetoresistance data and the analysis of the SdH oscillations for Bi\(_2\)Se\(_3\) thin films based on the extracted charge-carrier densities, where the channel contributions are quite unrelated to film thickness. We find that the bulk channel has a high mobility and is characterized by an ellipsoid Fermi pocket but a clear saturation in the angular dependence is absent. Due to the strong \( g \) factor in these materials we observe a Zeeman splitting in our oscillations which has been observed before in optical measurements and investigations on thermoelectric effects under high magnetic field. Furthermore, we observe a pronounced 2D state, either topologically trivial or nontrivial, which partially accounts for the low mobility channel’s charge-carrier density. Additional 2D states are observed but are often masked by the limited resolution of our analysis originating from the channel mobilities and charge-carrier densities. The limited resolution of the angular dependence and the difficulties to extract parameters like the Berry phase make it difficult to make a definitive statement on the origin of these states.

The authors would like to thank L. Tang (Radboud University) for useful discussions on the processing of the data. Furthermore, the authors would like to thank J. G. Holstein, H. M. de Roosz, and T. J. Schouten (University of Groningen) for the technical support. We acknowledge the support of the HFML, member of the European Magnetic Field Laboratory. Work at the University of Groningen is supported by a
COEXISTENCE OF BULK AND SURFACE STATES PROBED...
