The hole Fermi surface in Bi$_2$Se$_3$ probed by quantum oscillations

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Transport and torque magnetometry measurements are performed at high magnetic fields and low temperatures in a series of p-type (Ca-doped) Bi$_2$Se$_3$ crystals. The angular dependence of the Shubnikov-de Haas and de Haas-van Alphen quantum oscillations enables us to determine the Fermi surface of the bulk valence band states as a function of the carrier density. At low density, the angular dependence exhibits a downturn in the oscillations frequency between $0^\circ$ and $90^\circ$, reflecting a bag-shaped hole Fermi surface. The detection of a single frequency for all tilt angles rules out the existence of a Fermi surface with different extremal cross-sections down to 24 meV. There is therefore no signature of a camel-back in the valence band of our bulk samples, in accordance with the direct band gap predicted by $GW$ calculations.

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I. INTRODUCTION

Bi$_2$Se$_3$ is a narrow gap layered semiconductor which together with Bi$_2$Te$_3$ have been studied for decades for their thermo-electric properties.$^{11}$ The interest in this class of materials has recently surged because of the prediction$^{1}$ and observation$^{2}$ of a unique type of charge carriers existing at their surface, the so-called “helical Dirac fermions”, which behave as massless relativistic particles with a spin locked to their translatational momentum. Bi$_2$Se$_3$ therefore now belongs to the 3D topological insulators family characterized by a bulk gap coexisting with 2D conducting surface states. As a matter of fact, the existence of gapless states at the boundary of the material is related to a well defined change in the bulk band structure. In Bi$_2$Se$_3$, this originates from a parity inversion of the valence and conduction bands at the $\Gamma$ point of the Brillouin zone in the presence of a large spin orbit coupling.$^{2}$ The linear-in-momentum dispersion relation which characterizes the 2D surface states thus emerges from the general Hamiltonian of massive Dirac fermions$^{2}$ theoretically expected to describe the bulk states in Bi$_2$Se$_3$.

Pioneering experimental studies of the bulk conduction band at low energy$^{2}$ have reported an ellipsoidal electron Fermi surface, which was described within a simple model of massive carriers with a parabolic (non-parabolic) dispersion in the $k_|$ ($k_{||}$) direction, where $k_|$ ($k_{||}$) is the momentum in the direction perpendicular (parallel) to the $c$-axis of the crystal. This is accompanied by an increasing anisotropy of the Fermi surface observed as the Fermi level increases in the conduction band. More recent transport$^{5}$ NMR$^{6}$ and magneto-optics$^{10}$ measurements have confirmed the original parameters phenomenologically describing the bulk conduction band, and in some cases$^{10}$ connected them to the 3D Dirac Hamiltonian for massive fermions applied to topological insulators.

