Manifestation of Topological Protection in Transport Properties of Epitaxial Bi$_2$Se$_3$ Thin Films

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The massless Dirac fermions residing on the surface of three-dimensional topological insulators are protected from backscattering and cannot be localized by disorder, but such protection can be lifted in ultrathin films when the three-dimensionality is lost. By measuring the Shubnikov-de Haas oscillations in a series of high-quality Bi$_2$Se$_3$ thin films, we revealed a systematic evolution of the surface conductance as a function of thickness and found a striking manifestation of the topological protection: The metallic surface transport abruptly diminishes below the critical thickness of $\sim$6 nm, at which an energy gap opens in the surface state and the Dirac fermions become massive. At the same time, the weak antilocalization behavior is found to weaken in the gapped phase due to the loss of $\pi$ Berry phase.

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In topological insulators (TIs) the energy states are fundamentally modified from ordinary insulators by strong spin-orbit interactions, giving rise to a topologically distinct state of matter with a gapped insulating bulk and a gapless metallic surface [1]. Various interesting phenomena, including surface transport of spin-filtered Dirac fermions that are immune to localization, have been predicted and raised expectations for novel applications [2-4]. However, the progress in real applications of TIs crucially relies on the ability to manipulate the surface current in transport experiments. At present, such basic characterization as the surface conductance measurement has been possible only in a few cases in single crystals [5-12] because of the dominance of bulk transport caused by unintentional doping due to defects. Molecular beam epitaxy (MBE) is a promising technique for the synthesis of TIs [13-15] owing, in part, to the relatively low deposition temperature at which defect concentrations can be reduced from those in bulk crystals grown in thermal equilibrium. So far, using MBE-grown films, angle-resolved photoemission spectroscopy (ARPES) [19] and scanning tunneling spectroscopy (STS) [20] have provided useful information about the topological surface state (SS), and in transport experiments, such phenomena as weak antilocalization (WAL) and gate-controlled ambipolar transport have been reported [21, 22]. Also, since the MBE technique gives a precise control over film thickness, transport measurements for widely varying surface-to-bulk conductivity ratio have been performed [23, 24], although a reliable separation of surface Dirac electrons from bulk carriers has been hindered by a relatively low mobility of carriers in available thin films. Recently, we have succeeded in growing high-quality epitaxial films of Bi$_2$Se$_3$ that have a sufficiently high surface electron mobility to present pronounced Shubnikov-de Haas (SdH) oscillations. This made it possible to directly probe the surface conductance and the topological protection of the SS.

The immunity of the surface Dirac fermions to localization has a twofold origin [1-3]. One is the $\pi$ Berry phase associated with massless Dirac fermions [26], which protects them from weak localization through destructive interference of time-reversed paths. The other is the peculiar spin-momentum locking which nulls the backscattering probability [1-3]. Those mechanisms are collectively called topological protection. Recently, it was found [19, 27-31] that when TIs are thinned to the extent that the top and bottom surface states feel each other, their hybridization leads to an opening of the gap at the Dirac point and results in a degenerate, massive Dirac dispersion. This gapped state obviously violates the topological protection, but its consequence in the surface transport properties has not been duly addressed. In fact, this question is important because recently a lot of attention has been paid to the way to open a gap at the Dirac point [32] to realize topological magnetoelectric effects [33]. One may expect that unless the chemical potential is located exactly within the gap, the metallic surface transport is largely unaffected by the gap opening because states are kept being available for transport at the Fermi level. However, in the present work it turned out that the change in the Dirac spectrum deep in the occupied state has a profound effect on the physics at the Fermi level.

