Topological Surface Transport in Epitaxial SnTe Thin Films Grown on Bi₂Te₃

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The topological crystalline insulator SnTe has been grown epitaxially on Bi₂Te₃ buffer layer by molecular beam epitaxy. Thanks to the $p$-$n$-junction nature of the interface, the Bi₂Te₃ layer is electrically decoupled and makes negligible contribution to the transport properties when measured with contacts on the SnTe surface. In a 30-nm-thick SnTe film, $p$- and $n$-type carriers are found to coexist, and Shubnikov-de Haas oscillations combined with the Hall resistivity data provide compelling evidence that the $n$-type carriers are Dirac fermions residing on the top SnTe (111) surface, where a downward band bending necessarily occurs to avoid polar catastrophe. This is the first transport observation of the topological surface state in a topological crystalline insulator.

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The energy band inversion and time-reversal symmetry (TRS) are the main ingredients for realizing a nontrivial topology in $Z₂$ topological insulators (TIs) [1–4]. Recently, the family of TIs has been extended by the introduction of topological crystalline insulators (TCIs) [5, 6] where the topology is protected by a point-group symmetry of the crystalline lattice rather than by TRS. The first material predicted to be a TCI was SnTe [7], in which the band inversion predicted at an even number of time-reversal-invariant momenta (TRIMs) leads to a trivial $Z₂$ topological invariant, but its mirror symmetry gives rise to a nontrivial mirror Chern number $n_{M} = -2$ to guarantee the existence of topologically protected gapless surface states (SSs) on any surface containing a mirror plane. Angle-resolved photoemission spectroscopy experiments have confirmed the existence of Dirac-like SSs on the (001) surface of SnTe [7] and related compounds [8, 9], generating a lot of interest in these materials. Naturally, an important next step is to elucidate the topological SSs with transport experiments, as was done for $Z₂$ TIs [10–13].

However, probing the SSs in SnTe by transport experiments is a challenge, because of a high concentration of bulk holes ($10^{20} - 10^{21}$ cm$^{-3}$) [20]. Nevertheless, in thin films, an enhanced surface-to-bulk ratio and a high surface mobility expected for topologically-protected SSs [21, 22] might make it possible to probe them in quantum oscillations. To obtain high-quality thin films by molecular beam epitaxy (MBE) [22, 30], lattice matching of the substrate is crucial. In this regard, while BaF₂ is the usual choice of substrate for SnTe [31] with its $\sim 1.6\%$ lattice matching, we noticed that rhombohedral Bi₂Te₃ may be a better choice, at least for the (111) growth direction, with the lattice matching of $\sim 1.5\%$. Furthermore, the building block of Bi₂Te₃ is a Te-Bi-Te-Bi-Te quintuple layer (QL) terminated with a hexagonal Te plane, which naturally accommodates the Sn layer of the SnTe in the (111) plane [see Fig. 1(d)]. In addition, epitaxial Bi₂Te₃ grown by MBE is usually degenerately $n$-type [32, 33], which would make it electrically isolated from the $p$-type SnTe film grown on its top, due to the formation of a $p$-$n$ junction and the resulting tunnel barrier at the interface.

In this Letter, we show that high-quality SnTe thin films grown by MBE on Bi₂Te₃ indeed possess desirable properties and allow us to probe the topological SSs in transport experiments. We observed Shubnikov-de Haas (SdH) oscillations composed of a single frequency, and its dependence on the magnetic-field direction signifies that it stems from a two-dimensional (2D) Fermi surface (FS). Furthermore, the phase of the oscillations indicates that the 2D carriers are Dirac electrons. Careful consideration of the energy-band diagram in the present heterostructure makes it possible to conclude that the Dirac electrons reside on the top surface of SnTe. Intriguingly, the surface mobility is found to diminish strongly with increasing temperature.

The MBE growth was performed in a ultra-high vacuum chamber with the base pressure better than $5 \times 10^{-8}$ Pa. Before deposition of SnTe, a thin layer of high-quality Bi₂Te₃ was grown under Te-rich conditions on sapphire substrates [34] with a two-step deposition procedure similar to that used for Bi₂Se₃ films [27, 35, 36]. Both Bi (99.9999%) and Te (99.9999%) were evaporated from standard Knudsen cells. The Te₂/Te₄/Bi flux ratio was kept at $\sim 20$. The growth rate, which is determined by the Bi flux, was kept at 0.3 nm/min. After growing $\sim 30$ nm of Bi₂Te₃ layer, Sn (99.9999%) and Te were co-evaporated, keeping the Te₃/Te₄/Sn flux ratio at $\sim 40$, substrate temperature at 300°C, and the growth rate at 0.4 nm/min. The resistivity $\rho_{xx}$ and the Hall resistivity $\rho_{yx}$ of the films were measured in a Hall-bar geometry by a standard six-probe method on rectangular samples on which the contacts were made with silver paste or indium near the perimeter. The magnetic field was swept between $\pm 14$ T at fixed temperatures.

