Taking topological insulators for a spin

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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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Chapter 5

Charge transport under high magnetic fields in Bi$_2$Se$_3$

It is increasingly becoming clear that the surface transport channels in Bi-based topological insulators are often accompanied by a finite conducting bulk, as well as additional topologically trivial surface states. In order to investigate these parallel conduction channels, we have 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 magnetoresistance 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.

5.1 Introduction

As described earlier in this thesis, topological surface states have been well investigated by surface sensitive techniques such as (spin-resolved) ARPES, STM, and scanning tunneling spectroscopy (STS). Such techniques adequately describe the electronic properties of the (non)trivial surface states, but cannot account for additional transport features as observed in (magneto)transport experiments. In order to employ topological insulators in solid-state devices, direct access and understanding of these additional surface states in transport experiments are needed.

Studying Shubnikov–de Haas (SdH) oscillations can reveal the existence of such surface states where parameters such as the mobility, charge-carrier density, dimensionality, and the Berry phase of the states can be determined (see section 5.2). Earlier studies on various Bi-based topological insulators report on single or double frequency SdH oscillations [1–17]. Here, it is often claimed that these oscillations originate from the top and bottom topological surface state with the expected Berry phase and angular dependence. The magnetic field strength used in these studies is usually up to 15 T, which only allows to probe transport channels with a relatively high mobility. However, nonlinear Hall measurements indicate additional channels with a lower mobility to be present. Besides a finite conducting bulk, these additional, topologically trivial channels can originate from variations in the electrostatic potential near the surfaces and can be spin textured too [18–20], which we will refer to as 2D electron gas (2DEG). From earlier transport measurements, the mobilities of the different channels are found to be on the order of 50–500 and $\sim 3000$ cm$^2$/Vs where the low mobility channel has a higher charge-carrier density [9, 21]. Notably, from these numbers one can find that in terms of conductivity these channels can contribute equally to the electrical transport.

Motivated by these works, we have performed magnetotransport experiments up to 30 T and studied SdH oscillations to explore the most prominent conduction channels and additional channels with mobilities below 1000 cm$^2$/Vs, satisfying the requirement $\mu B \gg 1$ with $\mu$ the charge-carrier mobility and $B$ the magnetic field strength. The magnetotransport is studied in thin films of Bi$_2$Se$_3$ so as to minimize bulk effects and amplify the topologically trivial and nontrivial surface states. In contrast to earlier works, we will show that the bulk channel with a high mobility is present along with a prominent 2D channel that can be linked to the topological surface states. Our findings indicate the presence of additional channels with a lower mobility that cannot be precisely resolved from the oscillations. Similar to [22], we compare charge-carrier densities from the SdH oscillations with those from the magnetoresistance and study the dimensionality of the various channels in order to unravel the origin of these states.

5.2 Lifshitz–Kosevich theory

The observation of Shubnikov–de Haas oscillations is a result of Landau quantization of the energy levels in the two dimensions perpendicular to the applied magnetic field. The (de)population of these Landau levels gives rise to oscillations in the longitudinal resistance and its most obvious manifestation is that in the quantum Hall effect where
Lifshitz–Kosevich theory

A finite resistance can be measured when crossing a Landau level (see section 2.2). Crossing such a level gives rise to an increase in the density of states that yields an increase in the scattering (delocalization) and thus one obtains a finite resistance. In bulk systems, there is a dispersion in the third dimension parallel to the field, which gives rise to an ever finite resistance since the system is not fully localized. Furthermore, thermal and scattering effects that affect the mobility and the strength of the magnetic field control the density of states and the separation between the discrete Landau levels. The presence of 2D and 3D states in topological insulators under study, where the top and bottom surface states can have a different mobility, is therefore expected to yield a complicated pattern of oscillations in the resistance.

The oscillations in the longitudinal resistance $\Delta R_{xx}$ of each transport channel can be described by the Lifshitz–Kosevich formalism and is proportional to [7]:

$$\Delta R_{xx} \propto R_T R_D \cos \left[ 2\pi \left( f \frac{B}{B_{\text{sat}}} - \frac{1}{2} + \beta \right) \right],$$

with

$$R_T = \frac{2\pi^2 \left( \frac{k_B T}{\hbar \omega_c} \right)}{\sinh \left[ 2\pi^2 \left( \frac{k_B T}{\hbar \omega_c} \right) \right]},$$

and

$$R_D = e^{\frac{\pi}{\tau_D}},$$

where $f$ is the frequency of the oscillation, $B$ the applied magnetic field, $\beta$ the phase factor, $k_B$ the Boltzmann constant, $T$ the temperature, $\hbar$ the reduced Planck constant, $\omega_c = eB/m_c$ with $e$ the electron charge and $m_c$ the cyclotron mass, and $\tau_D$ the Dingle scattering time. By using equations (5.1), we can extract various parameters from the SdH oscillations. From the extracted frequency $f$, we can determine the Fermi surface area $A(E_F)$:

$$f = \frac{\hbar}{2\pi e} A(E_F)$$

If we assume a circular Fermi surface ($k_{F,a} = k_{F,b}$) as observed for the topological surface states from ARPES studies (2.3) then $A(E_F) = \pi k_{F}^2$. There are $L^2/(2\pi)^2$ allowed quantum states per unit area in $k$ space, where $L^2$ is the area of the material slab. From this, we can evaluate the total number of quantum states in $A(E_F)$ that should equal the total number of electrons $N$:

$$\pi k_{F}^2 \frac{L^2}{(2\pi)^2} = N,$$
which gives

\[ n_s = \frac{k_F^2}{4\pi} \]  

(5.3b)

where \( n_s = N/L^2 \). In contrast to the nondegenerate topological surface state, an ordinary 2D electron gas is degenerate and therefore we have to include a factor of 2 [this would yield 2\( \pi \) instead of 4\( \pi \) in equation (5.3b)]. For bulk systems with a spherical Fermi surface, we find \( k_{3D}^F = \left(3\pi^2 n_{3D}\right)^{1/3} \) and thus \( n_{3D} = \left(k_{3D}^F\right)^3 / 3\pi^2 \). In the case of an ellipsoidal bulk Fermi surface with \( k_{F,a} = k_{F,b} \neq k_{F,c} \), an extra factor \( \eta = k_{F,c}/k_{F,a} \) is included when calculating \( n_{3D} \). The extracted charge-carrier densities from the oscillations can then be compared with the two-carrier model as will be derived in section 5.5.2.

The shape (and dimensions) of the Fermi surface can be analyzed by studying the angular dependence of the magnetic field orientation on the frequency of the oscillations. 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 figure 5.2b). For bulk states in topological insulators, it is commonly observed that \( f(\theta) \) initially follows the similar behavior but saturates between 30 and 60\( \degree \), depending on the dimensions of the ellipsoidal pocket of these states [3, 12, 16, 23, 24]. However, few earlier works [4, 8, 25] 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, because the magnetic length \( l_B = \sqrt{\hbar/eB} \leq 8 \) nm at fields of 10 T from which we start observing the oscillations.

The temperature dependence as stated in equation (5.1b) describes the thermal broadening of the Fermi distribution relative to the cyclotron energy and from this expression the cyclotron mass can be determined for a single band. By plotting the peak amplitude at a specific magnetic field for different temperatures, estimates for the cyclotron mass can be extracted. For multiple oscillations, it is more difficult to determine the effective mass of the separate bands since the overlap between the oscillations yields a different value for the cyclotron mass, as will be discussed in the section 5.3. An alternative method to extract the cyclotron mass is by looking at the temperature dependence of the spectral peak’s amplitude and by including an effective magnetic field range \( B_{\text{eff}} \) over which the FFT was taken. However, the determination of \( B_{\text{eff}} \) is difficult, mainly due to the manifestation of the oscillations at different field ranges. Furthermore, the spectral peaks in the FFT spectra are not well defined such that there is overlap between the peaks. The overlap between the peaks as well as the difficulty to read off the peak amplitude of the poorly defined peaks, especially at higher \( T \), makes this method problematic. Another method will be presented in section 5.3 in which the evolution of the oscillations or FFT peaks will be connected to the change in effective mass. Importantly, since charge transport only involves charge carriers around the Fermi energy the measured cyclotron mass can only be determined for bands involved at that energy.