The growth of Bi$_2$Se$_3$ films occurs in a layer-by-layer manner, in which the 0.95-nm thick Se-Bi-Se-Bi-Se quintuple layer (QL) constitutes the basic unit [19]. Our systematic magnetotransport measurements for varying thickness reveal a sudden diminishment of the surface transport below the critical thickness of $\sim$6 QL, below which the energy gap opens in the Dirac spectrum [19]. We also observed that the weak antilocalization behavior [21, 22] quickly weakens below the critical thickness. We discuss that those striking effects are due to acquired degeneracy of the surface states [23] and loss of their $\pi$ Berry phase [34, 55] in the gapped phase.
Our MBE films were grown under Se-rich conditions on insulating sapphire (0001) substrates whose size was approximately 15 × 4 mm². To obtain films of high enough quality to present SdH oscillations, we employed a two-step deposition procedure. Both Bi (99.9999%) and Se (99.999%) were evaporated from standard Knudsen cells. The Se₂(Se₄)/Bi flux ratio was kept between 15 – 20. The growth rate, which is determined by the Bi flux, was kept at 0.2 – 0.3 QL/min. The resistance $R_{xx}$ and the Hall resistance $R_{yx}$ were measured in the Hall bar geometry by a standard six-probe method on rectangular samples on which the contacts were made with silver paste at the perimeter and cured at room temperature under pressure of ∼1 Pa. The magnetic field was swept between ±14 T at fixed temperatures and was always applied perpendicular to the films, except for the angular-dependence measurements of the SdH oscillations.

An atomic force microscopy (AFM) image of our relatively thick (50 QL) film is shown in Fig. 1(a), where a large, atomically flat area of ∼1 μm² and several sharp terraces with the height of exactly 1 QL can be clearly seen. The reflection high-energy electron diffraction (RHEED) pattern with sharp 1×1 streaks [Fig. 1(a) inset] and sharp x-ray diffraction peaks [Fig. 1(b)] are also indications of the high crystal quality of our films.

Temperature dependencies of the sheet resistance, $R_S(T)$, measured in films with systematically changed thickness $t$ down to 2 QL are shown in Fig. 1(c). The $R_S(T)$ behavior is metallic in thick films, but below $t = 5$ QL, it starts to show an upturn at low temperatures. In particular, the sharp divergence in $R_S$ for $T \to 0$ in the 2-QL film is indicative of strong Anderson localization and an insulating ground state.

The breakthrough in the present work is that our films exhibit pronounced two-dimensional (2D) SdH oscillations to provide a direct way to probe the surface charge transport. As an example, the analysis of the SdH oscillations in the 10-QL film is shown in Fig. 2. The 2D character of the oscillations is evident in Fig. 2(a), where the positions of the maxima and minima depend only on the perpendicular component of the magnetic field, $B_\perp$. The oscillation frequency $F = 106.8$ T is obtained from the Fourier transform [lower inset of Fig. 2(c)], and this is a direct measure of the Fermi wave number $k_F = 5.7 \times 10^6$ cm⁻¹. As we discuss in detail in the Supplemental Material, if the SdH oscillations are due to the trivial 2D electron gas (2DEG) which may form due to a band bending near the surface, this $k_F$ is so small that it imposes too strong a constraint on the possible bulk Fermi level, which makes it impossible to consistently explain the transport data. Hence, we identify the
oscillations to be due to surface Dirac fermions, and the obtained $k_F$ gives their density $n_s = 2.6 \times 10^{12}$ cm$^{-2}$.

In this 10-QL film, we observed only a single frequency, but we usually see two frequencies in other films (see Fig. S3 of the Supplemental Material [37]), suggesting that the top and bottom surfaces have somewhat different $n_s$. The temperature dependence of the oscillation amplitude [Fig. 2(b) inset] gives the cyclotron mass $m_c = 0.2m_0$ ($m_0$ is the free electron mass) [39] which in turn gives the Fermi velocity $v_F = 3.3 \times 10^7$ cm s$^{-1}$. This $v_F$ is consistent with the ARPES data [40] as well as the STS data [24, 41] for the Dirac cone. The obtained $k_F$ value corresponds to the Fermi level of $\sim$0.16 eV above the Dirac point for the topological surface state, which points to a slight upward band bending [42]. The Dingle analysis [Fig. 2(b)], see the Supplemental Material [37] for details, yields the mobility $\mu_s = 1330$ cm$^2$V$^{-1}$s$^{-1}$.

Finally, Fig. 2(c) shows the Landau-level fan diagram for the oscillations in conductance $G_{xx}$, where the positions of the minima in $\Delta G_{xx}$ (shown in the upper inset) are plotted as a function of $n$ [43]. Here, to minimize the error occurring from extrapolation [12], we fix the slope of the linear fitting by using $F = 106.8$ T obtained from the Fourier analysis and determine the intercept $n = 0.40 \pm 0.04$ (solid line); this is very close to the ideal value of 0.5 for Dirac electrons bearing the $\pi$ Berry phase [41], giving further confidence in the origin of the SdH oscillations. For comparison, a straight line with the same slope to give zero Berry phase is shown in Fig. 2(c) with a dashed line, which is obviously inconsistent with the experimental data.