A critical ingredient for the epitaxial SnTe growth in the present experiment is the high quality of the Bi₂Te₃ buffer layer. Figure 1(a) shows an atomic force microscopy (AFM) image of a 40-nm-thick Bi₂Te₃ thin film grown on sapphire substrate. Large equilateral triangles
The fringe distance $\Delta$ discernible kink in the data, suggesting that the structure from Bi$_2$Te$_3$ thick SnTe film. As we discuss in detail later, contribution of a series of SnTe films grown on Bi$_2$Te$_3$ step height of the terraces is $\sim 0.4$ nm. (c) Low-angle XRD patterns of a series of SnTe films grown on Bi$_2$Te$_3$ of different thickness. The total film thickness $d_t$ given by the distance $\Delta_1$ of Kiessig fringes at grazing angles is shown to the left. The fringe distance $\Delta_2$ near the (003) Bi$_2$Te$_3$ Bragg peak gives the thickness of the Bi$_2$Te$_3$ layer, $d_6$, which is shown near the peak. The SnTe layer thickness is given by $d_t - d_6$. Inset shows a wide-angle XRD pattern. (d) The (111) plane in the rock-salt lattice of SnTe is marked by triangles.

with atomically flat terraces, which have a height of exactly 1 quintuple layer (QL), can be easily recognized. An AFM image of a 30-nm-thick SnTe film grown on top of such Bi$_2$Te$_3$ buffer layer is shown in Fig. 1(b). Triangles are still clearly seen on the surface, giving evidence for an epitaxial growth. The height of the terraces is $\sim 0.4$ nm, which agrees with the periodicity of the rock-salt lattice along the (111) direction [Fig. 1(d)].

The high structural quality of both Bi$_2$Te$_3$ and SnTe films as well as the very smooth nature of the interface between them can be judged from the Kiessig fringes [33, 37] in the x-ray diffraction (XRD) measurements [Fig. 1(c)]. The inset of Fig. 1(c) shows the XRD pattern for a wider angle range, in which SnTe only yields (2n, 2n, 2n) Bragg peaks to confirm the (111) growth direction.

Figure 2(a) shows the behavior of $\rho_{xx}(T)$ in a 30-nm-thick SnTe film. As we discuss in detail later, contribution from Bi$_2$Te$_3$ buffer layer is negligible. There is no discernible kink in the data, suggesting that the structural phase transition observed in bulk SnTe [20, 38, 39] is absent in our thin films and that the mirror symmetry is kept intact down to low temperature [33]. In the magnetotransport properties, a downward cusp observed in $\rho_{xx}(B)$ at very low fields [Fig. 2(b)] is a reflection of the weak antilocalization behavior which is expected for topological materials [40, 42]. An interesting observation is that $\rho_{yz}(B)$ [Fig. 2(c)] shows a sign change in the slope, pointing to a coexistence of n- and p-type carriers in the system. Importantly, we found that both $\rho_{yx}(B)$ and $\rho_{xx}(B)$ present SdH oscillations at high magnetic fields. To remove a large background and make the oscillations more visible, we employed second derivatives. Figure 2(d) shows $d^2\rho_{yx}/dB^2$ measured in tilted magnetic fields and plotted as a function of $B \cos \theta$, where $\theta$ is the angle of the magnetic field from the surface normal. Since the maxima in the oscillations (marked by vertical dashed lines) appear at the same $B \cos \theta$ upon changing $\theta$, the observed SdH oscillations clearly have a two-dimensional (2D) character. An important question is which of the n- or p-type carriers are responsible for the oscillations, and this can answered in the following Landau-level (LL) fan diagram analysis.