From \( R_D \) [equation (5.1c)], we can extract the Dingle scattering time \( \tau_D \) which can be related to the quantum mobility of the charge carriers that influences the broadening of the Landau levels. However, due to the presence of multiple peaks, it is impossible to extract these parameters in our measurements.
Finally, we can look at the phase $\beta$ by constructing a Landau level fan diagram where we index the maxima and minima in the conductivity and plot those values against the magnetic field. The procedure for indexing maxima and minima is clearly described in [26]. By finding the intersection with the horizontal axis in this diagram, we can determine the phase $\beta$. For ordinary systems it is expected that $\beta = 0$, but for Dirac systems a phase $\beta = 1/2$ is expected because of the presence of a zero-energy Landau level at the Dirac point (see section 2.2). The phase analysis will be discussed in section 5.5.1.

### 5.3 Results and Discussion

In this study, we have used thin films of $n$-type Bi$_2$Se$_3$ with thickness $t = 10$, 20, 30, and 100 quintuple layers (QL) grown by MBE on Al$_2$O$_3$(0001) substrates in a custom designed SVTA MOS-V-2 system at a base pressure lower than $5 \times 10^{-10}$ Torr, following the methods as described in previous work [27]. The quality of the obtained films has been characterized through various techniques [9, 28–31]. The films have been patterned into Hall bars by using a combination of photolithography and Ar plasma dry etching. Contact pads consisting of Ti(5)/Au(70) are made by combining UV lithography with electron beam evaporation. The resulting Hall bars (inset figure 5.1a) have dimensions of $2400 \times 100 \ \mu m^2$ where the longitudinal resistance is measured over a probing length between 1400 and 2000 $\mu m$. 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 an ac current bias of 1 $\mu A$. In this section, 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 section 5.5.1.

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 figure 5.1a where $R_{xx}$ tends to saturate at high magnetic fields. From this data and those for larger thickness as well as from the fitting, 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 $R_{xy}$ (figure 5.1b). As also shown in section 6.3, 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 [3, 5, 32]. 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 semi-classical Drude model where contributions from two parallel channels are summed in the conductivity tensor $\sigma$, which relates to the resistivity $\rho$ as $\rho = \sigma^{-1}$ (more details on the analysis can be found in section 5.5.2):
Charge transport under high magnetic fields in Bi$_2$Se$_3$

Figure 5.1: (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 $-d^2R_{xx}/dB^2$ plotted vs $1/B$. A clear oscillatory pattern is present with multiple oscillations. Beyond 15.5 T (0.065 T$^{-1}$) the oscillatory pattern (black) can be reconstructed from oscillations with $f_\alpha = 0.122 \pm 0.003$ kT, $f_{2\alpha} = 0.236 \pm 0.003$ kT, and $f_\beta = 0.291 \pm 0.001$ kT (red). Inset: Resulting FFT spectrum. (d) Evolution of the (smoothed) FFT spectrum analyzed for increasing FFT ranges starting from 11 T towards higher fields with steps of 2 T, as schematically depicted in (c). The FFT amplitude $A_{FFT}$ is plotted vs frequency $f$ where the curves are offset by 0.07 for clarity.

\[
\sigma_{xx} = \frac{n_1 e \mu_1}{1 + \mu_1^2 B^2} + \frac{n_2 e \mu_2}{1 + \mu_2^2 B^2}, \quad (5.4a)
\]

\[
\sigma_{xy} = \frac{n_1 e \mu_1^2 B}{1 + \mu_1^2 B^2} + \frac{n_2 e \mu_2^2 B}{1 + \mu_2^2 B^2}. \quad (5.4b)
\]

As found from our analysis, simultaneous fitting of $R_{xx}$ and $R_{xy}$ is required since
**Results and Discussion**

$R_{xx}$ 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 figure 5.1a. A good agreement with the two-carrier model is obtained with a residual $\delta = (R_{data} - R_{fit}) \times 100\% / R_{data}$ between 1 and 5% for both $R_{xx}$ and $R_{xy}$. However, 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 equation (5.4).

An overview of the extracted charge-carrier properties for all film thicknesses can be found in table 5.1; the data and fits to the magnetoresistance for the samples with larger thickness can be found in section 5.5.1. We listed $n_1/t$ because of its correspondence to the 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. The correspondence between these extracted parameters and the information extracted from the SdH oscillations will be discussed in the remainder of this section.

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 [22]. 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$^2$/Vs. This is in agreement with estimates for $\mu_1$ as extracted from the magnetoresistance measurements (see table 5.1). In order to analyze the oscillations without the magnetoresistance background, the second derivative $-d^2R_{xx}/dB^2$ is taken after interpolation and adjacent averaging of the data (see also section 5.5.3). By plotting $-d^2R_{xx}/dB^2$ versus $1/B$, we find an oscillatory pattern that shows additional oscillations from 15 T ($\sim 0.067$ T$^{-1}$, figure 5.1c). 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, as shown in figure 5.1d. Below 15 T, as depicted by the black, red, and blue line in the figure, one main frequency is observed indicated by $\alpha$ as has been commonly reported in other works [4–6,8–10,13,14].

Beyond 15 T, we find the clear presence of the harmonic $2\alpha$ in the FFT spectrum, which is due to the strong Zeeman splitting because of the large $g$ factor in this material [33–36]. As shown in figure 5.2a, the occurrence of Zeeman splitting is

<table>
<thead>
<tr>
<th>$t$(QL)</th>
<th>$n_1/t$ ($\times 10^{19}$/cm$^3$)</th>
<th>$n_2$ ($\times 10^{13}$/cm$^2$)</th>
<th>$\mu_1$ (m$^2$/Vs)</th>
<th>$\mu_2$ (m$^2$/Vs)</th>
</tr>
</thead>
<tbody>
<tr>
<td>10±1</td>
<td>1.8±0.1</td>
<td>3.4±0.2</td>
<td>0.206±0.005</td>
<td>0.066±0.005</td>
</tr>
<tr>
<td>20±1</td>
<td>0.67±0.03</td>
<td>2.4±0.2</td>
<td>0.119±0.005</td>
<td>0.029±0.005</td>
</tr>
<tr>
<td>30±1</td>
<td>0.49±0.02</td>
<td>2.7±0.1</td>
<td>0.125±0.005</td>
<td>0.022±0.005</td>
</tr>
<tr>
<td>100±5</td>
<td>0.33±0.02</td>
<td>4.2±0.2</td>
<td>0.38±0.01</td>
<td>0.05±0.01</td>
</tr>
</tbody>
</table>

Table 5.1: Overview of the extracted charge-carrier densities $n_1/t$, $n_2$ and mobilities $\mu_1$, $\mu_2$ from the magnetoresistance measurements using the Drude model for two parallel channels.
justified by an enhancement in oscillation amplitude when studying its dependence on the perpendicular-to-the-sample-plane component of the magnetic field, $B_\perp$. The cyclotron energy is only 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 $\alpha$, we find a lower mobility channel denoted by $\beta$. The appearance of the oscillation linked to $\beta$ at higher magnetic field indicates that this channel has a mobility on the order of several times 100 cm$^2$/Vs. This is in agreement with the lower value $\mu_2$ found from the earlier analysis of the magnetoresistance (table 5.1). Using the extracted three frequencies, we can reconstruct the oscillatory pattern at high fields as shown in figure 5.1c 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 the magnetotransport is dominated by these three frequencies in the used magnetic field range. 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. From the considerations for $f(\theta)$, as described in section 5.2, 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 figure 5.2b, the angular dependence of the observed peaks for $t = 10$ QL is plotted from where we can trace the different channels $\alpha$, $2\alpha$, and $\beta$ 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 that 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 $f = (0.5\pm0.2)$ kT at $\theta = 90^\circ$ indicating that $f_\alpha$ and its harmonic saturate, which is due to the bulk channel with an elongated Fermi pocket. For the $\beta$ peak, we find a $1/\cos \theta$ behavior which can be linked to the appearance of a 2D state$^1$.

