Observation of ultrahigh mobility surface states in a topological crystalline insulator by infrared spectroscopy

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Topological crystalline insulators possess metallic surface states protected by crystalline symmetry, which are a versatile platform for exploring topological phenomena and potential applications. However, progress in this field has been hindered by the challenge to probe optical and transport properties of the surface states owing to the presence of bulk carriers. Here, we report infrared reflectance measurements of a topological crystalline insulator, (001)-oriented Pb1−xSnxSe in zero and high magnetic fields. We demonstrate that the far-infrared conductivity is unexpectedly dominated by the surface states as a result of their unique band structure and the consequent small infrared penetration depth. Moreover, our experiments yield a surface mobility of 40,000 cm² V⁻¹ s⁻¹, which is one of the highest reported values in topological materials, suggesting the viability of surface-dominated conduction in thin topological crystalline insulator crystals. These findings pave the way for exploring many exotic transport and optical phenomena and applications predicted for topological crystalline insulators.
Recently, a new class of insulators called topological crystalline insulators (TCIs) were predicted and observed in IV–VI semiconductors\(^1\)–\(^8\), which have attracted tremendous scientific interest\(^8\)–\(^17\). These materials host gapless Dirac-like surface states (SS) that are protected by crystalline symmetry\(^1\)–\(^2\), instead of time-reversal symmetry\(^1\)–\(^2\),\(^18\)–\(^20\). Consequently, breaking the crystal symmetry can open a band gap in these SS\(^1\)–\(^2\), offering new opportunities for band gap engineering by strain or structural distortion. Moreover, compared with \(Z_2\) topological insulators\(^1\)–\(^2\),\(^18\)–\(^20\), the (001) SS of TCIs have been predicted to exhibit a wider range of tunable electronic properties under many types of perturbations breaking the crystalline symmetry, such as interface superconductivity\(^3\), spin-filtered edge states\(^10\), quantum anomalous Hall effect\(^11\), Weyl fermions\(^13\), and valley-dependent optical properties\(^14\). Therefore, TCIs are emerging as a very versatile material system not only for exploring topological quantum phenomena, but also for potential device applications in electronics, spintronics, and optoelectronics\(^8\)–\(^14\).

Several novel characteristics of TCIs have been revealed by surface sensitive probes\(^3\)–\(^6\),\(^12\). However, the transport and optical properties of the (001) SS, which are fundamentally important and arguably most relevant to applications\(^8\)–\(^14\), have remained especially challenging to measure because of the overwhelming effects of bulk carriers in previous studies\(^13\)–\(^17\). This has seriously hampered the progress in this field.

Here, we present infrared (IR) reflectance measurements of a TCI, \(\text{Pb}_1-x\text{Sn}_x\text{Se}\) \((x = 0.23–0.25)\) single crystals with (001) surface in zero and high magnetic fields. From the band gap, Fermi velocity and Fermi energy of the bulk bands determined from our data, the bulk Drude spectral weight (SW) can be estimated, which is found to be much less than the measured Drude weight, indicating substantial contributions from surface carriers. Secondly, the spectral features in magneto-reflectance spectra below 25 meV can be attributed to a dominant resonance at \(\omega_{\text{ss}} \times B\) based on theoretical study of cyclotron resonance (CR) of the SS, the frequency of which obtained from our data \(\omega_{\text{ss}}\) is quantitatively consistent with those estimated from previous scanning tunneling microscopy (STM) and angle-resolved photoemission spectroscopy (ARPES) experiments. Moreover, the SW of the dominant resonance in magnetic field is in accord with the extra Drude weight in zero field besides the bulk contribution. Above all, we demonstrate that the resonance at \(\omega_{\text{ss}}\) in field is well below the energy range of all Landau level (LL) transitions (including CR) from the bulk states, so it can only be assigned to the SS. Therefore, all these findings taken together provide robust evidence for SS in \(\text{Pb}_1-x\text{Sn}_x\text{Se}\). Remarkably, we find that the IR conductivity of \(\text{Pb}_1-x\text{Sn}_x\text{Se}\) is dominated by the SS in the far-IR range \((7–25\,\text{meV} \text{ or } 2–6\,\text{THz})\) despite the presence of bulk carriers. We show that this unexpected property arises from the unique band structure of the SS and the resultant high surface carrier density and small IR penetration depth. Furthermore, our experiments yield a surface mobility of \(~40,000\,\text{cm}^2\,\text{V}^{-1}\,\text{s}^{-1}\) based on analysis of CR mode of the SS, which is 1–2 orders of magnitude higher than that in TCI thin films\(^21\)–\(^23\) and among the highest reported values in topological materials. The ultrahigh surface mobility and other transport parameters obtained here suggest that surface-dominated transport can be achieved in (001)-oriented \(\text{Pb}_{1-x}\text{Sn}_x\text{Se}\) crystals with sub-micron thickness. Our findings open up opportunities for exploring many exotic transport and optical phenomena and applications predicted for TCIs\(^8\)–\(^14\), ranging from quantum anomalous Hall effect and Weyl fermions to spintronics and valleytronics.