To properly construct the LL fan diagram, we use conductance $G_{xx}$ and Hall conductance $G_{xy}$ rather than $\rho_{xx}$ and $\rho_{yx}$. Figure 3(a) shows a plot of $d^2G_{xy}/dB^2$ vs $1/B$, and its Fourier transform (inset) gives the oscillation frequency of 12.3 T, which corresponds to an orbit on the FS with a radius of $k_F = 1.9 \times 10^6$ cm$^{-1}$, giving the 2D carrier density $n_s = 3 \times 10^{11}$ cm$^{-2}$ for each FS and each spin eigenvalue.
dex assignment for a minimum in d^2G_{xy}/dB^2 requires special care, because it depends on the sign of the carriers; if the carriers are electrons, the index should be $N + \frac{1}{2}$, while it should be $N + \frac{3}{2}$ if they are holes [33]. It turns out that the indices assigned according to the electron scenario becomes consistent with the results from $G_{xx}$ [see Fig. 3(b)], and hence one can conclude that the carriers responsible for the SdH oscillations are electrons in the present case. The constructed LL fan diagram crosses the $N$-index axis at 0.55, which gives evidence for the Berry of $\pi$ [43-45]. Therefore, the observed SdH oscillations are obviously due to $n$-type 2D Dirac fermions bearing $\pi$ Berry phase.

The temperature dependence of the SdH amplitude [upper inset of Fig. 3(b)] gives the cyclotron mass $m_c = 0.07m_0$ ($m_0$ is the free electron mass) [46], which in turn gives the Fermi velocity $v_F = 3.2 \times 10^5$ cm/s. For a Dirac-like surface state, this means that the Fermi level is $\sim 40$ meV above the Dirac point (DP). The Dingle analysis [lower inset of Fig. 3(b)] yields the Dingle temperature $T_D$ of 15 K, from which the mean free path of Dirac electrons $l_{SdH} = 26$ nm and their mobility $\mu_{SdH} = 2000$ cm²/Vs are calculated [4].

Now we discuss the origin of the $n$-type Dirac fermions in the present heterostructure system. Both Bi$_2$Te$_3$ and SnTe have topological surface states, and it is useful to consider the energy-band diagram of the heterojunction formed by degenerate $p^+$-SnTe and degenerate $n^+$-Bi$_2$Te$_3$ schematically shown in Fig. 4(a) [33]. Because of the degeneracy on both sides, a tunnel junction is formed at the interface, meaning that SnTe and Bi$_2$Te$_3$ layers are electrically isolated in resistivity measurements with contacts made on the top of the SnTe layer. Note that in Fig. 4(a) the band bending at the interface occurs primarily on the Bi$_2$Te$_3$ side, because the carrier density is lower and screening is much weaker in Bi$_2$Te$_3$ [33]; the hatched area in Fig. 4(a) corresponds to the insulating layer of the tunnel junction. Note also that in this diagram, the Fermi level at the interface lies below the top of the valence band (VB) of Bi$_2$Te$_3$; since the DP of the topological SS in Bi$_2$Te$_3$ is close to the top of the VB [47], it is unlikely that the $n$-type Dirac fermions observed in SdH oscillations come from the SS of Bi$_2$Te$_3$ [48]. Therefore, the only viable possibility is that the top SnTe surface has a sufficient downward band bending [Fig. 4(a)] to host $n$-type Dirac fermions.

Interestingly, such a band bending is actually unavoidable in materials with a partially ionic bonding. For Sn$^{2+}$Te$^{-2}$ films grown in the (111) direction, the stacking sequence of atomic planes is Sn$^{2+}$-Te$^{2-}$-· · ·, which brings about a dipole moment and leads to a diverging electrostatic energy [33]. This situation is known as polar catastrophe [72] and cannot be realized in real materials. In our system, to avoid polar catastrophe, some of the positive charge of the Sn$^{2+}$ atomic plane at the interface with Bi$_2$Te$_3$ is naturally compensated by $n$-type carriers in the $p$-n junction. On the free surface side, the termination is either with Te$^{2-}$ or Sn$^{2+}$ planes. When charge compensation is properly considered, the termination with Te$^{2-}$ plane costs more electrostatic energy, and Sn$^{2+}$ termi-
the bulk and surface mobilities of SnTe, respectively, obtained with the two-band model. (b, c) Temperature dependencies of compensation results in a downward band-bending at the from the two-band analysis.