However, experimental studies of the valence band bulk Fermi surface are to our knowledge scarce. The principal reason is that as-grown Bi$_2$Se$_3$ is electron-doped due to the presence of Se vacancies. The discovery of 2D surface states has nevertheless triggered large efforts to reach the topological insulator regime, where the Fermi level lies in the band gap of the bulk band structure. For instance, substituting trace amounts of Ca$^{2+}$ for Bi$^{3+}$ in as-grown Bi$_2$Se$_3$ can lower the Fermi energy of the native n-type crystals. Above a certain value of Ca-doping $\delta$, the electrical conduction in Bi$_2-\delta$Ca$_{\delta}$Se$_3$ is supported by hole carriers rather than electrons.$^{10,11}$ Further doping brings the Fermi level down in the previously inaccessible valence band. Very recently, Shubnikov-de Haas (SdH) measurements have been reported$^{13}$ in p-type Bi$_2$Se$_3$ samples with hole concentrations estimated between $5.7 \times 10^{18}$ cm$^{-3}$ and $1.6 \times 10^{19}$ cm$^{-3}$. A bag-like closed Fermi surface was observed at low concentration, with the suggestion of open tubes appearing in the Fermi surface at high carrier density. In spite of these first experimental advances, and several theoretical works,$^{12,13,14,15}$ the low-energy details of the valence band are still not unambiguously determined. In particular, a local minimum was suggested to form at the $\Gamma$ point as a consequence of the spin-orbit coupling. While a camel-back structure is observed in the valence band near the surface$^{12}$ it is absent in some bulk measurements$^{10}$ More recent GW calculations$^{15,16}$ show that the electron-electron interac-
In the parabolic band approximation, is obtained by
nal warping. The apparent hole effective mass, defined
at high energy are confirmed and attributed to trigono-
lar dependence of the oscillations frequency between
low Fermi energies, a downturn is observed in the an-
doped Bi$_2$Se$_3$ crystals are studied by magneto-transport
and torque magnetometry at low temperatures and un-
der magnetic fields up to 30 T. A high resolution angular
dependence of the quantum oscillations (both Shubnikov-
de Haas (SdH) and de Haas-van Alphen (dHvA)) en-
ables us to map out the Fermi surface of the bulk valence
band states, in the energy range $E_F \sim 20-60$ meV. At
low Fermi energies, a downturn is observed in the an-
gular dependence of the oscillations frequency between
$0^\circ$ and $90^\circ$, demonstrating a bag-shaped closed Fermi
surface. Importantly, a single frequency dominates the
FFT spectra regardless of the magnetic field orientation,
showing that no camel-back structure is observed for en-
ergies down to $\sim 24$ meV. The existence of a camel-back
structure for lower energies is hardly probable in respect
to the experimental $E(k)$ dependence, which points to
a direct band gap. The Fermi surface anisotropy in-
creases rapidly as the Fermi level goes higher in the va-
ence band, and pipe-like structures previously reported
at high energy are confirmed and attributed to trigonal
warping. The apparent hole effective mass, defined in
the parabolic band approximation, is obtained by tempera-
ture-dependent studies for $B \parallel c$-axis, and lies in
the $0.245 \pm 0.015 m_0$ range for $E_F \sim 23 - 45$ meV. High
magnetic fields measurement in the lowest density sam-
ple enables us to approach the quantum limit for holes
which is finally discussed.

II. QUANTUM OSCILLATIONS IN P-TYPE
Bi$_2$Se$_3$

A. Experimental details

The Bi$_{2-x}$Ca$_x$Se$_3$ samples studied here were grown via
a process of two-step melting described in Ref. 11. By
adding Ca, a transition to a p-type behavior is observed for
$x > 0.012$ [12] which is the regime our study focuses on.
The samples presented here are referred to as B2, B3, B6, E2, and E1, and their main characteristics are
summarized in Table I. Magneto-transport experiments
were conducted on $\mu$m-thick slices on which silver paste
contacts were deposited in a Hall bar-like configuration.
Measurements were performed using a standard low fre-
quency lock-in technique in a variable temperature insert
for temperatures ranging from 1.2 K to 40 K, up to mag-
netic fields of 30 T produced by a 20 MW resistive mag-
net. The data were initially symmetrized by changing the
polarity of the magnetic field to check that contact misalignement had a negligible impact on the analysis (no-
tably the oscillation frequency). For dHvA torque mea-
surements, samples of thickness varying between 40 and
230 $\mu$m were mounted on a CuBe cantilever which forms
the mobile plate of a capacitive torque meter. The torque
signal was measured with a lock-in amplifier and a capac-
tance bridge using conventional phase sensitive detection
at 5.3 kHz. The experiment was performed using a 16 T
superconducting magnet and a dilution fridge, equipped
with an in-situ rotation stage. The torque measurements,
which typically probe a larger number of particles, were
employed to further confirm the domination of the bulk
states in the present study.