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 $\sim 50$ K as shown in figure 5.2c. Because of the presence of multiple oscillations, it is difficult to extract the cyclotron mass from the FFT spectra. Inspired by recent work [40], 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 figure 5.2d 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 figure 5.1d where a single channel down to 0.07 T$^{-1}$ is observed, we can conclude that the channel corresponding to the $\alpha$ peak has a cyclotron mass $m_c = (0.15\pm0.01)m_e$, which is a typical value for the bulk conduction band [1]. Below 0.07 T$^{-1}$, we find a strong increase in the cyclotron mass up to $\sim 0.28m_e$ after which it lowers to $(0.20\pm0.01)m_e$ and saturates. This higher value of $m_c$ is probably due to the topological surface states [40,41], whereas a trivial two-dimensional electron gas is supposed to have a similar mass as the bulk [18]. The interplay of the different oscillations could give rise to an increase in the cyclotron mass.
Results and Discussion

Figure 5.2: (a) $B_\perp$ dependence on the oscillations above 0.06 T$^{-1}$ for increasing angle as indicated by the arrow. An enhancement of the oscillation amplitude is observed at locations indicated by the dashed lines. (b) Angular dependence of the frequencies extracted from the obtained FFT spectra. The dark gray, open square symbols designate additional peaks observed in the FFT spectra but which do not follow a clear angular dependent trend. The large error bars above 70° display the range of the peak position which cannot be determined accurately from the FFT spectrum. Insets: Remaining oscillation at $\theta = 90^\circ$ and schematics of the relative directions of current $I$ and magnetic field $B$. (c) Temperature dependence of the oscillations measured from 1.5 to 48 K. (d) Extracted cyclotron masses per peak position following from the data in (c).

mass because channels with lower mobility ($\propto 1/m_c$) start contributing, provided that the scattering times in the different channels are the same [40].

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 $\alpha$ peak makes up the high mobility channel where the charge-carrier density $n_\alpha = (1.26 \pm 0.06) \times 10^{19} \text{cm}^{-3}$ when assuming bulk states ($n_{3D} = k_{F,b}^2 k_{F,c}/3\pi^2$) with an ellipsoid pocket with ellipticity $k_{F,c}/k_{F,b} = 1.8$ [3]. The value for $n_\alpha$ is in reasonable agreement with $n_1/t$ found from the magnetoresistance analysis as displayed in table 5.1. Furthermore, due to reduced scattering compared to that at any of the surfaces it is most likely that the high
mobility channel corresponds to the unaffected bulk layer.

The state indicated by $\beta$ appears above 15 T and thus it is conceivable that this state is linked to the low mobility channel with $n_2$. The origin of the observed 2D surface state, trivial or nontrivial, cannot be concluded from the determination of the Berry phase (see section 5.5.1), but the extracted larger $m_c$ at high fields being different from bulk values hints at a topological surface state. Furthermore, it is not clear whether this state resides at the top or bottom surface because the characteristics of the electrostatics at both surfaces which would affect the mobility are unknown. Assuming $n_{\text{TSS}} = k_F^2/4\pi$ for a topological surface state and $n_{\text{2DEG}} = k_F^2/2\pi$ for a possible two-dimensional electron gas, the charge-carrier density related to $f_\beta$ varies between $n_{\beta,\text{TSS}} = (6.7 \pm 0.3) \times 10^{12}/\text{cm}^2$ and $n_{\beta,\text{2DEG}} = (1.34 \pm 0.05) \times 10^{13}/\text{cm}^2$, which makes up for 20 or 40% of $n_2$. We are careful to assume that this oscillation is linked to one surface state since it has been earlier reported that similar $n_\beta$ is present at the opposite surface [42], provided the mobilities at both surfaces are similar. Furthermore, as will be shown for $t = 20$ QL, an additional peak between $\alpha$ and $2\alpha$ occurs which shows that additional states exist, which adds to the low mobility charge-carrier density $n_2$.

The picture based on the charge-carrier densities for $t = 10$ QL also applies for the samples with $t = 20$ and 30 QL. However, the correction for the ellipsoidal asymmetry is most probably smaller compared to the sample with $t = 10$ QL, which can be related to a lower charge-carrier density [3]. Comparing the two films with $t = 10$ and 20 QL, we observe oscillations (figure 5.3a) with a similar spectrum but with the presence of an additional $\gamma$ peak for $t = 20$ QL (figure 5.3b), which could be a signature of a state at the surface opposite to where the channel linked to $\beta$ resides. Furthermore, the peak positions have changed, which is due to differences in charge-carrier density as also observed in the magnetoresistance measurements. From the similarities between these two samples, we can conclude that the thickness (i.e. bulk size) does not play a role but it is rather the relative mobilities and charge-carrier densities in these samples that are the decisive factors for the relative channel contributions.

For the thicker samples, as shown in figures 5.3c and 5.3d, we observe a dominant $\alpha$ peak in the spectrum while the $2\alpha$, $\beta$, and $\gamma$ peaks are present with a poor resolution. The reason for the decrease in amplitude is a lower signal-to-noise level of the measured voltage which generates a background and gives rise to a larger spectral width in the FFT spectrum. Furthermore, the oscillations show 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. The bulk state ($\alpha$ channel) can alone account for the total charge-carrier density $n_1 + n_2$. The poor fitting of the magnetoresistance data shows that the mobilities and charge-carrier densities could be different from the extracted values. Furthermore, stronger oscillations are expected for the extracted mobilities. 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 AFM images (see section 5.5.1), 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
Figure 5.3: (a) The second derivative of the resistance with respect to the magnetic field – \(d^2R_{xx}/dB^2\) plotted vs \(1/B\) for \(t = 20\) QL. A clear oscillatory pattern is present with multiple oscillations (b) Temperature dependence of the FFT spectrum (smoothed) based on the oscillations in (a). Colors correspond to the temperatures as depicted in (a). Temperature-dependent FFT spectra (smoothed) for (c) \(t = 30\) QL and (d) \(t = 100\) QL.

of \(2 \times 10^{17}/\text{cm}^3\). This is in the same range of charge-carrier densities found for the 2D states.

### 5.4 Conclusions

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. Here, the channel contributions are quite unrelated to film thickness but rather to the mobility and charge carrier density. We find that the bulk channel has a high mobility and is characterized by an ellipsoid Fermi pocket but a clear saturation of the oscillation frequency 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 mobil-
ity 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 such as the Berry phase make it difficult to make a definitive statement on the origin of these states.

5.5 Appendix

In this section, data in addition to the main part of this chapter will be provided. Furthermore, I will derive the expression for the two-carrier model, discuss the details on the analysis of the oscillations as well as on the possibilities to improve the FFT spectra. Finally, I will present some interesting negative magnetoresistance features and discuss weak antilocalization that we observed in our measurements.