For Te-terminated (111) surface, all DPs touch the bottom of the conduction band, and it is impossible to real-

FIG. 5: (Color online) (a) $R_{yx}(B)$ behavior at 1.5, 50, 100, 150, 200, 250, and 300 K. Solid lines are fitting of the data with the two-band model. (b, c) Temperature dependencies of the Dirac cones at $\bar{\Gamma}$ and three at $\bar{M}$ which is a TCI, there are four Dirac cones centered at $\bar{\Gamma}$ and three at $\bar{M}$ in the 3D BZ along the [111] direction as schematically shown in Fig. 4(c). The surface band calculations give different results for Te and Sn terminations [50, 51]. For Te-terminated (111) surface, all DPs touch the bottom of the conduction band, and it is impossible to realize $n$-type Dirac fermions irrespective of the position of the Fermi level. For Sn-terminated (111) surface, on the other hand, the DPs are close to the top of the valance band (see Fig. 4(b) adapted from Ref. [50]); in this case, SdH oscillations with a single frequency $F = 12.3$ T from $n$-type Dirac fermions are indeed expected if the Fermi level is at the dashed line in Fig. 4(b), which crosses the Dirac cones at $\bar{\Gamma}$ and $\bar{M}$ at $\sim 0$ and $\sim 40$ meV above the DPs, respectively; this causes the $\bar{\Gamma}$ Dirac cone to give negligible contribution and only the $n$-type carriers on the $\bar{M}$ Dirac cones manifest themselves. The slope of the SS at $\bar{M}$ gives $v_{F}^{M\rightarrow F} = 4.5 \times 10^{7}$ cm/s and $v_{F}^{M\rightarrow F} = 1.4 \times 10^{7}$ cm/s with the geometric average $\langle v_{F} \rangle = 2.5 \times 10^{7}$ cm/s which is close to the value extracted from the SdH oscillations.

Having clarified the origin of the $n$-type 2D Dirac fermions, we now discuss the nonlinear Hall effect. Figure 5(a) shows the Hall resistance $R_{yx}(B)$ measured at various temperatures from 1.5 to 300 K. Symbols are experimental data and solid lines are the results of the fitting of a simple two-band model [4, 13] to the data. The concentration of $n$-type surface carriers is known from the SdH frequency and the number of Dirac cones in the SSs, totaling to $n_{s} = 9 \times 10^{11}$ cm$^{-2}$. The $p$-type carriers must be due mostly to the bulk holes in SnTe as one can see in Fig. 4(a), and their concentration $p = 6.4 \times 10^{20}$ cm$^{-3}$ is determined with little ambiguity from the fitting of the $R_{yx}(B)$ at 300 K where the contribution of surface carriers to $R_{yx}(B)$ becomes very weak. Fixing both $n_{s}$ and $p$, the changes in the bulk and surface mobilities with temperature are reliably obtained from the two-band analysis, as one can infer from the excellent quality of the fittings in Fig. 5(a); note that, in those fittings, the parameters are always constrained so that they correctly reproduce the zero-field value of $\rho_{yx}$ [4].

The obtained mobilities are shown in Figs. 5(b) and 5(c). The bulk hole mobility $\mu_{s}^{H}$ changes only by a factor of two to reach $\sim 40$ cm$^{2}$/Vs at low temperature. On the other hand, the high mobility of surface Dirac electrons, $\mu_{s}^{H} \approx 4000$ cm$^{2}$/Vs at 1.5 K [52], is diminished dramatically with increasing temperature to $\sim 600$ cm$^{2}$/Vs at 300 K, which is the reason for the strong change in the $R_{yx}(B)$ behavior with temperature. The origin of this strong thermally-induced scattering of surface Dirac fermions observed in a TCI is an interesting question to be addressed in future. It is worth mentioning that inclusion of a third contribution to the transport from the $n$-type $\text{Bi}_{2}\text{Te}_{3}$ layer leads to an inconsistency in explaining $R_{yx}(B)$ [53], indicating that $\text{Bi}_{2}\text{Te}_{3}$ layer is really decoupled by the tunnel barrier.

In conclusion, we have epitaxially grown thin films of a TCI material, SnTe, on $\text{Bi}_{2}\text{Te}_{3}$ buffer layer using MBE technique. The high quality of our films and their built-in band bending allow us to probe the surface Dirac electrons residing on the Sn-terminated (111) plane by transport measurements. Those novel thin-film samples open new opportunities for experimentally exploring the physics of TCIs.

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Supplemental Material

S1. Resistivity behavior of Bi$_2$Te$_3$ buffer layer

Our Bi$_2$Te$_3$ films grown on sapphire substrates with MBE are always degenerately n-type doped. Figure S1 shows typical data of the temperature dependence of the resistivity $\rho_{xx}$. The bulk electron density $n_b$ of this films is known from the Hall effect to be $4 \times 10^{19}$ cm$^{-3}$, from which one obtains the bulk mobility of about 400 and 100 cm$^2$/Vs at 1.5 and 300 K, respectively.