B. Magnetic field dependence

Figure 1 shows representative quantum oscillations in
the valence band for the two extreme carrier densities
studied. In Fig. 1(a), we report on magneto-transport
(SdH) data on sample B3, one of our lowest density sam-
bles. Superimposed on a large monotonic background,
oscillations of the resistivity reflect the oscillatory density
of states of the system. The data in the upper left inset
are obtained by subtracting a smoothed (moving window
average) data curve. The resulting FFT is shown in the
bottom right inset. It shows a clear single peak at about
55 T, consistent with the apparent absence of splitting in
the data.

dHvA oscillations are shown for a sample with a higher
carrier concentration (sample E1) in Fig. 1(b), where the
torque signal $\tau$ is plotted as a function of the total mag-
netic field for a tilt angle $\theta = 16^\circ$. Small quantum os-
cillations in $\tau$, again on a large monotonous background,
are clearly visible and reflect the oscillatory magnetiza-
tion of the system. The dHvA oscillations can be better
observed in the oscillatory torque ($\Delta \tau$), obtained with
the same background removal procedure as for transport.
Here also a single frequency is observed, at a higher value
consistent with the higher carrier concentration of sample
E1. We note that the results obtained with the two dif-
ferent experimental techniques for a given carrier density
are fully consistent.

| Ca doping ($\delta$) | Expt F (T) | $m_h$ ($m_0$) | $E_F$ (meV) |
|--------------------|-----------|--------------|------------|
| B2 0.015           | SdH 48.7  | 0.238(0.01)  | 23.7       |
| B3 0.015           | SdH 55.8  | 0.248(0.01)  | 27.15      |
| B6 0.015           | dHvA 87.8 | -            | -          |
| B1 0.015           | SdH 96    | 0.249(0.01)  | 44.8       |
| D1 0.025           | SdH 97    | 0.249(0.01)  | 45.1       |
| E2 0.03            | dHvA 100.8| -            | 46.9       |
| E1 0.03            | dHvA 126  | -            | 58.6       |

TABLE I: Parameters of the Bi$_2$Se$_3$ samples. Calcium doping level $\delta$ defined by Bi$_{2-x}$Ca$_x$Se$_3$, experimental technique used, quantum oscillation main frequency, apparent valence band effective mass ($m_h$) and Fermi energy in the parabolic band approximation.
C. Temperature dependence

The temperature dependence of the SdH oscillations was measured for samples B1, D1, B2 and B3 between 1.3 K and 40 K for magnetic fields up to 30 T. In Fig. 2, we report typical results obtained on samples B1 and B2 up to 11 T, for temperatures between 1.3 K and 25 K. In the upper left inset, we plot the temperature dependence of the oscillation amplitude at the fixed magnetic field $B = 9.6$ T. The same type of data are reported in the lower left inset for a lower concentration sample (B2) at a similar magnetic field ($B = 10.6$ T). The amplitude of the oscillations has not fully saturated at $T = 1.3$ K, suggesting a higher hole effective mass compared to the well-documented n-type samples$^{6,9}$ in qualitative agreement with recent magneto-optics studies.$^{10}$ A non-trivial field dependence was observed as the magnetic field was increased, but is beyond the scope of the present paper where we aim at characterizing the (field-independent) hole Fermi surface. From the frequency $F$ of the quantum oscillations discussed in the previous section, one can deduce the extremal Fermi surface cross-section in the momentum space, $C_S = \frac{\pi k_F^2}{F}$ (where $k_F$ is the Fermi wave vector), given by $C_S = 2\pi eF/h$. In the case of an energy-independent effective mass in the $B \parallel c$-axis configuration, the Fermi energy can be written $E_F = (h^2C_S)/(2\pi m_h)$. Using this method, we have determined the Fermi energies reported in Table I which define the energy range ($E_F \sim 20$ to 60 meV) probed by our experiment.

III. HOLE FERMI SURFACE

A. Doping and angular dependences

In Fig. 3, we report the angular dependence of the FFT of the quantum oscillations in a color map for three samples with different hole concentrations.
FIG. 3: (color online) Color plot. Normalized amplitude of the quantum oscillations $1/B$ Fourier transform as a function of the frequency and the tilt angle $\theta$, for different samples. At each angle, the FFT signal is normalized to its maximal value (red color). The blue color corresponds to an intensity $\leq 70\%$ of the maximum value.

The displayed color map is a linear extrapolation of angular dependent data taken with a $5^\circ$ step, where $\theta$ is the angle between the sample $c$-axis and the magnetic field. The FFT signals have been normalized to their maximum value for each angle to focus only on the angular dependence. The frequency of the quantum oscillation is directly related to the cross-section of the Fermi surface in $k$-space. As the sample is rotated in the magnetic field, the angular dependence of this cross-section can be traced and related to the Fermi surface's geometry.