5.5.1 Additional data

In this section, additional data can be found for all the samples including the extracted charge-carrier densities from the SdH oscillations which can be compared with the values extracted from the Drude modeling. Furthermore, AFM images will be shown which give an idea about the growth quality and the error in the thickness, leading to the error in $n_1/t$. For $t = 10$ QL, we further performed a Berry phase analysis.

Additional data for sample with $t = 10$ QL

A typical AFM image for this sample is shown in figure 5.4a from which we assign a maximum uncertainty in the channel thickness $t$ of 1 QL as a conservative margin.

To understand the origin of the 2D states, we can extract the Berry phase from the oscillations. A careful analysis has been described in [26] on how to index the maxima and minima in $dG_{xx}/dB$. For our analysis, we looked at the maxima and minima in $d^2G_{xx}/dB^2$ where the maxima in $d^2G_{xx}/dB^2$ coincide with the minima in $G_{xx}$ (labeled with integer $n$) and minima in $d^2G_{xx}/dB^2$ correspond to maxima in $G_{xx}$ [labeled $n + 1/2 (n = 1, 2, 3, ..)$]$^2$. From figure 5.4b, it can be seen that the oscillation complies with the predicted behavior over a small range but due to the presence of multiple oscillations the data acquires a different phase compared to the theoretical curve. Therefore, the analysis can only be employed for the first three maxima. Plotting $1/B_n$ versus $n$ yields a phase between 0.04 and 0.38 depending on the frequency taken; $f_\beta = 0.277$ kT is the value found from the FFT analysis, $f_\beta = 0.285$ kT corresponds to the best fit of the data and is within the error for $f_\beta$ as given in table 5.2. Due to the presence of multiple oscillations with different frequency, the limited number of oscillations, and the error in $f_\beta$, a proper extraction of the Berry phase value cannot be made. Furthermore, Zeeman coupling and deviations from the linear dispersion limit the possibility to extract the Berry phase from linear extrapolation in systems with a low mobility [17,43].

---

$^2$This labeling is related to the presence of a zero-energy Landau level as introduced in section 2.2.
Appendix

Figure 5.4: (a) AFM image for sample with \( t = 10 \) QL. (b) \( d^2G_{xx}/dB^2 \) plotted vs 1/\( B \) for Berry phase analysis (black). The red curve shows the theoretical positions of maxima and minima in \( d^2G_{xx}/dB^2 \) with \( f = 0.277 \) kT. (c) Landau level fan diagram extracted from the maxima and minima in the oscillatory pattern as shown in (b).

<table>
<thead>
<tr>
<th>Label</th>
<th>( f ) (kT)</th>
<th>( n_{TSS}, n_{2D} \times 10^{12}/\text{cm}^2 )</th>
<th>( n_{2DEG} \times 10^{12}/\text{cm}^2 )</th>
<th>( n_{3D} \times 10^{19}/\text{cm}^3 )</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \alpha )</td>
<td>0.115±0.005</td>
<td>2.8±0.1</td>
<td>5.6±0.2</td>
<td>1.26±0.07</td>
</tr>
<tr>
<td>( \beta )</td>
<td>0.28±0.01</td>
<td>6.7±0.3</td>
<td>13.4±0.5</td>
<td>4.7±0.2</td>
</tr>
<tr>
<td>( n_1 )</td>
<td>18±1</td>
<td></td>
<td></td>
<td>1.8±0.1</td>
</tr>
<tr>
<td>( n_2 )</td>
<td>34±2</td>
<td></td>
<td></td>
<td>3.4±0.2</td>
</tr>
</tbody>
</table>

Table 5.2: Extracted values for \( n_{TSS}, n_{2DEG}, \) and \( n_{3D} \) (with \( k_{F,c}/k_{F,b} = 1.8 \)) from the oscillations and \( n_1 \) and \( n_2 \) from the magnetoresistance (as can also be found in table 5.1 in section 5.3) for \( t = 10 \) QL.

At last, the values for \( n_{TSS} = k_{F,b}^2/4\pi, n_{2DEG} = k_{F,b}^2/2\pi, \) and \( n_{3D} = k_{F,b}^2 k_{F,c}/3\pi^2 \) that we extract from the frequencies in the FFT spectra are displayed in table 5.2. Here, we assume \( \eta=1.8 \) which has been reported for samples with similar charge-carrier density by Kulbachinskii et al. [3]. These values can be compared to \( n_1 \) and \( n_2 \) which are calculated for the 2D and 3D case and included in table 5.2. As also
discussed in the main text, we observe a reasonable agreement between $n_\alpha$ (with $f = 0.115 \, kT$) and $n_1/t$. It is important to realize that when considering all channels to be 2D, the charge-carrier density from the oscillations clearly underestimates the values found from the two channel fit. The presence of a bulk channel therefore seems feasible and in agreement with the presence of a residual oscillation at $\theta = 90^\circ$ and a Zeeman-split bulk state. The contribution of $n_\beta$ to $n_2$ is 20\% when this channel is linked to a topological surface state. From band structure calculations [44], we find that the next bulk band is located around 1 eV higher than the first conduction band minimum which makes the origin of the state $\beta$ to be related to a second bulk band unlikely. In addition, the FFT spectra that we observe in comparison with the work by Kulbachinskii et al. [3] are different such that a second bulk state is more improbable. The presence of additional channels with an even lower mobility could account for the difference between the values extracted from the magnetoresistance and those from the SdH oscillations.

**Additional data for sample with $t = 20$ QL**

In figures 5.5a and 5.5b, the data and fits for the out-of-plane field dependence of the sheet resistance $R_{xx}$ and the Hall resistance $R_{xy}$ for the sample with $t = 20$ QL are shown. Here, a good agreement between data and fit is observed. Importantly for this sample, the resistance has been measured for fields up to 33 T in order to include the last clear oscillation, enhancing slightly the resolution in our FFT spectrum.

When studying the FFT range progression analysis, additional bands start to appear already below 15 T, as shown in figure 5.5d. This is striking considering the low extracted mobility $\mu_2$ ($\mu B \gg 1$) and is inconsistent with the trend observed for the sample with $t = 10$ QL. A direct assignment of the mobility to the appearance of the bands at certain fields is thus not straightforward. The larger amplitude of the oscillation contributes to a good resolution in FFT such that the spectral peaks already appear at lower fields. The order of appearance of the peaks as displayed in figure 5.5d is similar to that described in the main text with the additional $\gamma$ peak starting to appear around similar fields as the harmonic peak. By reconstructing the oscillatory pattern, as shown in figure 5.5c, $f_\alpha$, $f_\beta$, and $f_\gamma$ are found from the fit, whereas additional inclusion of $f_{2\alpha}$ does not improve the fit much and only modifies the amplitude slightly, in agreement with the weak appearance in figure 5.5d.

