

Link between superconductivity and a Lifshitz transition in intercalated Bi_2Se_3

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Topological superconductivity is an exotic phase of matter in which the fully gapped superconducting bulk hosts gapless Majorana surface states protected by topology. Intercalation of copper, strontium or niobium between the quintuple layers of the topological insulator Bi_2Se_3 increases the carrier density and leads to superconductivity that is suggested to be topological. Here we study the electronic structure of strontium-intercalated Bi_2Se_3 using angle resolved photoemission spectroscopy (ARPES) and Shubnikov-de Haas (SdH) oscillations. Despite the apparent low Hall number of $\sim 2 \times 10^{19} \text{cm}^{-3}$, we show that the Fermi surface has a shape of an open cylinder with a larger carrier density of $\sim 10^{20} \text{cm}^{-3}$. We suggest that superconductivity in intercalated Bi_2Se_3 emerges with the appearance of a quasi-2D open Fermi surface.

A promising avenue towards achieving topological superconductivity is to induce superconductivity in materials whose electronic structure is topologically non-trivial. Intercalated Bi_2Se_3 has been suggested as a possible candidate for a topological superconductor based on the appearance of a zero-bias feature in the tunnelling conductance [1–4] and nematicity in the superconducting state [4–11]. A possible explanation to the zero bias peak is the existence of Majorana modes [1, 3, 4]. Also, nematicity can be accounted for by a two-component Ginzburg-Landau theory with nematic solutions corresponding to topological-superconductivity with odd-parity E_u symmetry (the order parameter is odd under the operation $x \rightarrow -x$ and $y \rightarrow -y$). Such nematic solutions require an open, quasi-2D, Fermi-Surface [12–15].

A simple criterion has been put forth for determining whether superconductivity in Bi_2Se_3 -derived systems is 3D topological time-reversal-invariant (TRI): the Fermi surface (FS) has to enclose an odd number of time-reversal-invariant-momenta (TRIM) points [16]. Superconducting $\text{A}_x\text{Bi}_2\text{Se}_3$ compounds are slightly electron-doped ($n \sim 10^{19} - 10^{20} \text{cm}^{-3}$) with the bottom of the conduction band located at the center of the Brillouin zone [17]. In $\text{Cu}_x\text{Bi}_2\text{Se}_3$, ARPES, Shubnikov-de Haas (SdH) and de Haas-van Alphen [18, 19] experiments found an open FS that encloses two TRIM points (Γ and Z) thus not fulfilling the aforementioned criterion but suggesting the possibility of weak 2D topological superconductivity.

Increasing the carrier density of Bi_2Se_3 via Cu intercalation results in a quick expansion of the FS towards the Z point and eventually a Lifshitz transition into an open cylindrical-like FS [18]. Interestingly, Sr intercalated Bi_2Se_3 is reported to have an order of magnitude smaller carrier concentration as compared to the Cu intercalation, while still exhibiting superconductivity with similar critical temperature T_c [6, 20, 21]. This raises two questions: 1) Does $\text{Sr}_x\text{Bi}_2\text{Se}_3$ have a closed Fermi surface

that allows for a 3D topological TRI superconductivity? 2) How can such a small amount of carriers produce a $T_c \simeq 3\text{K}$?

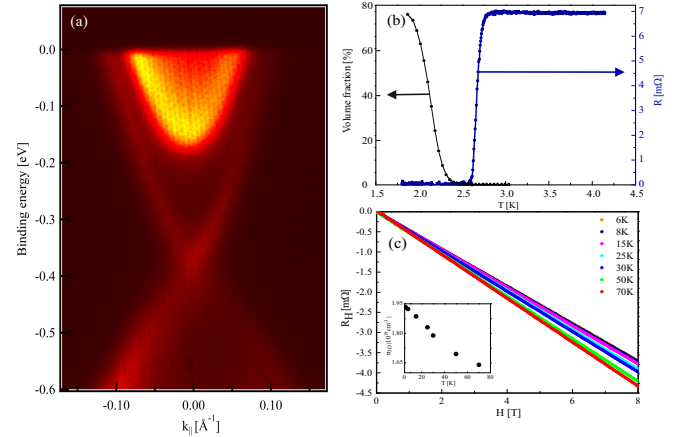


FIG. 1. Bulk superconductivity coexists with topological surface states in $\text{Sr}_x\text{Bi}_2\text{Se}_3$. (a) Typical ARPES spectra, taken using a 20 eV photon energy at $T = 28.5\text{ K}$, displays the electron-like band and the surface states. The Dirac point lies about 375 meV below the Fermi-level. (b) Resistance and magnetization measurements of the sample measured in the Shubnikov-de-Haas part. The sharp resistive transition at 2.7K, along with the large superconducting volume fraction, clearly demonstrate a 3D bulk superconductivity. (c) Hall measurements at various temperatures. The linearity of the Hall coefficient with respect to the magnetic field is visible, while a clear temperature dependence is apparent. Inset: The carrier density inferred from the Hall measurements vs. temperature.