S2. Kiessig fringes

Low-angle XRD patterns shown in Fig. 1(c) in the main text demonstrate clear Kiessig oscillations. Generally, such low-angle intensity oscillations are the result of x-ray interference between two interfaces of a thin film, indicating its homogeneity (uniform structure) and smoothness of the interfaces [36, 37]. The position of the $m$-th maximum $\theta_m$ in Kiessig fringes follows [37]

$$\theta_m^2 = \alpha_c^2 + m^2 \left( \frac{\lambda}{2t} \right)^2,$$

where $\alpha_c$ is the critical angle for the total external reflection, $\lambda$ is the x-ray wavelength, and $t$ is the thickness of a film. Since $\alpha_c \approx 0$ for x-rays, the film thickness can be accurately estimated as

$$t \approx \frac{\lambda}{2\Delta},$$

where $\Delta$ is the period of Kiessig oscillations (i.e. fringe distance) in radian unit.

Our samples consist of sapphire (Al$_2$O$_3$) substrate, Bi$_2$Te$_3$ buffer layer, and SnTe layer. The fringe distance observed at grazing incidence angles reflects the total thickness $d_t$ of the film, namely, the thickness of Bi$_2$Te$_3$ layer plus SnTe layer, and indeed, it exactly matches the thicknesses measured by AFM near a sharp edge. Interestingly, in our samples, Kiessig fringes are seen not only at grazing angles, but also near the (003) Bragg peak of Bi$_2$Te$_3$ layer. They have larger fringe distances which are the results of the interference within the Bi$_2$Te$_3$ buffer layer; hence, they testify to the high quality of both Al$_2$O$_3$/Bi$_2$Te$_3$ and Bi$_2$Te$_3$/SnTe interfaces and allow us to estimate the thickness $d_b$ of the Bi$_2$Te$_3$ layer alone. [The top curve in Fig. 1(c) shows the data for Bi$_2$Te$_3$ film without deposition of SnTe, and hence the fringe distances at grazing angles and near the (003) peak are the same.] Since both $d_t$ and $d_b$ are known from those Kiessig fringes, the SnTe layer thickness is accurately estimated by calculating $d_t - d_b$.

S3. Ferroelectric transition in SnTe

SnTe is known to undergo a ferroelectric transition at low temperature, which is associated with a structural phase transition from cubic to rhombohedral [20, 38, 59]. In bulk single crystals, this ferroelectric transition manifests itself

![Graph](image)

FIG. 1: Temperature dependence of $\rho_{xx}$ of a 38-nm-thick Bi$_2$Te$_3$ film without SnTe deposition.
in the temperature dependence of $p_{xx}$ as a kink \cite{38}. In our thin films, however, the resistivity data do not present any feature ascribable to the ferroelectric transition, which suggests that the epitaxial strain suppressed the structural transition. The absence of the ferroelectric transition in strained films implies that the mirror symmetry is kept intact down to low temperature. Actually, even if the ferroelectric transition happens, it will not break the mirror symmetry on the surface of our films, because the structural distortion occurs along the (111) direction which is the same as the growth direction.

S4. Assignment of Landau-level indices

As discussed in detail in Ref. \cite{4}, for the assignment of Landau-level (LL) indices in the data of Shubnikov-de Haas (SdH) oscillations, the best practice is to start with the principle that an integer index $N$ be assigned to a minimum in $G_{xx}$. The index assignment for $G_{xy}$ depends on the sign of the carriers, and the correct assignment is most easily understood by considering the integer quantum Hall effect: For electrons, $G_{xy}$ increases with $B$ between the Hall plateaus and hence $dG_{xy}/dB$ shows a maximum at $N + \frac{1}{2}$, whereas for holes $G_{xy}$ decreases between plateaus to cause $dG_{xy}/dB$ to show a minimum at $N + \frac{1}{2}$. Therefore, when the index assignment is to be done to a minimum in $d^2G_{xy}/dB^2$, the index should be $N + \frac{1}{2}$ if the carriers are electrons, while it should be $N + \frac{3}{2}$ if they are holes. (Note that the LL index decreases with increasing $B$, which is the reason why a minimum in $d^2G_{xy}/dB^2$ should be at $N + \frac{1}{2}$ when a maximum in $dG_{xy}/dB$ is at $N + \frac{3}{2}$.)