To estimate the contribution of the SS in the overall transport in this 10-QL film, we use the magnetic-field dependence of the Hall resistivity, $R_{yx}(B)$ [Fig. 2(d)], which is not linear in $B$ and thus signifies the presence of at least two types of carriers. The fitting of a standard two-band model [6, 8, 15] to the data, in which $n_s$ is fixed by the SdH frequency, gives the surface contribution to the total conductance, $G_s/G_{tot}$, of 28%. The $\mu_s$ value obtained from this fitting is close to the SdH result, assuring the consistency of our analysis.

The same analysis can be applied to all measured films with $t \geq 8$ QL, in which we consistently observed SdH oscillations [37]. Evolutions of the transport parameters with changing $t$ are summarized in Figs. 3(a)-3(c). We note that by tracing the evolution of the SdH oscillations starting from thick films, we can distinguish the topological SS from the 2D quantum-well (QW) state of the bulk origin [37] as the source of the SdH oscillations.

Our main finding is that the surface transport abruptly diminishes below a critical thickness $t_c$ which is located between 5 and 8 QL. This change is most convincingly manifested in the behavior of $R_{yx}(B)$, which suddenly becomes $B$-linear in films with $t \leq 5$ QL [Fig. 3(d)]; this indicates that the transport becomes suddenly dominated by only one type of carriers. Correspondingly, the SdH oscillations disappear for $t < t_c$. More quantitatively, assuming that $n_s$ is essentially unchanged through $t_c$ [dashed squares in Fig. 3(a)], one can estimate that $\mu_s$ must be suddenly degraded by more than an order of magnitude in samples with $t < t_c$ [dashed squares in Fig. 3(b)] for $R_{yx}(B)$ to become linear and be governed by bulk carriers [37]. The magnetoresistance behavior shown in Fig. 3(e) also presents a qualitative change below $t_c$, showing a negative slope at high fields. This evolution is best represented in the $t$ dependence of $G_s/G_{tot}$ [Fig. 3(c)], which shows a steady increase with decreasing $t$ to reflect the change in the surface-to-bulk ratio, but it drops sharply below $t_c$ to signify that the surface transport is abruptly diminished.

This observation naturally calls for the question whether the observed diminishment of the surface transport in ultrathin films might be related to a lowering of the quality in thinner films. In this respect, our ultrathin films remain essentially flat and smooth across $t_c$, with the surface bumpiness of only $\sim$1 QL (see Fig. S10 of the Supplemental Material [37]). This observation, combined with the fact that $G_s/G_{tot}$ increases steadily with decreasing $t$ until it reaches $t_c$, testifies against the above concern.

In addition to the above results, we found a striking change in the WAL behavior [21, 22] below $t = 5$ QL. Fig.
α coherence length. The prefactor analysis, connected though bulk electrons [21, 22]. The change to Fig. 4(b) shows the transport channel that either carries a lines are the fittings using Eq. (1). (b) Thickness dependence of α. Inset shows schematic energy bands above and below the critical thickness.

ure 4(a) shows the magnetoconductance of our films measured in perpendicular magnetic fields at 1.6 K. Dashed lines are the fitting with the Hikami-Larkin-Nagaoka formula [46],

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

(1)

where $\Psi$ is the digamma function and $L_\phi$ is the phase coherence length. The prefactor $\alpha$ should be $-\frac{1}{2}$ for each transport channel that either carries a π Berry phase [20] or bears a strong spin-orbit interaction [10]. In our analysis, $\alpha$ and $L_\phi$ are the only fitting parameters and Fig. 4(b) shows the $t$ dependence of obtained $\alpha$ (see the Supplemental Material [32] for details). For $t \geq 5$ QL, we observed $\alpha \approx -\frac{1}{2}$ similar to that reported for metallic Bi$_2$Se$_3$ thin films where top and bottom surfaces are connected though bulk electrons [21, 22]. The change to $\alpha \approx 0$ observed for $t \leq 3$ QL is in accord with the diminishing of the surface transport channel and the eventual localization of the bulk state. A similar tendency was also observed in previous studies [23, 25], although the decrease in $\alpha$ was less pronounced probably due to a larger metallicity of the measured samples.