Table 5.3 shows the extracted values for $n_{\text{TSS}}$, $n_{\text{2DEG}}$, and $n_{\text{3D}}$ with $k_{F,c} \approx k_{F,a}$

<table>
<thead>
<tr>
<th>Label</th>
<th>$f$ (kT)</th>
<th>$n_{\text{TSS}}$</th>
<th>$n_{\text{2DEG}}$ $\times \times 10^{12}$/cm$^2$</th>
<th>$n_{\text{3D}}$ $\times \times 10^{19}$/cm$^3$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\alpha$</td>
<td>0.077±0.002</td>
<td>1.86±0.05</td>
<td>3.7±0.1</td>
<td>0.38±0.02</td>
</tr>
<tr>
<td>$\gamma$</td>
<td>0.125±0.007</td>
<td>3.0±0.2</td>
<td>6.0±0.4</td>
<td>0.79±0.07</td>
</tr>
<tr>
<td>$\beta$</td>
<td>0.186±0.005</td>
<td>4.5±0.2</td>
<td>9.0±0.3</td>
<td>1.43±0.06</td>
</tr>
<tr>
<td>$n_1$</td>
<td>13±1</td>
<td></td>
<td></td>
<td>0.67±0.03</td>
</tr>
<tr>
<td>$n_2$</td>
<td>24±2</td>
<td></td>
<td></td>
<td>1.1±0.3</td>
</tr>
</tbody>
</table>

Table 5.3: Extracted values for $n_{\text{TSS}}$, $n_{\text{2DEG}}$, and $n_{\text{3D}}$ (with $k_{F,c} \approx k_{F,a}$) and $n_1$ and $n_2$ from the magnetoresistance (as can also be found in table 5.1 in section 5.3) for $t = 20$ QL.
Figure 5.5: (a) Out-of-plane magnetic field dependence of (a) the longitudinal sheet resistance $R_{xx}$ and (b) the Hall resistance $R_{xy}$ for $t = 20$ QL at $T = 1.4$ K. The data (black) can be fitted with the two channel model (red) in good agreement as can be seen in the insets showing the residuals $\delta$ as defined in the main text. Oscillations in $R_{xx}$ are again clearly visible beyond 15 T. (b) The second derivative of the resistance with respect to the magnetic field $-d^2R_{xx}/dB^2$ plotted vs $1/B$. Beyond 16 T (0.062 T$^{-1}$) the oscillatory pattern (black) can be reconstructed from oscillations with $f_\alpha = 0.078 \pm 0.004$ kT, $f_\gamma = 0.118 \pm 0.004$ kT, and $f_\beta = 0.188 \pm 0.001$ kT (red). (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 depicted in (c). The FFT amplitude $A_{FFT}$ is plotted vs frequency $f$ where the curves are offset by 0.07 for clarity.

as we have a lower charge-carrier density in this sample [3]. These values can be compared to $n_1$ and $n_2$ as also displayed in table 5.3. We find a good agreement between $n_{\alpha,3D}$ and $n_1/t$ giving good confidence on the origin of this state. As shown in figure 5.6a, we observe hardly any thickness variation and therefore the error in $t$ is expected to be small. Furthermore, we find that the channel linked to $\beta$ makes up 20% of $n_2$, assuming it to be a topological surface state. The additional $\gamma$ state accounts for 13% of $n_2$ which only partially explains the difference in values found from the oscillations and the magnetoresistance.

Furthermore, we display the angular dependence for the sample with $t = 20$ QL in where we see a similar behavior as for the $t = 10$ QL sample, i.e. all the four peaks
follow a $1/\cos \theta$ dependence up to the angles where we are able to observe oscillations, as shown in figure 5.6b. For this sample, it is not possible to observe any remaining bulk oscillations at $90^\circ$, which can be related to the lower mobility compared to that of the sample with $t = 10$ QL. At last, we show the cyclotron mass extracted per peak from the temperature dependence in figure 5.6c. In agreement with the data for $t = 10$ QL, we observe a steady increase in $m_c$ towards higher magnetic field but without a structural trend as for the data for $t = 10$ QL. Furthermore, the cyclotron mass seems to be slightly higher compared to the sample with $t = 10$ QL.

**Additional data for sample with $t = 30$ QL**

In figures 5.7a and 5.7b, the data and fits for the out-of-plane field dependence of the sheet resistance $R_{xx}$ and the Hall resistance $R_{xy}$ for the sample with $t = 30$ QL are shown. A good agreement between data and fit is observed. In this case, the SdH oscillations are not as clear as seen in the previous samples. Nevertheless, upon plotting $-d^2R_{xx}/dB^2$ versus $1/B$, we find a clear oscillatory pattern with an increased
Figure 5.7: Out-of-plane magnetic field dependence of (a) the longitudinal sheet resistance $R_{xx}$ and (b) the Hall resistance $R_{xy}$ for $t = 30$ QL at $T = 1.4$ K. The data (black) can be fitted with the two channel model (red) in good agreement as can be seen in the insets showing the residuals $\delta$ as defined in the main text. (c) The second derivative of the resistance with respect to the magnetic field $-d^2R_{xx}/dB^2$ plotted vs $1/B$. Beyond 16 T (0.062 T$^{-1}$) the oscillatory pattern (black) can be partially reconstructed from oscillations with $f_\alpha = 0.100 \pm 0.005$ kT, $f_\gamma = 0.14 \pm 0.01$kT, and $f_\beta = 0.219 \pm 0.005$ kT (red), but a clear deviation from the data is present. (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 depicted in (c). The FFT amplitude $A_{\text{FFT}}$ is plotted vs frequency $f$ where the curves are offset by 0.08 for clarity.

<table>
<thead>
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<th>Label</th>
<th>$f$ (kT)</th>
<th>$n_{\text{TSS}}$, $n_{\text{2D}}$ ($\times 10^{12}$/cm$^2$)</th>
<th>$n_{\text{2DEG}}$ ($\times 10^{12}$/cm$^2$)</th>
<th>$n_{\text{3D}}$ ($\times 10^{19}$/cm$^3$)</th>
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<tbody>
<tr>
<td>$\alpha$</td>
<td>0.092±0.006</td>
<td>2.2±0.2</td>
<td>4.5±0.3</td>
<td>0.50±0.05</td>
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<td>$\gamma$</td>
<td>0.13±0.01</td>
<td>3.2±0.3</td>
<td>6.4±0.5</td>
<td>0.9±0.1</td>
</tr>
<tr>
<td>$\beta$</td>
<td>0.23±0.01</td>
<td>5.5±0.3</td>
<td>10.9±0.5</td>
<td>1.9±0.2</td>
</tr>
<tr>
<td>$n_1$</td>
<td>15±1</td>
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<td></td>
<td>0.49±0.02</td>
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<tr>
<td>$n_2$</td>
<td>27±1</td>
<td></td>
<td></td>
<td>0.91±0.03</td>
</tr>
</tbody>
</table>

Table 5.4: Extracted values for $n_{\text{TSS}}$, $n_{\text{2DEG}}$, and $n_{\text{3D}}$ (with $k_{F,c} \approx k_{F,a}$) and $n_1$ and $n_2$ from the magnetoresistance (as can also be found in table 5.1 in section 5.3) for $t = 30$ QL.
Figure 5.8: (a) AFM image of sample with $t = 30$ QL from which we defined an error in the thickness of 1 QL. Clear contamination is visible. (b) Angular dependence of the frequency extracted from the obtained FFT spectra. (c) Temperature dependence of the oscillations. (d) Extracted cyclotron masses per peak position based on the oscillations in (c).

noise in the signals compared to the previously discussed samples, as displayed in figure 5.7c. This increase in noise might be linked to the contamination that we find from the AFM image (figure 5.8a). We find that this oscillatory pattern shows three spectral peaks but not as clearly resolvable as previous samples where the $2\alpha$ peak is missing, as shown in figure 5.7d. These three peaks can be used to fit the data, but a clear discrepancy is observed indicating that additional channels should be present beyond the experimental resolution. Interestingly, we observe that the appearance of the peaks commences at rather low fields when considering the mobilities that we find from the magnetoresistivity fitting, similar to that observed for $t = 20$ QL.

The presence of the peaks can be further checked by the angular dependence as plotted in figure 5.8b. Spectral peaks $\alpha$, $\beta$, and $\gamma$ show a clear angular dependence following a $1/\cos \theta$ behavior; again it is not possible to observe any remaining oscillation at $\theta = 90^\circ$. Comparing the values extracted from the oscillations and the magnetoresistance (table 5.4), we conclude that the $\alpha$ peak corresponds to the bulk channel with density $n_1$. The other channels contribute to a total charge-carrier density of about $(1.0 \pm 0.1) \times 10^{13}/\text{cm}^2$ which is on the same order of magnitude as $n_2$, thereby assuming that these states are linked to topological surface states. From the cyclotron mass analysis (figure 5.8d), we cannot see an evolution of $m_c$ because of the large errors and the limited number of peaks that could be analyzed due to the
Appendix

presence of a beating pattern.