S5. Energy band lineups

A heterostructure is formed when a semiconductor is grown on top of another semiconductor. The lineups of the conduction and valence bands at the interface are of fundamental importance for engineering semiconductor devices \cite{33 53}. Since the dawn of semiconductor technology, numerous models and theories have been developed for calculating the energy band offsets. One of the first attempts was the Anderson’s electron affinity rule \cite{54}, which is based on the consideration of the energy balance for an electron moving from the vacuum energy level to the first semiconductor (gaining $\chi_1$), then to the second semiconductor (losing $\Delta E_c$), and then to the vacuum level again (losing $\chi_2$). From the energy conservation, one obtains

$$\Delta E_c = \chi_1 - \chi_2,$$

where $\Delta E_c$ is the conduction-band offset and $\chi_1$ ($\chi_2$) is the electron affinity of the first (second) semiconductor. The valence-band offset follows automatically as

$$\Delta E_v = E_{g2} - E_{g1} - \Delta E_c,$$

where $E_{g1}$ ($E_{g2}$) is the energy gap of the first (second) semiconductor. The electron affinity rule was successful in explaining the band discontinuities in many semiconductor heterostructures, but failed for some, which is due to the following limitations of this model \cite{57}: First, it idealizes the surface of a semiconductor. In real crystals, the surface undergoes a reconstruction in order to reduce the surface energy. The rearrangement of atoms leads to the formation of a surface dipole layer which affects the electron affinity of a semiconductor. Surface defects and surface energy states also play roles. As a result, experimentally measured electron affinities are not very reliable. (This is actually the case with both SnTe and Bi$_2$Te$_3$, for which very different results have been reported in the literature \cite{58 64}.) Second, the surface reconstruction and surface energy states at the interface of two semiconductors are not necessarily the same as those on their free surfaces, making the energy balance consideration to be more complicated. Third, electron correlation effects also influence the electron affinity and have to be taken into consideration.

Over the years, there were many attempts to improve the description of the band offsets at interfaces. One of them is known as “common anion rule” \cite{65 67} and deals with heterostructures which consist of semiconductors having a common anion element. This is exactly the situation in the SnTe/Bi$_2$Te$_3$ interface. This approach is based on the assumption that the valence band is mostly built from the atomic wave functions of cations, while the conduction band is mostly built from the atomic wave functions of anions. Therefore, the valence band of materials with the same anion element should be similar, implying that their valence-band offset in the heterostructure will be smaller than the conduction-band offset. In the energy-band diagram shown in Fig. 4(a) of the main text, we adapted this common anion rule. To plot the band diagram, we used the full depletion approximation with $p = 6 \times 10^{20}$ cm$^{-3}$ and $\epsilon = 1200$ for SnTe, and $n = 4 \times 10^{19}$ cm$^{-3}$ and $\epsilon = 290$ for Bi$_2$Te$_3$. Those parameters are chosen to reflect the known carrier densities in the two layers, and they lead to the situation that the band bending occurs primarily on the Bi$_2$Te$_3$ side of the interface.
it becomes immediately clear that either the density $p$ increased from the two-band case, to compensate for the additional contribution of the bulk moderately high density and mobility. However, the resulting combination of the parameters will never be consistent oscillations, and hence the only parameters that can be significantly varied are those for the bulk $R$ bulk carriers of Bi. However, if the semimetal situation is realized, one needs to consider the additional contribution from the $n$-type carriers would contribute to the transport. We know from measurements of our 38-nm-thick Bi$_2$Te$_3$ at 1.5 K is about 400 cm$^2$/Vs. Hence, if the semimetal situation is realized, those electrons provide the bulk n-type channel. Note that the observed SdH oscillations from $n$-type carriers cannot be ascribed to those bulk electrons of the Bi$_2$Te$_3$ layer because of their 2D-like angle dependence and, more importantly, their low frequency which is two orders of magnitude smaller than what is expected from the 3D FS of Bi$_2$Te$_3$ with $n_b \simeq 4 \times 10^{19}$ cm$^{-3}$. Therefore, if the semimetal situation is realized, there should be two different $n$-type channels, along with the $p$-type bulk channel of SnTe. However, as we show below, the existence of such three channels cannot be consistent with the transport data.