Now we discuss the origin of our observations. As was already found in Bi$_2$Se$_3$ ultrathin films by ARPES [10], an energy gap in the SS opens at the Dirac point below $t_c \simeq 6$ QL. Obviously, our transport measurements reflect this change in the Dirac dispersion. The gap is due to hybridization between top and bottom surfaces [schematically shown in the Fig. 4(b) inset], and such a hybridization gap $\Delta$ changes the massless Dirac dispersion $E = \pm \hbar v_F k$ into a massive one $E = \pm \sqrt{(\hbar v_F k)^2 + (\Delta/2)^2}$. For Bi$_2$Se$_3$, Δ is about 0.25 eV at $t = 2$ QL [19]. In our films, the Fermi level is estimated to be $\sim 0.16$ eV above the Dirac point, so even in our thinnest film the Fermi level is not in the gap but crosses the SS as schematically shown in the inset of Fig. 4(b). Hence, the observed drastic suppression of the surface transport is not due to the disappearance of surface carriers but is likely due to an enhanced scattering of the carriers [47].

The gap opening also has a profound effect on the Berry phase $\phi_B$ of the surface band $\psi_k(r)$. In the simplest case, $\phi_B$ is given by [34]

$$\phi_B = -i \int_0^{2\pi} d\varphi \left| \psi_k(r) \right| \frac{\partial \psi_k(r)}{\partial \varphi} = \pi \left( 1 - \frac{\Delta}{E_F} \right),$$

(2)

and hence the Berry phase is reduced from $\pi$ when a gap opens. This lifts the immunity of the SS from weak localization. Also, this change in $\phi_B$ weakens the WAL in the SS [33]. It is useful to note that in Fig. 4(a) the WAL was still observed for $t = 5$ QL which is below $t_c$, but this is natural because the small $\Delta$ at this $t$ makes the deviation of $\phi_B$ from $\pi$ to be small.

More importantly, in the gapped phase, the SS becomes degenerate [20], which means that now for each momentum both up and down spin states are available. This opens the backscattering channel and significantly reduces the surface mobility. Actually, the hybridization of top and bottom surfaces means that the system is no longer truly three-dimensional (3D), so it is natural that the topological properties of 3D TIs are lost in the gapped phase, in full agreement with recent ab initio density functional studies of Tl thin films [33, 31]. In this respect, the present observation is a spectacular manifestation of the topological protection of the SS in 3D TIs.

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Supplemental Material

S1. Van der Waals epitaxy and two-step deposition

The building block of the layered Bi$_2$Se$_3$ is a quintuple layer (QL) as depicted in Fig. S1(a). The surface is terminated by Se atoms without dangling bonds, interacting only by weak van der Waals forces. Due to the weak interaction with a substrate, an epitaxial growth with its own lattice constant is possible at the very beginning (with a drastically relaxed lattice matching condition) as schematically shown in Fig. S1(b). Thus, the growth of ultrathin films starting from one QL is possible [48]. This is called van der Waals epitaxy.

One of the key parameters of the MBE growth is the temperature of deposition, which determines the rate of adsorption/desorption of Bi and Se as well as their surface diffusion. We found that increasing the substrate temperature greatly improves the size of grown terraces. Unfortunately, at high temperatures, the growth of thin films is virtually impossible due to a weak interaction of adatoms with the substrate. The crucial improvement was achieved by using a two-step deposition method [49]: In the first step, the initial 1 QL of Bi$_2$Se$_3$ is deposited at a low temperature of 110–120°C; then, the temperature of the substrate is slowly raised up to 300–320°C, and, in the second step, the desired number of QLs is deposited. We obtained the best quality thin films at the temperature of the second step of around 300°C.