**Results**

**IR spectra of \(\text{Pb}_1-x\text{Sn}_x\text{Se}\).** In this work, \(\text{Pb}_1-x\text{Sn}_x\text{Se}\) single crystals with an actual composition of \(x = 0.23–0.25\) were investigated, which are in the TCI phase and host gapless SS\(^4\) (Fig. 1a). The actual composition is determined from the bulk band gap\(^4\) discussed below. These materials have a direct bulk band gap located at four L points (valleys) in the three-dimensional (3D) Brillouin zone. Our samples are n-doped with the Fermi energy \(E_F\) in the bulk conduction band\(^3\). The IR reflectance spectra \(R(\omega)\) of \(\text{Pb}_1-x\text{Sn}_x\text{Se}\) crystals with (001) surface were measured in zero field and in magnetic field applied perpendicular to the surface of the samples. Figure 1 depicts the \(R(\omega)\) spectrum and the dissipative part of the optical conductivity \(\sigma(\omega)\) in zero field at \(T = 8\,\text{K}\) (see “Methods”). The \(R(\omega)\) spectrum shows a typical metallic behavior: \(R(\omega)\) is very close to 1 below 15 meV; at higher energy the reflectance is gradually depressed toward a plasma minimum at about 30 meV, followed by a peak around 120 meV. The energy of the plasma minimum observed here is much lower than those in earlier IR studies\(^16\),\(^17\), indicating much lower total carrier density in our samples due to low density of Se vacancy defects (see “Methods” and Supplementary Note 1). The zero field \(\sigma(\omega)\) spectrum exhibits a Drude component below 25 meV and a threshold-like feature around 100 meV. The narrow plasma minimum in \(R(\omega)\) with a half width \(~3\,\text{meV} \text{ or } 0.3\,\text{THz}\) is a direct manifestation of the very narrow Drude peak in \(\sigma(\omega)\), which is corresponding to a very low scattering rate. The threshold feature in \(\sigma(\omega)\) can be assigned to the onset of inter-band transitions for the bulk around \(E_{\text{inter}} \approx \Delta + E_F \{1 + \frac{m_s}{m}\}\) as illustrated in the inset of Fig. 1c, where \(\Delta\) is the bulk band gap, the Fermi energy \(E_F\) is defined with respect to the bottom of the conduction band, \(m_s\) and \(m\) are effective masses of the conduction and valence band, respectively. The experimental absorption coefficient spectrum

![Fig. 1](https://example.com/fig1.png)
also exhibits a similar threshold feature in the same energy range. It is shown that in doped semiconductors the absorption coefficient can be written as
\[ \alpha(\omega) \propto \alpha_0(\omega) |1 - f(\omega, E_F, T)|, \]
where \( \alpha_0(\omega) \propto \sqrt{\hbar \omega - \Delta/\hbar} \) is the absorption coefficient for the undoped material, \( f(\omega, E_F, T) \) is Planck’s constant divided by \( 2 \pi k_B T \) and is the Boltzmann constant. Fitting the experimental absorption coefficient using the equation above, we find \( E_{\text{inter}} \sim 100 \text{ meV} \) as indicated by the dashed line in Fig. 1c.