The overall observed non-monotonous behaviour of the frequency as a function of the tilt angle $\theta$ is characteristic of an anisotropic Fermi surface. At low energies ($E_F < 30 \text{ meV}$) the angular dependence is rather mild, suggesting an almost isotropic Fermi surface. Anisotropy progressively develops with increasing Fermi energy and, for a frequency of 120 T at $\theta = 0^\circ$, the $\theta = 90^\circ$ frequency approximatively doubles. It should be noted that a cosine behavior, usually associated with a 2D system, was not observed in any sample even up to very high magnetic fields. This confirms that bulk states dominate both the transport and magnetization properties in samples of relatively large thicknesses. It is worth stressing that, as the system becomes anisotropic at high doping level, a complete angular dependence ($0^\circ - 90^\circ$ at least) is absolutely required to probe the dimensionality of the system (see the example of graphite, a very anisotropic 3D system showing a $\cos \theta$ behavior up to $\sim 70^\circ$, in Ref. [23]).

At variance with previous results obtained on $n$-type Bi$_2$Se$_3$ in the low energy region of the conduction band ($E_F < 30 \text{ meV}$), the cross section cannot be reproduced accurately by assuming a purely ellipsoidal Fermi surface. A perhaps even more striking difference is the occurrence of a downturn in the angular dependence above a density-dependent angle (e.g. $\theta \sim 70^\circ$ for sample B6). This demonstrates a “bag-shape” Fermi surface, where the cross section increases until it reaches the bag’s diagonal axis from where it starts to decrease with further increasing the angle. A closer look at the low density data (sample B2) shows the downturn is also present on the apparently flat angular dependence (see section III B for a better representation). The intensity of the FFT signal (which can not be assessed in Fig. 3 because of the normalization) can to some extent also be informative. The raw FFT intensity reflects a severe drop in the oscillation amplitude where the downturn in frequency occurs for example around $\theta = 60^\circ$ for sample B2 and B3. The FFT signal then clearly reappears from $\theta = 75^\circ$ to $\theta = 90^\circ$ [23]. This drop in the amplitude may be correlated to the downturn in the angular dependence. The FS strong curvature change in this region probably leads to the loss of the phase coherence of the oscillations. We note that the downturn appears at higher angle for higher density. Finally, our data in the high energy limit (sample E1) are in good quantitative agreement with the previously reported SdH measurements in high hole density samples. In particular, oscillations are lost for specific $\theta$ ranges (see e.g. the sudden drop in frequency from $\theta = 90^\circ$), corresponding to open orbits in the momentum space. We attribute these open orbits to the pipes emerging in the Fermi surface at high carrier density in the presence of trigonal warping [25].

B. Theoretical model and discussion

In Fig. 4(a), we report the angular dependence of the quantum oscillation main frequency (FFT maximum) for different hole doping levels. These are the same data as the ones in Fig. 3 with additional samples at intermediate carrier concentrations. For each sample we computed the extremal cross-section of the Fermi surface in $k$-space from the $F(\theta)$ dependence. By assuming that the Fermi surface is closed and has rotational symmetry around the c-axis, the constant $k_x - k_z$ energy surface, where $k_z$ is parallel to the trigonal axis of the Brillouin zone (c-axis) and $k_\perp = (k_x^2 + k_z^2)^{1/2}$ is in the plane perpendicular to $k_z$, was derived (not shown) [23]. The $\theta$ angular dependence of the Fermi radius was then fitted by solving an analytical $E(k)$ dependence for each experimental Fermi energy reported in Table I. The empirical dispersion relation up
this “squashed” ellipsoid the in-plane Fermi wave-vector results from the bag-shape nature of the Fermi surface. In oscillation frequency is well reproduced, and clearly re-

At low energy (sample B2), the downturn of the energy bands along \( k_\perp \) and \( k_z \) obtained for the \( 4 \times 4 \) Hamiltonian of Zhang et al\(^\text{[23]} \) and its original parameters. The camel-back is prominent with a band maximum occurring at finite \( k_x \approx 0.07 \AA^{-1} \) and a \( k_x = 0 \) depth varying in theoretical works from about 80 meV\(^\text{[22]} \) to 140 meV\(^\text{[23]} \).