In this letter we combine the SdH effect and ARPES measurements to determine the shape of the Fermi surface, motivated by the relatively small Hall number of $\text{Sr}_x\text{Bi}_2\text{Se}_3$. We study $\text{Sr}_x\text{Bi}_2\text{Se}_3$ samples with

$x_{\text{nominal}} = 0.15$, crystal growth and characterization are described in Ref. [6, 23] and in the supplementary. The samples display Dirac-like surface states as well as a parabolic bulk band. A naive estimate of the carrier density from the Hall slope yields a carrier concentration of $\sim 2 \times 10^{19} \text{cm}^{-3}$. This estimate is in good agreement with the binding-energy of the Dirac-point [18], but has a noticeable temperature dependence (see Fig.1). Bulk superconductivity is evident from the large SC volume fraction, which extrapolates to 100% at zero temperature, with $T_c = 2.7\text{K}$.

The frequency of the resistance-oscillations as a function of inverse magnetic field corresponds to the cross-sectional areas of the extrema of the FS perpendicular to the magnetic field [24]. We therefore rotate the magnetic field with respect to the c-axis (θ) to resolve the shape of the FS. Our main finding using this method is the clear observation of two frequencies when the magnetic field is parallel to the c-axis, $\theta = 0$ (see Fig. 2(a)).

This finding is a strong evidence for an open FS, as two frequencies will only show up if the FS have two extrema with different cross-sectional area. Therefore, A closed Fermi-surface would have two frequencies only for a complicated shape, which is inconsistent with previous ARPES measurements and theoretical DFT and $k \cdot p$ calculations [17, 25].

The two frequencies persist over a wide temperature range. However, as we increase θ only one frequency remains visible in the SdH data beyond 10° . The disappearance of the second frequency is probably due to a strong damping of the oscillations, possibly a consequence of different effective masses. The remaining frequency increases when rotating the field away from the c-axis, becoming unresolved for $\theta > 57^\circ$. This frequency is well described by $1/\cos(\theta)$, as expected for an open cylindrical-type FS (see Fig. 2).

Due to the larger uncertainties for $\theta > 50^\circ$, we found it necessary to support our analysis suggesting an open FS in $\text{Sr}_x\text{Bi}_2\text{Se}_3$. We turn to ARPES measurements in which we vary the photon energy and thus scan the entire Brillouin zone at different k_z [18, 26, 27]. The inner-potential that determines the conversion of photon-energy to momentum normal-to-the-surface is derived in the supplementary material.

Each panel in Fig. 3 displays the dispersion of the conduction band with respect to the momentum parallel to the surface for different photon energies. We find intensity corresponding to the bulk band for all measured k_z value, including the Γ and Z points. Our data showing finite density of states at both Z-points support our conclusion of open FS. For comparison, we note that no intensity is seen by ARPES at the k_z corresponding to the Z points for doped Bi_2Se_3 having a closed FS [18, 28]. The bandwidth changes with k_z and is maximal at the Γ -point, similarly to the case of $\text{Cu}_x\text{Bi}_2\text{Se}_3$ and consistent with our SdH data. Furthermore, the Fermi-momentum