The difficulty is most easily understood by considering the situation at 1.5 K. In Fig. 5(a) of the main text, the $R_{yx}(B)$ data at 1.5 K is strongly nonlinear and shows a sign change in slope with $B$. This behavior is already excellently explained by the coexistence of a low density ($n_s = 9 \times 10^{11}$ cm$^{-2}$) of high-mobility ($\mu^H_s \simeq 4000$ cm$^2$/Vs) $n$-type 2D carriers and a high density ($\rho = 6.4 \times 10^{20}$ cm$^{-3}$) of low-mobility ($\mu^L_b \simeq 40$ cm$^2$/Vs) $p$-type bulk carriers. However, if the semimetal situation is realized, one needs to consider the additional contribution from the $n$-type bulk carriers of Bi$_2$Te$_3$, whose density ($n_b \simeq 4 \times 10^{19}$ cm$^{-3}$) and mobility ($\sim 400$ cm$^2$/Vs) are reasonably known and are moderately high. Such an additional contribution demands a significant revision of the analysis of the $R_{yx}(B)$ behavior. Nevertheless, the parameters of the 2D $n$-type channel is constrained from the analysis of the SdH oscillations, and hence the only parameters that can be significantly varied are those for the bulk $p$-type channel.

When one tries to reproduce the $R_{yx}(B)$ behavior at 1.5 K using a three-band model with the above constraints, it becomes immediately clear that either the density $\rho$ or the mobility $\mu^L_b$ of the $p$-type carriers must be significantly increased from the two-band case, to compensate for the additional contribution of the bulk $n$-type carriers with moderately high density and mobility. However, the resulting combination of the parameters will never be consistent.

Another approach to improve the simple electron affinity rule is the effective dipole model [68-70], which includes the effects of the dipole charge formation due to a local difference in the atomic (and electronic) structures at the interface in comparison with the bulk structures of constituent semiconductors.

Generally, there are only three possible band lineups [54] which are shown in Fig. S2. The straddling lineup with conduction- and valence-band offsets of opposite sign is the most common one. In the main text, we consider the heterostructure of this type. The staggered lineup has conduction- and valence-band offsets of the same sign, which does not make any qualitative difference in our discussions. The broken-gap lineup is an extreme case of the staggered one, in which the bottom of the conduction band of one semiconductor goes below the top of the valence band of another semiconductor. This is the most exotic lineup and it is realized in at least one nearly-lattice-matched heterostructure, InAs/GaSb [71]. Another possible example, where the broken-gap lineup has been invoked to explain the coexistence of $n$- and $p$-type carriers, is the case of quantum-well structures and superlattices made of IV-VI semiconductors (such as PbTe/SnTe) for thermoelectric applications [61–63]. Interestingly, after the discovery that SnTe is a topological crystalline insulator (TCI) [7], we now understand that PbTe/SnTe or PbSe/SnTe heterostructures must carry a (possibly $n$-type) metallic state at the interface, because it is a boundary between topologically trivial and nontrivial phases. However, this possibility was never considered because the idea of TCI was not yet developed at the time.

S6. Problem with the broken-gap-lineup scenario

It is important to know whether we can understand the transport data described in the main text if SnTe/Bi$_2$Te$_3$ interface were of the broken-gap type. In this case, there will be no electrical isolation between the two layers and the heterostructure will behave as a semimetal [63], in which both $p$-type bulk SnTe carriers and $n$-type bulk Bi$_2$Te$_3$ carriers would contribute to the transport. We know from measurements of our 38-nm-thick Bi$_2$Te$_3$ films (without deposition of SnTe) that the Bi$_2$Te$_3$ buffer layer contains $n_b \simeq 4 \times 10^{19}$ cm$^{-3}$ of bulk electrons and their mobility at 1.5 K is about 400 cm$^2$/Vs. Hence, if the semimetal situation is realized, those electrons provide the bulk $n$-type channel. Note that the observed SdH oscillations from $n$-type carriers cannot be ascribed to those bulk electrons of the Bi$_2$Te$_3$ layer because of their 2D-like angle dependence and, more importantly, their low frequency which is two orders of magnitude smaller than what is expected from the 3D FS of Bi$_2$Te$_3$ with $n_b \simeq 4 \times 10^{19}$ cm$^{-3}$. Therefore, if the semimetal situation is realized, there should be two different $n$-type channels, along with the $p$-type bulk channel of SnTe. However, as we show below, the existence of such three channels cannot be consistent with the transport data.