Additional data for sample with \( t = 100 \) QL

In figure 5.9a, the data and fits for the out-of-plane field dependence of the sheet resistance \( R_{xx} \) and the Hall resistance \( R_{xy} \), respectively, for the sample with \( t = 100 \) QL are shown. Here, a disagreement is found between data and fit, which means that for this thickness the two-channel model is too limited to describe the magnetoresistance. Upon plotting \(-d^2R_{xx}/dB^2\) versus \(1/B\), we find a clear oscillatory pattern with an increased noise in the signals compared to the previously discussed samples (figure 5.9c). In figure 5.9d, we find that this oscillatory pattern contains

![Figure 5.9: Out-of-plane magnetic field dependence of (a) the longitudinal sheet resistance \( R_{xx} \) and (b) the Hall resistance \( R_{xy} \) for \( t = 100 \) QL at \( T = 1.4 \) K. The data (black) and the two channel model fit (red) show disagreement, especially for \( R_{xx} \) as also can be seen in the insets showing the residuals \( \delta \) as defined in the main text. (c) The second derivative of the resistance with respect to the magnetic field \(-d^2R_{xx}/dB^2\) plotted vs \(1/B\). Oscillations are clearly visible. Beyond 17 T (0.06 T\(^{-1}\)) the oscillatory pattern (black) can partially be reconstructed from oscillations with \( f = 0.110\pm0.005, 0.15\pm0.01, 0.175\pm0.005, \) and \(0.272\pm0.005\) kT (red). (d) Evolution of the (smoothed) FFT spectrum analyzed for different regions as depicted in (c) where the FFT amplitude \( A_{FFT} \) is plotted vs frequency \( f \).](image-url)

91
Figure 5.10: (a) AFM image (2×2µm²) of sample from where we defined an error in the thickness of 5 QL. (b) Angular dependence of the frequency extracted from the obtained FFT spectra for t = 100 QL. (c) Temperature dependence of the oscillations. (d) Extracted cyclotron masses per peak position based on oscillations (c).

<table>
<thead>
<tr>
<th>Label</th>
<th>f (kT)</th>
<th>n_{TSS}, n_{2D} (×10^{12}/cm²)</th>
<th>n_{2DEG} (×10^{12}/cm²)</th>
<th>n_{3D} (×10^{19}/cm³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>α</td>
<td>0.12±0.01</td>
<td>2.8±0.3</td>
<td>5.6±0.5</td>
<td>0.7±0.1</td>
</tr>
<tr>
<td>γ</td>
<td>0.188±0.003</td>
<td>4.55±0.08</td>
<td>9.1±0.2</td>
<td>1.46±0.04</td>
</tr>
<tr>
<td>β</td>
<td>0.25±0.02</td>
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<td>12±1</td>
<td>2.3±0.3</td>
</tr>
<tr>
<td>n₁</td>
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<td>27±2</td>
<td></td>
<td>0.33±0.02</td>
</tr>
<tr>
<td>n₂</td>
<td></td>
<td>42±2</td>
<td></td>
<td>0.42±0.02</td>
</tr>
</tbody>
</table>

Table 5.5: Extracted values for n_{TSS}, n_{2DEG}, and n_{3D} (with k_{F,c} ≈ k_{F,a}) and n₁ and n₂ from the magnetoresistance (as can also be found in table 5.1 in section 5.3) for t = 100 QL.

three main spectral peaks and a weak harmonic 2α peak. By reconstructing the oscillatory pattern, the four given frequencies are not sufficient enough, which indicates more channels might be present but are beyond our experimental resolution. A hint for an additional peak is given by the presence of an additional trace in the angular dependence as shown in figure 5.10b at a frequency f = 0.046 kT. However, this was considered to be an artifact in the FFT analysis because of the unclear temperature dependence in figure 5.3d in section 5.3.

Due to the beating features in the oscillation pattern (figure 5.10c), it is difficult to
extract the cyclotron mass for this sample, as shown in figure 5.10d. Nevertheless, we extract a cyclotron mass \( m_c = 0.15 m_e \) which increases to 0.20 \( m_e \) after 15 T, which is in agreement with the trend as observed for \( t = 10 \) QL. At last, we show in table 5.5 the extracted charge-carrier densities where we observe that any extracted bulk value from the oscillations is larger than \( n_1/t \) and \( n_2/t \). As mentioned already in section 5.3, the charge-carrier density from \( f_\alpha = 0.12 \) kT could account for \( n_1 + n_2 \). One uncertainty is the effective thickness \( t \) of the sample as shown in the AFM image (figure 5.10a), where large triangular undulations are present at the surface. Nevertheless, the exact reason for the disagreement between magnetoresistance measurements and the SdH oscillations analysis is yet unclear.

5.5.2 Derivation of the two-carrier model

As observed in the longitudinal magnetoresistance \( R_{xx} \) and the Hall resistance \( R_{xy} \) in section 5.3, we have parallel conduction channels with different charge-carrier densities and mobilities. Because of the limited dataset, we can only approximate the observed magnetoresistance with two parallel channels. In this section, a derivation of the equations as used in section 5.3 will be given. The steady-state Drude equation of motion of charge carriers in a material with effective mass \( m^* \) and charge \(-e\) is given by:

\[
\frac{m^*v}{\tau} = -e (E + v \times B),
\]

where \( \tau \) is the scattering time, \( v \) the drift velocity, \( E \) and \( B \) the applied electric and magnetic field, respectively. Now, the charge carriers are subject to an electric field \( E = (E_x,E_y,0) \) and a magnetic field \( B = (0,0,B) \). This yields for \( v = (v_x,v_y,v_z) \):

\[
v_x = -\frac{e\tau}{m^*}(E_x + v_y B), \tag{5.6a}
\]
\[
v_y = -\frac{e\tau}{m^*}(E_y - v_x B), \tag{5.6b}
\]
\[
v_z = 0 \tag{5.6c}
\]

Substituting (5.6b) into (5.6a) yields

\[
v_x = -\frac{e\tau}{m^*} \left[ E_x - \frac{e\tau B}{m^*} (E_y - v_x B) \right], \tag{5.7a}
\]

which gives

\[
v_x = \frac{-\frac{e\tau}{m^*} E_x + \left( \frac{e\tau}{m^*} \right)^2 B E_y}{1 + \left( \frac{e\tau B}{m^*} \right)^2} = -J_x/\nu e, \tag{5.7b}
\]

and can be rewritten as

\[
J_x = \sigma_{xx} E_x + \sigma_{xy} E_y = \frac{\nu e \mu}{1 + (\mu B)^2} E_x - \frac{\nu e \mu^2 B}{1 + (\mu B)^2} E_y, \tag{5.7c}
\]

93
Charge transport under high magnetic fields in Bi$_2$Se$_3$

where the mobility $\mu$ is given by:

$$\mu = \frac{e\tau}{m^*} \quad (5.7d)$$

By substituting the result of (5.7b), we find for $v_y$:

$$v_y = -\frac{e\tau}{m^*} \left\{ E_y - B \left[ \frac{-\frac{e\tau}{m^*} E_x + B \left( \frac{e\tau}{m^*} \right)^2 E_y}{1 + \left( \frac{e\tau}{m^*} \right)^2 B^2} \right] \right\}, \quad (5.8a)$$

which can be simplified to

$$v_y = -\frac{e\tau}{m^*} E_y - \frac{(e\tau/m^*)^2 B E_x}{1 + (e\tau/m^*)^2 B^2} = \frac{-J_y}{ne} \quad (5.8b)$$

and yields

$$J_y = \sigma_{yy} E_y + \sigma_{yx} E_x = \frac{ne\mu}{1 + (\mu B)^2} E_y + \frac{ne\mu^2 B}{1 + (\mu B)^2} E_x \quad (5.8c)$$