More insights into the band structure of Pb\textsubscript{1-x}Sn\textsubscript{x}Se can be provided by a systematic investigation of the magneto-reflection \( R(\omega, B) \) spectra displayed in Fig. 2 (see “Methods”). Strikingly, the zero field \( R(\omega) \) spectrum is strongly modified by the magnetic field. The \( R(\omega, B) \) spectra are strongly suppressed below 25 meV in magnetic fields and no longer extrapolate to unity in the limit of \( \omega \rightarrow 0 \). Moreover, a series of new resonance features are observed in \( R(\omega, B) \) extending up to 300 meV, all of which evolve systematically with magnetic fields. In order to acquire a complete understanding of the features in \( R(\omega, B) \) spectra, the optical conductivity in magnetic field was extracted from an analysis of \( R(\omega, B) \) using the magneto-Drude-Lorentz model\textsuperscript{26-28} (see “Methods”). Figure 3a depicts the real part of the optical conductivity \( \sigma_0(\omega, B) \) from 50 to 300 meV, which exhibits several resonance (absorption) peaks systematically shifting to higher energies with increasing magnetic field. The energies \( E(k) \) of the absorption features in \( \sigma_0(\omega, B) \) are displayed in Fig. 3b, which show good agreement with the energies of the resonance features in \( R(\omega, B) \). The field-dependent absorption features can be assigned to LL transitions. Interestingly, the observed transitions are not equally spaced in energy in any spectrum, which is in stark contrast to the behavior of systems with quadratic energy-momentum dispersion. We find that the observed resonance features in \( \sigma_0(\omega, B) \) can be well described by LL transitions of 3D massive Dirac fermions\textsuperscript{8, 15}, whose LL energies have the form:

\[
E_n(k_z) = \pm \delta \delta_{n,0} \sqrt{\left( \frac{\Delta}{2} \right)^2 + (\hbar v_F k_z)^2} + \text{sgn}(n) \sqrt{2 \hbar v_F B |n| \left( \frac{\Delta}{2} \right)^2 + (\hbar v_F k_z)^2}
\]

where have two zeroth LLs labeled as \( n = +1 \) and \( n = -1 \). Here, the integer \( n \) is LL index, \( k_z \) is momentum along the direction of the magnetic field, \( e \) is the elementary charge, \( v_F \) is the Fermi velocity, \( \delta \) is the Kronecker delta function, and \( \text{sgn}(n) \) is the sign function. Theoretical studies showed that the optical conductivity has sharp peaks at energies of LL transitions at \( k_z = 0 \) because of singularities in the joint-density-of-states between LLs at these energies\textsuperscript{29}. From the selection rule\textsuperscript{29, 30} for allowed optical transitions from \( \text{LL}_n \) to \( \text{LL}_{n'} \), \( \Delta n = |n| - |n'| = \pm 1 \), we find that all the observed resonances can be assigned to allowed LL transitions based on Eq. (1) with \( k_z = 0 \) and \( \Delta \approx 64 \pm 3 \text{ meV} \), and \( v_F \approx (0.400 \pm 0.005) \times 10^6 \text{ m s}^{-1} \). Figure 3b, c, which are determined from least squares fit of the observed transition energies. This analysis shows that the sharp resonances in Fig. 3a mainly arise from massive Dirac fermions of the bulk states\textsuperscript{8, 15}. Since the Sn substitution level directly leads to changes in the bulk band gap, the actual composition of our samples can be determined from \( \Delta \) to be \( x = 0.23 - 0.25 \).

**IR signatures of SS.** Our data allow us to identify the signatures of SS in Pb\textsubscript{1-x}Sn\textsubscript{x}Se because all spectroscopic features of the bulk states can be determined. The first evidence for the SS is from an analysis of the Drude SW (area under \( \sigma_1(\omega) \)) in zero field. The low-energy bulk states can be described by massive Dirac fermions:

\[
E(k_z) = \pm \sqrt{\hbar^2 v_F^2 k_z^2 + (\Delta/2)^2},
\]

where \( \Delta \) and \( v_F \) are given above. We can estimate \( E_F \) of the bulk states from \( E_{\text{inter}} \approx \Delta + 2E_F \) (note that \( m_c = m_v \) for massive Dirac fermions) shown in Fig. 1c. Alternatively, the transition \( \text{LL}_0 \rightarrow \text{LL}_1 \) disappears at \( B < 6 \text{ T} \) (Figs. 2, 3), so the Fermi energy can be estimated as \( E_F \approx \text{LL}_1(B = 6 \text{ T}) - \Delta/2 \) using Eq. (1). Either method yields \( E_F \approx 17 \pm 2 \text{ meV} \). From the band dispersion and \( E_F \), we obtain \( k_F \approx 0.014 \pm 0.001 \text{ Å}^{-1} \). Recent transport studies found that the bulk Fermi surface pockets of Pb\textsubscript{1-x}Sn\textsubscript{x}Se in the TCI phase are nominally spherical\textsuperscript{15}, so the bulk carrier density can be estimated by