If the Fermi energy lies in the camel-back region, there should be no carriers around \( k_\perp = 0 \) and the Fermi surface should exhibit a “donut-like” shape. This should give two frequencies corresponding the the outer (maximal) and inner (minimal) orbits. Our observation of a single frequency around \( \theta = 0^\circ \) suggests that there is no “camel-back” structure deeper than the lowest energy investigated (\( \sim 23.7 \) meV). Let us now consider the case where the residual “camel back” depth is lower than \( \sim 23 \) meV. For our lowest Fermi energy, one would expect a single oscillation frequency at low angles, splitting into two frequencies above a certain angle. The low angle frequency would correspond to an orbit along the horizontal cross section of a “dumbbell-like” Fermi surface, and the two frequencies at high angles would correspond to the two extremal transversal section of the dumbbell. The size of the frequency splitting at \( \theta = 90^\circ \) is in this case related to the depth of the “camel-back” structure. At variance with these expectations, our measurements reveals that a single oscillation frequency persists up to \( \theta = 90^\circ \). This shows that no camel back structure can be observed at all within our experimental resolution (our FFT full-width-half-maximum at \( \theta = 90^\circ \) is \( \Delta F \sim 13 \)T).

\( F(\theta) \) dependencies for the \( 4 \times 4 \) Hamiltonian of Zhang et al\(^\text{[23]} \).

\( E(k) = C_1 k_\parallel^4 + C_2 k_\parallel^2 + C_3 k_\perp^4 + C_4 k_\perp^2 + C_5 k_z^4 + C_6 k_x (k_\parallel^2 - 3k_z^2) + C_7 k_\perp k_y (k_\parallel^2 - 3k_z^2) \) \quad (1)

From symmetry considerations, \( E(k) = E(-k) \), which sets \( C_6 = 0 \). Neglecting non-parabolicity effects at low density leads to \( C_4 = 0 \) and \( C_5 = 0 \). The last term accounts for the trigonal warping of the Fermi surface, which we neglect in our first approach. The simplified expression we used can therefore be written as:

\( E(k) = C_1 k_\parallel^4 + C_2 k_\parallel^2 + C_3 k_\perp^4 \) \quad (2)

where the numerical values of the coefficients \( C_1 \), \( C_2 \) and \( C_3 \) are listed in the supplemental material, section III. The theoretical \( F(\theta) \) dependencies are plotted as solid lines on top of the experimental points in Fig. 4(a) and three computed Fermi surfaces are shown in Fig. 4(b). At low energy (sample B2), the downturn of the oscillation frequency is well reproduced, and clearly results from the bag-shape nature of the Fermi surface. In this “squashed” ellipsoid the in-plane Fermi wave-vector is longer than along the \( c \)-axis, leading to a negative anisotropy factor\(^\text{[25]} \).

FIG. 4: (color online) (left panels) Quantum oscillation frequency as a function of the tilt angle \( \theta \) for different carrier concentrations in the valence band. The error bar is defined to be the FFT width at 90% of the maximum signal. From bottom to top, sample B2 (SdH), B3 (SdH), B6 (dHvA), E2(dHvA) and E1(dHvA). Theoretical fits obtained by the analytical model of Eq.2 (see text) (solid lines). (Right panels) Fermi surfaces computed at \( E_F = 23.7 \) meV, \( 42 \) meV, and \( 58.6 \) meV.

FIG. 5: (color online)(a) and (b) Valence band along \( k_{x,y} \) and \( k_z \) respectively. Experimental data (dots), parabolic dispersion (solid lines) and camel-back model (dotted lines).
The absence of the camel back structure can more generally be seen in figure 5(a), where the experimental points $E_F$ vs $k_z$ are plotted, showing that camel-back structure is experimentally absent. The $k_F^2$ approximation used in Eq. 2 is justified by the good parabolic fit (red solid line) and the almost energy-independent mass measured experimentally with the temperature-dependent mass $m_h = 0.24m_0$. In Fig. 5(b), we plot the experimental dispersion along $k_z$ obtained from our simulations. Importantly, the $k_z$ dispersion is far from being parabolic, $E(k_z)$ showing a slower increase, in agreement with the observation made in previous ARPES measurements focusing on the $\Gamma - Z$ dispersion in the Brillouin zone. In contrast to the $k_z$ dispersion, higher order terms in the data analysis as it improved the $F(\theta)$ curves plotted in Fig. 4(a) only slightly.