S2. Anderson localization

An upturn in the sheet resistance in ultrathin Bi$_2$Se$_3$ films has always been seen at low temperatures [23–25], in spite of the difference in concentration of charge carriers and their mobilities. Figure S2 shows the diverging trend of $R_s$ at low temperatures we observed in the 2-QL film. A rapid decrease in the mobility with decreasing temperature is also shown in the inset of Fig. S2, which points to a localization of charge carriers in the zero-temperature limit. Similar behavior of $R_s(T)$ was studied in Ref. [24] on films with a higher conductance, where the insulating ground state was proposed to be due to strong interactions of surface electrons, which were assumed to dominate the transport in ultrathin films. However, for ultrathin films with lower conductance, which is determined by the concentration of charge carriers and the strength of disorder, the Anderson localization of a disordered 2D metallic conductor seems to be a better (more natural) explanation for the observed insulating state. Such localization behavior in low-conductance ultrathin films highlights the disappearance of topological protection of the surface state. This conclusion is also supported by a recent study of field-effect devices fabricated from ultrathin (∼3.5 nm) Bi$_2$Se$_3$ crystals obtained by mechanical exfoliation on SiO$_2$/Si substrates [50]. By measuring the temperature and gate-voltage dependences of the conductance, the authors of Ref. [50] observed clear insulating behavior with an activated energy gap when the chemical potential is tuned into the gap, which made them conclude that their 3.5-nm thick Bi$_2$Se$_3$ crystals are conventional insulators.

S3. SdH oscillations

Transport properties of surface carriers are obtained mostly from the SdH oscillations observed in films with the thickness $t$ above 6 QL (Fig. S3). The frequency $F$ identified in SdH oscillations in $R$ vs. $1/B$ gives the size of a 2D Fermi surface (FS) via the Onsager relation $F = (h/2\pi e)A$, where $A = \pi k_F^2$ is the FS cross section; $h$ and $e$ are the reduced Plank’s constant and the electron charge, respectively; $k_F$ is the Fermi wave number. The concentration of spin-filtered surface charge carriers is $n_s = k_F^2/4\pi$.

The cyclotron mass $m_c$ is obtained from the temperature damping factor of the first harmonic, $R_T = \pi\lambda/\sinh(\pi\lambda)$, where $\lambda = 2\pi k_B T/\hbar\omega_c$ and $\omega_c \equiv eB/m_c$ is the cyclotron frequency [39]. From $k_F$ and $m_c$, we obtain the Fermi velocity $v_F = \hbar k_F/m_c$.

The quantum scattering time $\tau = h/(2\pi k_B T_D)$ and the mean free path $\ell_s = v_F\tau$ are obtained from the Dingle damping factor $R_D = \exp(-2\pi^2 k_B T_D/\hbar\omega_c)$, which accounts for scattering effects [39]; the surface mobility $\mu_s$ is estimated from the relation for the surface conductance $G_s = e\mu_s n_s = (e^2/h)(k_F\ell_s)$.

S4. Sudden drop in the surface mobility below $t_c$

As has been discussed in the main text, the SdH oscillations disappear below $t_c$, which coincides with the disappearance of topological protection of the surface state (SS) when the energy gap opens and the Dirac fermions become massive. Here we present additional discussions that the disappearance of the SdH oscillations with decreasing film thickness cannot be simply due to a smooth decrease of the surface mobility below the observational limit.
Figures S4 and S5 show $R_{xx}(B)$ and $R_{yx}(B)$ data measured in films with $t = 8$ and 5 QL, respectively, together with their fittings considering the surface and bulk channels, similar to what has been described in the main text for $t = 10$ QL (Fig. 2(d)). For the 8-QL film (Fig. S4), where we observed two SdH frequencies (Fig. S3), the concentrations of surface carriers $n_1 = 1.7 \times 10^{12} \text{ cm}^{-2}$ and $n_2 = 5.0 \times 10^{12} \text{ cm}^{-2}$ are fixed by the SdH frequencies. For the 5-QL film (Fig. S5), where SdH oscillations were not detected, we assume $n_1 = n_2 = 3 \times 10^{12} \text{ cm}^{-2}$ following the trend in $n_x$ vs $t$ for the surface channel [see Fig. 3(a)].

A three-band model analysis for the 8-QL film gives the mobilities $\mu_{s1} = 1600 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ and $\mu_{s2} = 1000 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ for two different surfaces and $\mu_3 = 161 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ for the bulk carriers (Fig. S4). Note that this combination of carrier densities and mobilities for the three channels reproduce the curved $R_{yx}(B)$ behavior as well as the $B = 0$ T value of $R_{xy}$, but the observed $B$ dependence of $R_{xx}$ is stronger than the three-band model prediction; this is understandable, because the simple multi-band model employed here neglects the intrinsic magnetoresistance (i.e. the $B$ dependence of the mobility) which is known to exist in Bi$_2$Se$_3$.