From this result, we can now construct the tensor $\sigma'$ for two parallel resistance channels, i.e. two conductance channels in series:

$$\sigma = \begin{pmatrix} \sigma'_{xx} & \sigma'_{xy} \\ \sigma'_{yx} & \sigma'_{yy} \end{pmatrix} = \begin{pmatrix} \sigma'_{xx} & \sigma'_{yx} \\ \sigma'_{xy} & \sigma'_{yy} \end{pmatrix} \quad (5.9a)$$

with

$$\sigma'_{xx} = \frac{n_1 e \mu_1}{1 + (\mu_1 B)^2} + \frac{n_2 e \mu_2}{1 + (\mu_2 B)^2} \quad (5.9b)$$

$$\sigma'_{yx} = \frac{n_1 e \mu_1^2 B}{1 + (\mu_1 B)^2} + \frac{n_2 e \mu_2^2 B}{1 + (\mu_2 B)^2} \quad (5.9c)$$

Now, the resistivity $\rho'$ and conductivity $\sigma'$ are related as $\rho' = \sigma'^{-1}$:

$$\rho = \begin{pmatrix} \rho'_{xx} & \rho'_{xy} \\ \rho'_{yx} & \rho'_{yy} \end{pmatrix} = \frac{1}{\sigma'^{2}_{xx} + \sigma'^{2}_{yx}} \begin{pmatrix} \sigma'_{xx} & \sigma'_{yx} \\ -\sigma'_{yx} & \sigma'_{xx} \end{pmatrix} \quad (5.10)$$

By accounting for the geometrical factors for a slab defined by $(l, w, t)$, we can relate the measured (sheet) resistances $R_{xx}$ and $R_{xy}$ to $\rho'_{xx}$ and $\rho'_{yx}$:

$$E_x = \frac{V_x}{l} = \rho'_{xx} J_x = \frac{\rho'_{xx} I_x}{wt}, \quad (5.11a)$$
Appendix

which gives

\[ \frac{\rho'_{xx}}{t} = \frac{R_{xx}w}{l} \]  \hspace{1cm} (5.11b)

and

\[ E_y = \frac{V_y}{w} = \rho'_{yx}J_x = \frac{I_x}{wt}, \]  \hspace{1cm} (5.11c)

which gives

\[ \frac{\rho'_{yx}}{t} = R_{xy} \]  \hspace{1cm} (5.11d)

Note: matrix indices are defined according to the basic rules of linear algebra, whereas indices for \( R_{xy} \) are inverted because in electronics the first index usually indicates the source direction and the second index the probe direction. And thus we find for sheet resistance \( R_{xx} \) and Hall resistance \( R_{xy} \):

\[ R_{xx} = \frac{1}{t} \frac{1}{\left( \frac{n_1e\mu_1}{1+(\mu_1B)^2} + \frac{n_2e\mu_2}{1+(\mu_2B)^2} \right)^2 + \left( \frac{n_1e\mu_1^2B}{1+(\mu_1B)^2} + \frac{n_2e\mu_2^2B}{1+(\mu_2B)^2} \right)^2}, \]  \hspace{1cm} (5.12a)

\[ R_{xy} = -\frac{1}{t} \frac{1}{\left( \frac{n_1e\mu_1}{1+(\mu_1B)^2} + \frac{n_2e\mu_2}{1+(\mu_2B)^2} \right)^2 + \left( \frac{n_1e\mu_1^2B}{1+(\mu_1B)^2} + \frac{n_2e\mu_2^2B}{1+(\mu_2B)^2} \right)^2}, \]  \hspace{1cm} (5.12b)

where the negative sign for \( R_{xy} \) indicates that the charge carriers are electrons. In the measurements, the voltage probe contacts have been inverted in such a way that this negative sign can be removed for the analysis. In our fitting procedure we used equations (5.12) but without the correction for \( t \) such as to obtain the 2D charge-carrier densities \( n_i \). However, the choice of 2D or 3D does not influence the final result of extracted mobilities and charge-carrier densities. Simultaneous fitting is done via Matlab R2016a by minimizing the sum of errors between data and fit of both \( R_{xx} \) and \( R_{xy} \) without any weighing. Here, the weak antilocalization (WAL) feature observed close to zero field does not affect the fitting procedure since the range over which this is observed is less than 1% of the total field range. Furthermore, at elevated temperatures where WAL is absent, the Drude model still shows a good agreement with the data and therefore we can rule out any fitting errors due to the presence of WAL. In this way, the fitting is most reliable due to inclusion of the full data set at once.

5.5.3 Details on analysis of oscillations

In this section, we would like to briefly elaborate on the analysis procedure for the Shubnikov–de Haas oscillations. In order to decouple the oscillations from the strong varying background, we have taken the second derivative \(-d^2R_{xx}/dB^2\) since from
equation (5.12) we have a $B^2$ dependence on the resistance in the limit of high field. By taking the second derivative, we are indeed successful to remove the background completely, whereas the first derivative still shows a strong residual background originating from the original background, which makes the FFT analysis difficult.

Taking the second derivative requires a low noise level which can be realized by adjacent averaging of the data. The averaging procedure is performed over 0.5 T intervals which are much shorter compared to the oscillation period such that this will not affect the FFT analysis; it only will slightly change the oscillation amplitude. Furthermore, the second derivative requires equidistant intervals and this is done by interpolation where the number of points is kept constant with respect to the original data. This interpolation is further used before the FFT is taken (in the $1/B$ range), which does not yield any artifacts because of the relatively large oscillation period in these measurements.

### 5.5.4 Improvement of the FFT spectra

The sampling range $\Delta 1/B_s$ in which the oscillations are observed limits the resolution in which separate peaks in a FFT spectrum can be resolved [45]. This resolution is denoted as waveform frequency resolution $\Delta R_{\text{WFR}} = B_s$, which can be described as the fundamental minimum spacing between two adjacent frequency spectral points that can be resolved. Furthermore, we can define the FFT resolution $\Delta R_{\text{FFT}} = f_s/N_{\text{FFT}}$ which defines the spacing between two data points in an FFT spectrum where $f_s$ is the sampling frequency and $N_{\text{FFT}}$ the number of data points included for the FFT analysis. In the normal case, these two resolution definitions yield the same number.

![Figure 5.11: The effect of zero padding.](image)

Figure 5.11: The effect of zero padding. (a) Zero-padded signal: the data has been cut off at 0.15 T$^{-1}$ and zeros have been added between 0 and 0.4 T$^{-1}$. (b) Comparison of FFT spectra of original signal for $t = 100$ QL (black) and the zero-padded signal (red) as shown in (a).

However, we can ‘improve’ $\Delta R_{\text{FFT}}$ by a process called ‘zero padding’ where zeros are included to the data sets as to increase $N_{\text{FFT}}$. This process yields an improvement of the peak shapes to for example determine the peak position more accurately.
Nevertheless, it will not allow one to observe additional frequencies because this is fundamentally limited by $\Delta R_{WFR}$. As an example, we show the effect of zero padding on data obtained for $t = 100$ QL as shown in figure 5.11. Although we observe a smoother spectrum, the amplitude changes by a factor of 4.

Figure 5.12: (a) Generated cosine waveform with $f = 5$ Hz, amplitude $A = 1$, sampling frequency $f_s = 100$ Hz over a sampling range of 0.5 s. (b) Comparison of FFT (black) and MEM PSD (red) spectra of signal in (a). (c) Dependence order $M$ on MEM PSD spectrum. (d) Comparison of FFT and MEM PSD for oscillation data for $t = 10$ QL.