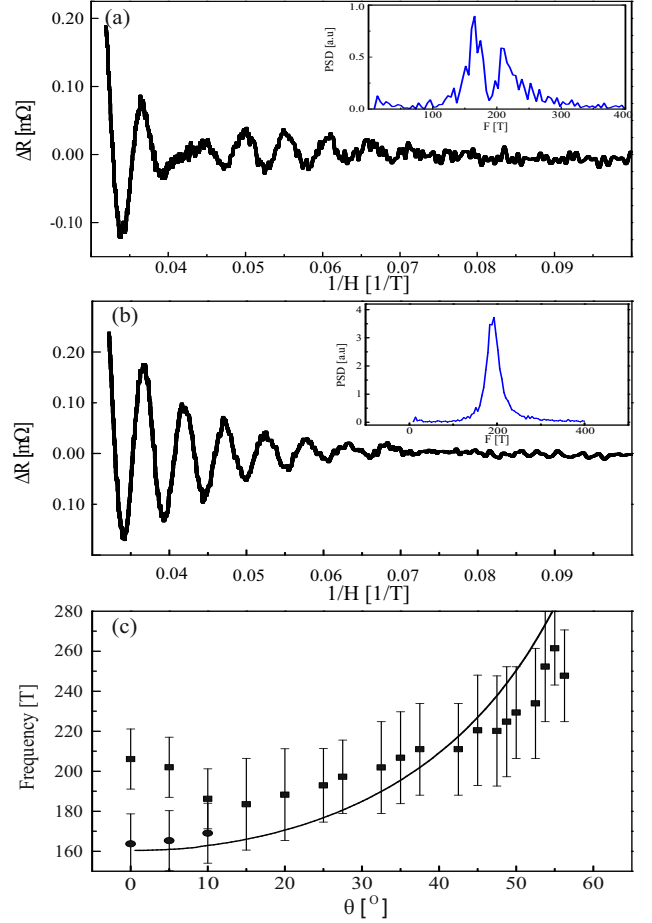


FIG. 2. Angular dependence of the SdH frequency at 0.4 K. (a) Two frequencies are observed for $\theta \simeq 0^\circ$. The two frequencies point to an open Fermi surface as explained in the text. (b) As we rotate the sample to larger out of plane angles, we observe only one frequency above a certain angle, as can be seen, for example, at $\theta \simeq 20^\circ$. (c) SdH frequency as a function of angle. The solid line represent the expected angular dependence of one SdH frequency for an open cylinder. The insets in (a) and (b) are the Fourier power spectrum of the quantum oscillations vs inverse magnetic field ($1/H$). θ is the angle between the magnetic field direction and the c-axis.

is independent of k_z , suggesting a nearly perfect cylinder. The constant Fermi-momentum in the $k_x - k_y$ plane for different k_z suggests that the effective mass of the band depends on k_z (see supplementary data).

The temperature and intercalation dependence of the Hall coefficient casts a doubt whether it can be used for an accurate determination of the carrier density. The Hall number is almost independent of Sr intercalation, albeit this intercalation is the main mechanism for doping mobile electrons in the sample [23, 29]. Together with the unexpected large temperature dependence of the Hall number we conclude that the Hall-inferred density is unreliable. Doped Bi_2Se_3 has a single parabolic band close to the Fermi-energy, and if we assume that the SdH signal