In contrast, the $R_{yx}$ data for the 5-QL film is almost perfectly linear in $B$, and to reproduce this linear behavior while keeping the consistency with the $R_{xx}(0)$ value, one needs to consider a very low mobility for the surface channel. For example, a combination of the surface mobility $\mu_{1,2} = 50 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ and the bulk mobility $\mu_3 = 140 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ gives reasonable reproductions of $R_{xx}(0)$ and $R_{yx}(B)$, as shown in Fig. S5. On the other hand, if one assumes a gradual change in the surface mobility across $t$, and hypothetically put $\mu_{1,2} = 500 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$, it is impossible to make $R_{xx}(0)$ to be consistent with the data nor to make $R_{yx}(B)$ linear, as shown in Fig. S6. Therefore, one can conclude that the surface mobility indeed dropped by more than one order of magnitude when $t$ was reduced from 8 to 5 QL.

S5. Quantum confinement effects

A. Formation of quantum-well states in ultrathin films

In thin films, the effects of quantum confinement modify the 3D bulk energy states to 2D quantum-well (QW) states when the film thickness becomes comparable to the de Broglie wavelength of the carriers. In the case of very thin films, it is usually difficult to distinguish between a genuine SS and a 2D QW state if one simply measures the angular dependence of the SdH oscillations. Fortunately, we were able to measure SdH oscillations in all films with $t > 6$ QL, including those in which bulk states are certainly in the 3D limit. As an example, Fig. S7 shows the 2D character of SdH oscillations measured in a 120-QL film. It is worth noting that in none of the measured films we saw SdH oscillations coming from the 3D bulk states. This is most likely because of unfavorable conditions for their observations, that is, the relatively low mobility and a high bulk-carrier concentration together make the oscillation amplitude to become small. The systematic measurements of SdH oscillations in a wide range of thickness make it possible to reliably distinguish the SSs from bulk states or the 2D QW states by tracing them from thick samples.

B. Surface band bending and resulting 2D electron gas

The surface bend bending can create a 2D electron gas (2DEG) on the surface of TIs, and this could be responsible for the observed 2D SdH oscillations instead of the topological SSs. Spectroscopically, coexistence of the topological state and a 2DEG state on the surface of Bi$_2$Se$_3$ has been observed by ARPES a few hours after cleaving [38, 52], which is believed to be due to gradual formation of defects (Se vacancies) on a freshly cleaved surface in ultrahigh vacuum. Figure S8 reproduces the energy dispersions of the SS and the 2DEG state measured along the $\bar{\Gamma}-\bar{\Gamma}-\bar{K}$ direction in Ref. [38]. Solid lines are our fittings of the equations shown in Fig. S8 to the experimental data.

Now we show that it is not possible to ascribe the observed 2D SdH oscillations to the 2DEG state. The frequency of the SdH oscillations in the 10-QL film shown in Fig. 2 of the main text corresponds to $k_F = 5.7 \times 10^6 \text{ cm}^{-1}$. As discussed in the main text, if the SdH oscillations are coming from the topological SS, this $k_F$ gives the estimate for $E_F$ of 160 meV measured from the Dirac point. On the other hand, if this $k_F$ is associated with the 2DEG state shown in Fig. S8, $E_F$ should be 82 meV from the bottom of this 2DEG state and the corresponding density of the 2DEG, $n_{2\text{DEG}}$, is $5.2 \times 10^{12} \text{ cm}^{-2}$. One should remember that for this 2DEG state to form, the conduction band must be bent downward near the surface, which dictates that the $E_F$ of the bulk state measured from the bulk conduction band edge must be smaller than 82 meV. This means that the maximum possible bulk carrier density is $5 \times 10^{18} \text{ cm}^{-3}$ if we use the effective mass $m^* = 0.13m_0$ for the bulk conduction band [33]. (A larger bulk $E_F$ would mean that the band is bent upward near the surface, which precludes the formation of the 2DEG state from the beginning.)

However, the high-field slope of the $R_{yx}(B)$ data, which simply gives a sum of the densities of electrons from all bands [33], points to the existence of a total of $4 \times 10^{19} \text{ cm}^{-3}$ of electrons in our 10-QL film; since the above 2DEG density corresponds to the volume density of only $5.2 \times 10^{18} \text{ cm}^{-3}$, the maximum possible bulk carrier density of $5 \times 10^{18} \text{ cm}^{-3}$ is not enough to account for the Hall data. (Note that the density of Dirac electrons is $1 \times 10^{13} \text{ cm}^{-2}$ at most in the above 2DEG scenario (see later), and hence the topological SS can only provide up to $2 \times 10^{19} \text{ cm}^{-3}$ of
carriers from the two surfaces, and this is still not enough.] Therefore, one cannot achieve a consistent understanding of the transport data in the 2DEG scenario.