If one would want to resolve frequencies that are closer separated than $\Delta R_{WFR}$ and could not perform the measurements at higher fields, one has to predict how the oscillations would evolve outside the sampling range (as to increase $B_s$). Motivated by the work of Terashima [46], analysis via the Maximum Entropy Method (MEM) can be used as a solution to increase the resolution where this method estimates how the data evolves outside the sampling range and in this way finer spectral peaks can be obtained. For our case, we approximated the autoregressive coefficients by the Yule–Walker method which is included in the Matlab package. The order $M$ has been chosen such that there is a good agreement with the FFT spectrum and $M$ is usually well below the safe maximum of $N/2$ with $N$ the number of data points ($M \sim 0.35 \times N/2$). Notably, the peak heights in the FFT and MEM spectrum cannot be compared. Only the peak positions can be determined since the MEM spectrum displays the power spectral density (PSD) instead of amplitude. In order to demonstrate this procedure,
we have analyzed the FFT and MEM PSD spectra of a cosine wave with $f = 5$ Hz, amplitude $A = 1$, sampling frequency $f_s = 100$ Hz over a sampling range of 0.5 s (see figure 5.12a). As shown in figure 5.12b, we observe that PSD gives a sharp spectral feature around 5 Hz compared to the FFT spectrum. However, as shown in figure 5.12c, we observe that the exact peak position is strongly dependent on the order $M$ which makes the analysis problematic.

Furthermore, we have compared the PSD and FFT spectrum of an oscillatory pattern measured for $t = 10$ QL as shown in figure 5.12d (the smoothed version of the FFT spectrum can be found in figure 5.1). The obtained PSD spectrum by the MEM analysis with an optimized $M = 500$ (with $N = 1560$) shows a good agreement with the FFT spectrum, where the MEM PSD shows slightly sharper spectral peaks. The small difference between both methods tells us that the FFT analysis performed already describes the oscillatory pattern very well and that extrapolation via this method does not contribute much.

### 5.5.5 Negative magnetoresistance

From the angular dependence, we have observed negative magnetoresistance (NMR) features when the sample is oriented at $\theta = 90^\circ$. More specifically, these features are only observed when the in-plane magnetic field is oriented parallel to the current bias direction. Because of the specific Hall bar geometry that we used, the magnetoresistance can be measured both for $I \perp B$ and $I \parallel B$ simultaneously, but has not been measured for $t = 10$ QL. As an example, the $MR = [R_{xx}(B) - R_{xx}(0)]/R_{xx}(0)$ for $t = 30$ QL has been plotted for both $I \perp B$ and $I \parallel B$ in figure 5.13a where a clear difference for the different orientations is present. The curves both show a clear WAL feature at low field (see section 5.5.6) whereafter the curves saturate in different directions. Furthermore, this NMR feature has a strong temperature dependence, as shown in figure 5.13b. Here, WAL disappears and the NMR increases at higher temperatures. Interestingly, an opposite trend is observed for $t = 100$ QL (figure 5.13c) where the NMR decreases with increasing temperature. Another deviation from the observed trend for $t = 30$ QL is seen for $t = 20$ QL (figure 5.13d), where the magnetoresistance of $I \parallel B$ is different from $I \perp B$, but still positive. Furthermore, the data for low temperatures show strong undulations compared to those at higher temperatures.

Similar NMR features have been observed in the same material earlier by our collaborators [24], other groups [47] and in clean metallic systems [48]. From these extensive works and related theoretical works [49, 50], the current understanding is that this NMR effect is rather related to scattering than specific properties of the band structure (NMR has also been proposed for the axial anomaly in Weyl semimetals, see also [51] for alternative scenarios for the topological semimetal Cd$_3$As$_2$). It is important to realize that the observed features are different from experimental [52–54] and theoretical works [55] in which NMR features are observed for both $I \parallel B$ and $I \perp B$, which are related to the spin texture of the surface states. Dependence on the scattering strength in the system might explain the partial absence of NMR features in our measurements since the measured mobilities are different from sample to sample. However, the current dataset is too limited to make definite conclusions.
on the exact origin of this effect as could be done through the extensive studies from previous works cited here.

### 5.5.6 Weak antilocalization

WAL is a manifestation of phase coherence of charge carriers in a material. For the topological surface states, Dirac fermions gain a $\pi$ Berry phase when the charge carriers finish a closed trajectory (see section 2.2). This additional phase will lead to destructive interference at the original position which reduces the probability of the fermion to localize [56]. The phase of the charge carriers is usually tuned by an applied magnetic field which can affect the obtained $\pi$ Berry phase of the Dirac fermions in such a way that the localization can be increased. Therefore, the conductance decreases (resistance increases) upon increasing magnetic field and this effect is known as weak antilocalization.

The theoretical framework for this effect in 2D systems has been established by Hikami et al. [57] in which an analytic expression for the change in conductance
\( \Delta \sigma_{xx}(B) \) due to WAL is given:

\[
\Delta \sigma_{xx}(B) = -\frac{Ae^2}{\pi \hbar} \left[ \ln \left( \frac{1}{\tau_e a} \right) - \Psi \left( \frac{1}{2} + \frac{1}{\tau_e a} \right) \right],
\]

(5.13a)

where \( a \) is given by

\[
a = \frac{4DeB}{\hbar}.
\]

(5.13b)

Plugging this into the first equation:

\[
\Delta \sigma_{xx}(B) = -\frac{Ae^2}{\pi \hbar} \left[ \ln \left( \frac{\hbar}{4eL_{\phi}^2 B} \right) - \Psi \left( \frac{1}{2} + \frac{\hbar}{4eL_{\phi}^2 B} \right) \right],
\]

(5.13c)

where \( \tau_e \) is the energy relaxation time, \( \Psi \) the Digamma function, \( D \) the diffusion coefficient and \( L_{\phi} \) the phase coherence length which is similar to the diffusion length since spin–orbit scattering plays an important role in our system. \( A \) is a coefficient related to scattering events that can affect the phase and can ideally have three values: 0 in case of the presence of strong magnetic scattering, 1/2 in the case of spin–orbit scattering, and 1 in the case of absence of any scattering events or the presence of two channels with \( A = 1/2 \). The parameter \( A \), often referred in literature as \( \alpha \), can be tuned by gating [58] and varies depending on the sample quality and thickness [11, 38, 59–62].

From the measurements presented in this chapter, we find clear WAL features for \( t = 10, 20, \) and 30 QL, whereas for \( t = 100 \) QL such features are absent. This absence could be related to the low data point density for this sample when taking into account the sharp feature for this thickness as observed by our collaborators [61]. The low point density makes the fitting of equation (5.13) to the data difficult. Nevertheless, we find \( L_{\phi} \) between 150 and 170 nm and \( A \) of about 0.25, as shown in figure 5.14. The obtained values for \( L_{\phi} \) and considering the thickness \( t \) indicate that charge carriers can scatter from one surface to the opposite surface through the bulk states diffusively with the phase information being retained [26]. The interplay between the different states is further displayed in the extracted values for \( A \) that do not resemble the expected (multiples of) 0.5. This indicates that the system is not fully 2D and bulk effects come into play [58]. In that sense, the findings from WAL are in agreement with the information from the SdH oscillations. However, since the WAL was not the main focus of this study and therefore the dataset is limited, we will be careful with making definitive statements on the outcome of the WAL analysis.
Figure 5.14: WAL features for (a) $t = 10$ QL, (b) $t = 20$ QL, and (c) $t = 30$ QL. Fits are given by the red line.
5.6 References


References