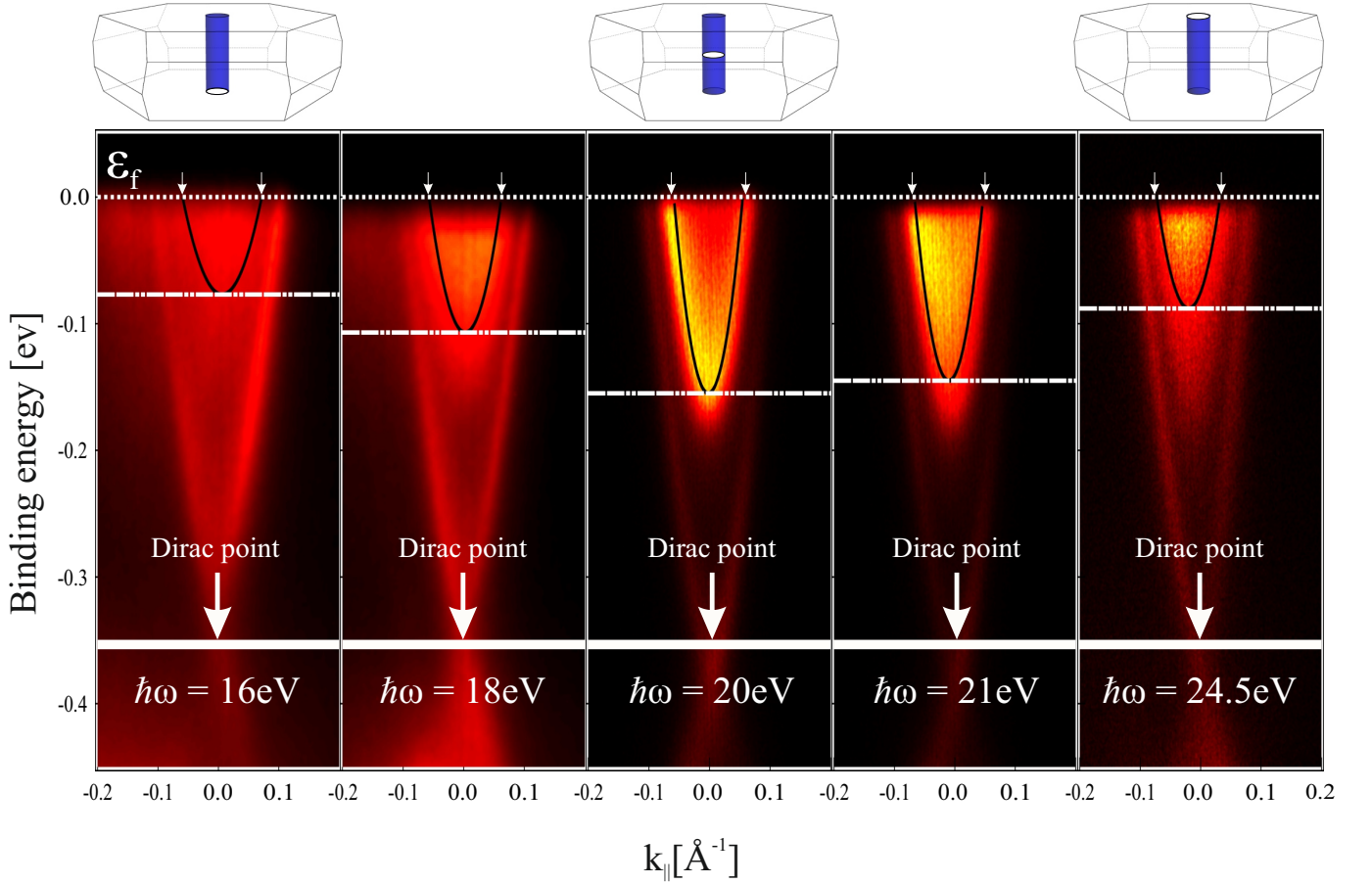


FIG. 3. Band structure of $\text{Sr}_x\text{Bi}_2\text{Se}_3$ samples measured at various photon energies, each photon energy corresponds to a different k_z value (see supplementary material). The two outmost panels are measurements at the two Z points, while the middle panel is a measurement at $k_z=0$. The 16 and 18 eV data were measured at the BaDElPh beam line at Elettra, and the 20, 21, 24.5 eV data were measured at the SIS beamline at the SLS, PSI. The small arrows mark the Fermi momentum, and the large arrow marks the Dirac point. ϵ_F and the minimum of the band are marked by dashed and dashed-dotted lines, respectively. In all panels the energy of the Dirac point is the same, as expected. Drawings on top: An illustration of the constant k_z plane at which the spectra in the panel below it was measured.

originates only from the conducting bulk, the interpretation of the data is straightforward and gives an exact information about the volume of the Fermi-surface and the carrier density. We therefore revisit our previous results and calculate the carrier density from the volume in momentum space as inferred from ARPES and SdH. The Fermi momentum from the ARPES, which is a direct method, gives $n = 6.5 \pm 1 \times 10^{19} \text{ cm}^{-3}$ for a non corrugated cylinder. The SdH numbers for this sample and for other samples are summarized in Table I.

A similar Fermi-surface topology is found in *all* intercalated Bi_2Se_3 samples exhibiting superconductivity [18, 30]. This observation raises the question of why does superconductivity appear concomitantly with an open Fermi-surface?

Intuitively, one might expect that the opening of the FS will result in a quasi-2D band structure that has a

larger density of states compared with the 3D case, for same carrier density. In the framework of weak-coupling BCS theory, the increase in density of states would enhance the critical temperature T_c . We use a simple model to quantify this intuitive idea, using weak-coupling limit BCS theory and the band-structure of Bi_2Se_3 , based on DFT calculations for pure Bi_2Se_3 [1, 31]. To account for the effect of the intercalation the coupling amplitudes in the \hat{z} direction are varied as a function of the chemical potential [25]. The FS of this band-structure is a narrow ellipsoid *closed* surface at low carrier densities and a cylindrical *open* surface at high carrier densities, as shown in Fig. 4, and in agreement with the experimental observation.