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When one tries to reproduce the $R_{yx}(B)$ behavior at 1.5 K using a three-band model with the above constraints, it becomes immediately clear that either the density $\rho$ or the mobility $\mu^L_b$ of the $p$-type carriers must be significantly increased from the two-band case, to compensate for the additional contribution of the bulk $n$-type carriers with moderately high density and mobility. However, the resulting combination of the parameters will never be consistent.

FIG. 2: Schematic picture of three possible types of band alignments: (a) straddling, (b) staggered, and (c) broken-gap band lineups.
with the zero-field value of $\rho_{xx}$, since we now have an additional $n$-type conduction channel and also the conductivity of the $p$-type channel has been significantly enhanced, both of which make the calculated $\rho_{xx}$ value to be significantly lower than is actually observed. Therefore, it is impossible to consistently analyze the transport data by assuming that a bulk $n$-type channel with moderately high density and mobility contributes to the transport. This in turn means that the broken-gap-lineup scenario, which necessarily gives rise to a bulk semimetal behavior, is inappropriate for the present system.

**S7. Te-termination vs Sn-termination**

SnTe is a material with a partially ionic bonding. For films grown in the (111) direction, the stacking sequence of atomic planes is $\text{Sn}^{2+}$-$\text{Te}^{2-}$-$\text{Sn}^{2+}$-$\text{Te}^{2-}$-$\cdots$. Each uniformly charged atomic plane generates the electric field

$$E = \frac{\sigma}{2\epsilon\epsilon_0}$$

where $+\sigma (-\sigma)$ is the charge density on the $\text{Sn}^{2+}$ ($\text{Te}^{2-}$) plane, $\epsilon_0$ is the permittivity of vacuum, and $\epsilon$ is the permittivity of SnTe. Such a charge distribution brings about a finite electric field between each pair of $\text{Sn}^{2+}$ and $\text{Te}^{2-}$ planes, and, hence, a monotonic increase in the electrostatic energy as shown in Fig. S3. For increasing number of atomic layers along the polar direction, the electrostatic energy diverges. This situation is known as a polar catastrophe and cannot be realized in real materials [72]. One way to avoid the polar catastrophe is to partially compensate the charge on the surface of SnTe by mobile carriers as shown in Figs. S4 and S5.

In our system, the growth of SnTe film starts with a $\text{Sn}^{2+}$ atomic plane, which should be compensated with a negative charge. This is naturally realized due to a $p-n$ junction formed with the $n$-type Bi$_2$Te$_3$ buffer layer. If the free surface is terminated with Te, the negative charge of the outermost $\text{Te}^{2-}$ plane must be compensated by positive charge (Fig. S4). The charge compensation makes $E$ to change between positive and negative values along $z$, and the resulting profile of $V$ just oscillates between zero and a finite positive value. Nevertheless, in this Te-terminated case, there remains a finite electrostatic potential $V_e$ at the outer surface (Fig. S4).

On the other hand, if the free surface is terminated with Sn, the positive charge of the outermost $\text{Sn}^{2+}$ plane must be compensated by negative charge (Fig. S5). Moreover, for the charge-compensated Sn-terminated surface, the remaining electrostatic potential can be exactly zero (Fig. S5). Compared with a finite $V_e$ in the case of Te-terminated surface (Fig S4), the Sn-termination results in lower electrostatic energy and thus is energetically favored. This means that our films are most likely terminated with a $\text{Sn}^{2+}$ atomic plane.

We note that our transport data points to the existence of a small density of high-mobility $n$-type carriers in the heterostructure, which is only possible when the outer surface has a downward band bending to cause the topological

![FIG. 3: The polar catastrophe. A schematic charge distribution in idealized ionic SnTe along the (111) direction is shown together with the resulting variation of the electric field $E$ and the electrostatic potential $V$ along the (111) direction denoted as $z$. Note the monotonic increase in $V$ with increasing number of atomic planes.](image-url)
FIG. 4: The situation for Te-terminated SnTe film with charge compensation. A finite electrostatic potential $V_e$ remains at the outer surface.

FIG. 5: The situation for Sn-terminated SnTe film with charge compensation. Remaining electrostatic potential $V_e$ is zero.

Surface state to be doped with $n$-type carriers. Such a band bending is actually expected for the Sn-terminated surface which is compensated with negative charge, but the Te-terminated surface will have an upward band-bending due to its compensation with positive charge. Therefore, the conclusion about the termination based on the electrostatic energy argument is supported by the transport results.
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