To illustrate the difficulty of the 2DEG scenario in a more quantitative manner, let us assume, for simplicity, the same band bending as observed in Ref. [38] (i.e., the main 2DEG state located 40 meV below the bottom of the bulk conduction band and 90 meV above the conduction-band edge of the surface, which gives the total of 130 meV downward band bending, see the inset of the left panel of Fig. S9 [54]). Then the position of the Dirac point (DP) is about 370 meV below the surface Fermi level, giving the concentration of Dirac electrons \( n_{\text{Dirac}} = 1 \times 10^{13} \text{ cm}^{-2} \).

The bulk \( E_F \) should be 42 meV in this situation, and the corresponding bulk carrier density is \( n_{\text{bulk}} = 1.8 \times 10^{18} \text{ cm}^{-3} \). Therefore, all carrier densities are fixed and the only parameters that can be tuned to reproduce \( R_S(0) \) and \( R_{\text{xy}}(B) \) are the mobilities of 2DEG, Dirac, and bulk electrons. Figure S9 shows the result of the best fitting, which actually does not fit the \( R_{\text{xy}}(B) \) behavior at all and obviously the slope of the calculated \( R_{\text{xy}}(B) \) is way too steep. This is essentially because the concentrations of both Dirac and bulk electrons are too low in the 2DEG scenario. As presented in the main text, the alternative scenario to ascribe the SdH oscillations to the topological SS and consider a slight upward band bending (which precludes the formation of the 2DEG state) naturally explains the relatively large bulk carrier density and gives a totally consistent description of our data.

It is worth noting that the above-estimated \( E_F \) of 82 meV for the 2DEG state in the downward band-bending scenario is already significantly smaller than that seen in the ARPES data shown in Fig. S8, where \( E_F \) is about 200 meV for the 2DEG state. This means that in our 10-QL film, even if one assumes that the 2DEG state were formed and were responsible for the SdH oscillations, the accumulated charge on the surface must be much smaller than in the case observed in ARPES experiments [39], and hence the band bending, if existed, must be much weaker [54]. In other words, the SdH data dictate that the band bending as strong as that shown in Fig. S9 cannot actually be conceived for our 10-QL film.

In addition, we note that we did not observe any sign of progressive downward band bending in our transport data, even though our samples are not protected by any passivation layer and we applied a couple of different sample preparation procedures: some samples were transferred into an inert atmosphere immediately after the growth and were exposed to air for only several minutes before measurements, other samples spent hours in air during the contact preparation, and several samples have been remeasured months after their first measurement. So far no relaxations were exposed to air for only several minutes before measurements, other samples spent hours in air during the contact preparation, and several samples have been remeasured months after their first measurement. It is worth noting that the above-estimated \( E_F \) of 82 meV for the 2DEG state in the downward band-bending scenario is already significantly smaller than that seen in the ARPES data shown in Fig. S8, where \( E_F \) is about 200 meV for the 2DEG state. This means that in our 10-QL film, even if one assumes that the 2DEG state were formed and were responsible for the SdH oscillations, the accumulated charge on the surface must be much smaller than in the case observed in ARPES experiments [39], and hence the band bending, if existed, must be much weaker [54]. In other words, the SdH data dictate that the band bending as strong as that shown in Fig. S9 cannot actually be conceived for our 10-QL film.

S6. Film quality across \( t_c \)

To confirm that the observed diminishment of the surface transport in ultrathin films is not caused by a lowering of the quality in thinner films, we have characterized the morphology of our films using atomic force microscopy. As shown in Fig. S10, our ultrathin films remain essentially flat and smooth across \( t_c \) (surface bumpiness of only \( \sim 1 \text{ QL} \)). This observation suggests that it is not very likely that there is a drastic degradation in the film quality as a function of \( t \).