We can gauge the effect of the Lifshitz transition by plugging the density of states $g(\mu)$ into the standard weak coupling BCS [32] expression for the critical tem-

perature T_c in the weak coupling regime:

$$k_B T_c \approx 1.14 \hbar \omega_D \exp \left(-\frac{1}{V \cdot g(\mu)} \right), \quad (1)$$

where ω_D is the Debye frequency and V is the electron-phonon coupling constant. We use the known ω_D and adjust V to match the critical temperature of the system at large enough carrier densities where $g(\mu)$ is approximately constant. Assuming that V is independent of carrier density, we can plug it back into Eq. (1) and estimate T_c for different carrier densities. As shown in Fig. 4, the critical temperature of the open FS ($n > 5 \times 10^{19} \text{ cm}^{-3}$) is approximately constant, but drops sharply at carrier densities below the Lifshitz transition once the Fermi surface closes. Two experimental points $T_c(n)$ are indicated on the plot, matching the critical temperatures and carrier densities for $\text{Cu}_x\text{Bi}_2\text{Se}_3$ and $\text{Sr}_x\text{Bi}_2\text{Se}_3$ deduced from SdH measurements.

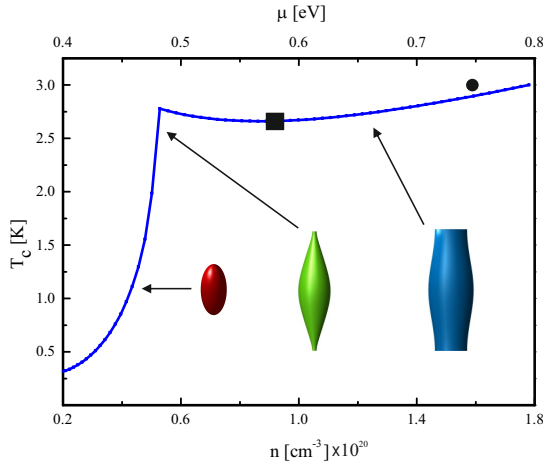


FIG. 4. Theoretical chemical potential/carrier density dependence of the superconducting critical temperature, obtained by combining DFT-based band structure with the BCS theory. Note the chemical potential calculated is about 3 times larger compared to the maximum bandwidth obtained by ARPES. Two experimental points are presented, a black dot for $\text{Cu}_x\text{Bi}_2\text{Se}_3$ [18] and a black square for $\text{Sr}_x\text{Bi}_2\text{Se}_3$, showing the consistency of the theoretical model.

Sample	$n_{\text{SdH}} \text{ cm}^{-3}$	$n_{\text{Hall}} \text{ cm}^{-3}$	ref.
Bi_2Se_3 (not SC)	1.9×10^{18}	3.7×10^{17}	[18]
Bi_2Se_3 (not SC)	1.8×10^{19}	2.3×10^{19}	[18]
$\text{Cu}_x\text{Bi}_2\text{Se}_3$ (SC)	1.6×10^{20}	2.7×10^{20}	[18]
$\text{Sr}_x\text{Bi}_2\text{Se}_3$ (SC)	9.3×10^{19}	$2 \times 10^{19*}$	This paper

TABLE I. Comparison between the Hall carrier density and the SdH carrier density. * - The Hall density of $\text{Sr}_x\text{Bi}_2\text{Se}_3$ is taken at $T = 6 \text{ K}$.

The main finding is that superconducting samples always have large ($\geq 6.5 \times 10^{19} \text{ cm}^{-3}$) carrier density, which

results in an open FS, consistent with our simplified model and measurements done using different techniques. From the theoretical point of view [16], a 3D topological superconductor is not achievable with our suggested Fermi-surface. This still leaves the door open for other scenarios of weak 2D topological superconductivity [33].

An open Fermi-surface is consistent with two more observations. First, the most realistic physical mechanism of superconducting pairing in $\text{Sr}_x\text{Bi}_2\text{Se}_3$ is the electron-phonon interaction, that occurs at small phonon momenta [13, 34]. An open Fermi-surface allows this mechanism to be energetically favourable [13], as confirmed recently by inelastic neutron scattering [35]. Second, as shown in Ref. [36], for an odd-parity E_u pairing, the nodeless superconducting gap observed in tunnelling measurements in $\text{Cu}_x\text{Bi}_2\text{Se}_3$ [1, 3, 4] and $\text{Sr}_x\text{Bi}_2\text{Se}_3$ [37] is consistent only with an open Fermi-surface.

In addition, the suggested mean-field picture with the very weak dependence of T_c on the density is in agreement with the observation of a Hall density independent T_c in Cu-co-doped $\text{Sr}_x\text{Bi}_2\text{Se}_3$ crystals [21].

Finally, we note that according to our simplified model a small region in the carrier density ($4\text{-}5 \times 10^{19} \text{ cm}^{-3}$) - temperature phase diagram may allow for a 3D topological superconductivity. This would require precise tuning of carrier density, possibly using gate voltage or proper co-doping.

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