Our main observation that no camel-back structure is formed in the Bi$_2$Se$_3$ valence band is in qualitative agreement with other experimental work probing the bulk band structure of this compound. This can be related to the recent conclusion of GW numerical computations that the band gap of Bi$_2$Se$_3$ is direct and reduced by the electron-electron interaction.

**IV. HOLE TRANSPORT TOWARDS THE QUANTUM LIMIT**

For the lowest density samples (B2 and B3), magneto-transport experiments have been extended to high magnetic fields up to 30 T. Typical results are reported in Fig. 6 for sample B3. Simulations of the magnetoresistance were performed based on the simple phenomenological approach developed for the conduction band in which one considers a 3D electron gas with a large (spin-orbit-enhanced) spin gap $g^*_e [fH_B]$, where $g^*$ is the effective hole $g$-factor. As can be seen in Fig. 6, the measured (background-removed) oscillations exhibit a minimum at $B \sim 22.5$ T, instead of the maximum expected from the $g^* = 0$ (no spin-splitting) simulation based on the low field oscillation frequency ($F = 55.8$ T). One could tentatively attribute this additional minimum to a spin-split feature only visible in the lowest Landau level/highest magnetic fields. The temperature dependence however shows that the energy gaps associated with the consecutive minima follow a continuous trend and have therefore most likely the same origin. This suggests that Landau levels of different orbital and spin quantum numbers nearly coincide in energy, with consequently no visible spin splitting in the quantum oscillations. A similar scenario is observed in n-type Bi$_2$Se$_3$ where the spin gap is about twice the value of the cyclotron gap. In such cases the quantum limit is repelled to higher magnetic fields due to a large field-dependent spin splitting shifting down the Landau levels energies.

In the present case, when using the effective hole mass $m_h = 0.24m_0$ determined in section III C, the best simulation is obtained for a $g$-factor of $g^* \sim 35$, for which spin-up and spin-down subbranches of the $N$ and $N + 4$ Landau level coincide. As can be seen in Fig. 6, the agreement with the data is not perfect (small dephasing can be seen at low magnetic fields) suggesting that some more complex field dependencies of the band parameters should be considered for a better description. The high value of the effective hole $g$-factor is nevertheless confirmed by simply analyzing the magnetoresistance peaks positions. The so-called “1/$B$ phase plots” show a severe deviation from linearity in high magnetic fields, which can be seen in the inset of Fig. 6, where we focus on the product $nB_n$ where $n$ is an integer and $B_n$ the magnetic field value corresponding to the $n$$^{th}$ SdH extremum. SdH oscillations extend to high ratio between the Fermi energy and the cyclotron gap ($\propto B$) equals $n/2$, and thus the product $nB_n$ is proportional to the Fermi energy value. The $y$-axis value of the blue circles in the inset of Fig. 6 is proportional to the measured $nB_n$ value and show that the Fermi energy is reduced in high magnetic fields. This deviation is consistent with the Fermi energy drop expected in the presence of a high Zeeman energy, which is simulated by our model (dashed line). These observations stress, again, how crucial the contribution of the Zeeman energy is when conducting a phase oscillation analysis in such materials, in particular when searching for the (phase) signature of 2D surface carriers. Our results suggest that the effective $g$-factors could be similar in the bulk conduction and valence bands, which should be confirmed by a more direct measurement in...
p-type samples.

In conclusion, we have reported an energy-dependent study of the Bi$_2$Se$_3$ valence band based on quantum oscillations measurements in high magnetic fields and low temperatures. At low energies, a downturn observed in oscillations measurements in high magnetic fields and low study of the Bi$_2$Se$_3$ p-type samples.

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