It is worth noting that in our films the sheet density of bulk carriers [shown in Fig. 3(a) of the main text by circles] changes smoothly but it is not proportional to \( t \), which is similar to what was previously reported [23]; the observed trend suggests that the density of Se vacancies gradually increases in thinner films [23].

S7. Weak antilocalization

The fitting parameters of the WAL behavior measured in our films are shown in Table I. One can clearly see that both \( \alpha \) and \( L_\phi \) rapidly decrease below the critical film thickness. A similar tendency has also been observed in previous thickness-dependence studies [23, 25], although the decrease of \( \alpha \) was less pronounced and more gradual, which is not a surprise for more metallic films where the suppression of the topological SS is less crucial.

We also measured the angle dependence of the WAL behavior. Figure S11 shows the low-field magnetoresistance measured in the 10-QL film in tilted magnetic fields. The WAL response plotted as a function of \( B \cos \theta \) clearly demonstrates its entirely 2D character and is well fit by the Hikami-Larkin-Nagaoka (HLN) formula [Eq. (1) of the main text].
| Thickness (QL) | α   | $L_\alpha$ (nm) |
|---------------|-----|-----------------|
| 2             | 0.007 | 101             |
| 3             | 0.04  | 153             |
| 5             | 0.5   | 256             |
| 8             | 0.43  | 409             |
| 9             | 0.45  | 584             |
| 10            | 0.5   | 578             |
| 15            | 0.45  | 573             |
| 21            | 0.5   | 794             |
| 32            | 0.4   | 405             |
| 50            | 0.57  | 905             |

TABLE I: Parameters of the WAL behavior described by the HLN formula.

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FIG. S1: (a) Schematic picture of the Bi$_2$Se$_3$ crystal structure (two quintuple layers are shown). (b) Schematic picture of the van der Waals MBE growth on sapphire.
FIG. S2: Temperature dependence of the sheet resistance in a 2-QL film showing a diverging trend at low temperatures. Inset shows the temperature dependence of the Hall mobility $\mu = 1/eNR_s$.

FIG. S3: SdH oscillations in $dR_{xx}/dB$ measured in films with various thicknesses. Insets show the Fourier transform of the data in the main panel.
FIG. S4: Fitting of a three-band model to $R_S(B)$ (left panel) and $R_{yx}(B)$ (right panel) measured in the 8-QL film at 1.6 K. Symbols are experimental data. Solid lines are calculations.

FIG. S5: Two-band model calculations of $R_S(B)$ (left panel) and $R_{yx}(B)$ (right panel) for the 5-QL film with the parameters shown in the left panel, together with the experimental data measured at 1.6 K.
FIG. S6: Two-band model calculations of $R_S(B)$ (left panel) and $R_{yx}(B)$ (right panel) for the 5-QL film with a hypothetical surface mobility of 500 cm$^2$V$^{-1}$s$^{-1}$ which is lower than that for $t > t_c$ but is still reasonably high. The experimental data measured at 1.6 K are shown by symbols for comparison.

FIG. S7: $dR_{yx}/dB$ (shifted for clarity) measured in tilted magnetic fields in the Bi$_2$Se$_3$ film with the thickness of 120 QL and plotted as a function of $1/B \cos \theta$, demonstrating the 2D character of oscillations with two frequencies. Inset shows the Fourier transform of the data at $\theta = 0^\circ$ indicating the two-frequency nature of the oscillations.
FIG. S8: Energy dispersions of the SS and the 2DEG state measured on an aged surface of a cleaved Bi$_2$Se$_3$ crystal in Ref. 38. Solid lines are our fitting results.

FIG. S9: $R_S(B)$ and $R_{yx}(B)$ measured in the 10-QL film (symbols) and their fitting (solid lines) using parameters constrained in a 2DEG scenario (see text). Inset in the left panel shows a schematic picture of the band bending considered for the calculations, where $E_C$ is the bottom of the conduction band, $E_V$ is the top of the valence band, $E_F$ is the Fermi level, $E_0$ is the main (first) 2DEG energy state appeared due to the confinement of the electron motion perpendicular to the surface of the film.
FIG. S10: (a-c) Atomic force microscope images and height profiles of the 3, 5, and 8 QL films; the white lines indicate the 5-µm segment for which the profiles are shown.

FIG. S11: The WAL behavior measured for various magnetic field directions in the 10-QL film. θ is the angle between the magnetic field and the surface normal. The dashed line is the fitting of the HLN formula.