\[
\frac{n_{\text{bulk}}}{2} = \frac{\pi \kappa_F^3 g_s}{2} \approx 3.7 \pm 1.1 \times 10^{12} \text{ cm}^{-3},
\]

where \( g_s = 2 \) and \( g_v = 4 \) are the bulk spin and valley degeneracy, respectively. Similarly low carrier density has been reported by previous transport experiments\textsuperscript{15}. Moreover, the effective mass of massive Dirac fermions at \( E_F \) can be calculated from

\[
n_{\text{bulk}} = \frac{\hbar^2 + \Delta^2}{2} \approx 0.054 \pm 0.003 m_v,
\]

where \( m_v \) is bare electron mass. The SW of the Drude absorption is related to the bare plasma frequency \( \omega_p^2 = 4 \pi e^2 n/m \) by\textsuperscript{31}:

\[
\text{SW} = \int_{0}^{\omega_0} \sigma_1(\omega) d\omega = \frac{\pi \Omega^{-1}}{120} \omega_p^2
\]

where \( \omega_0 \) is a cut-off frequency separating the Drude component from the inter-band transitions, \( \Omega \) is Ohm, \( \sigma_1 \) is in
\( \Omega^{-1} \text{cm}^{-1} \), and all frequencies are in \( \text{cm}^{-1} \). From \( \rho_{\text{bulk}} \), \( \rho_{\text{bulk}} \), and Eq. (2), we estimate that \( \Delta \Omega_{\text{bulk}} \approx (1.6 \pm 0.5) \times 10^{4} \Omega^{-1} \text{cm}^{-2} \) for the bulk states from calculating \( \rho_{\text{bulk}} \). On the other hand, the total Drude SW can be determined by the total plasma frequency \( \omega_{p} \) of the Drude component in the experimental data using \( \Delta \Omega_{\text{bulk}} = \frac{2\pi e^{2}}{m_{e}^{*}} \), where \( \omega_{p} \) is related to the screened plasma frequency \( \omega_{p} \) by \( \omega_{p} = \omega_{p} / \varepsilon_{\infty} \), with \( \omega_{p} \approx 260 \text{cm}^{-1} \) (32.4 meV) corresponding to the plasma minimum in \( \varepsilon_{\infty} \) and \( \varepsilon_{\infty} \) representing all electronic contributions to the dielectric constant other than the Drude component. Therefore, the total plasma frequency \( \omega_{p} \) can be obtained from \( \varepsilon_{\infty} \), which is determined by all Lorentzian oscillators obtained from Drude-Lorentz fit of the entire \( \varepsilon(\omega) \) spectrum (see Supplementary Note 5). We find that \( \varepsilon_{\infty} \approx 45 \pm 9, \) so \( \Delta \Omega_{\text{bulk}} \approx (7.9 \pm 1.6) \times 10^{4} \Omega^{-1} \text{cm}^{-2} \). A comparison between \( \Delta \Omega_{\text{bulk}} \) and \( \Delta \Omega_{\text{total}} \) shows that the bulk contribution can only account for about 20% of the total Drude SW of the system. We emphasize that the total Drude SW and that from bulk states are both accurately determined from our data, which strongly suggests dominant surface contribution to the total Drude absorption in zero field.

The low-energy magneto-reflection of \( \text{Pb}_{1-x}\text{Sn}_{x}\text{Se} \) provides further evidence for the SS. The systematic suppression of \( \varepsilon(\omega, B) \) spectrum with \( B \) field below 25 meV suggests that the system becomes more insulating with increasing field. This is reminiscent of the typical behavior of CR. For instance, similar low-energy behaviors in \( \varepsilon(\omega, B) \) were also observed in graphite \(^{27} \) due to CR, which manifests itself in \( \varepsilon(\omega, B) \) as a peak at energies of \( E \alpha B \). We now show that the spectral feature below 25 meV in \( \varepsilon(\omega, B) \) and its evolution with \( B \) field are unambiguous signatures of CR from the SS (Supplementary Notes 2 and 3). As shown in Figs. 1a and 4a, the band structure of SS on the (001) surface features two generations of Dirac fermions \(^{7} \), starting from a pair of Dirac cones located at the \( \Gamma \) points of the surface Brillouin zone with their Dirac points at \( E_{\Gamma}^{\text{DP}} \) and \( E_{\Gamma}^{\text{DP}} \). The hybridization between these two Dirac cones leads to a gap in all directions except along the \( \Gamma\Delta \) line (Fig. 1a), where a pair of Dirac points exist that are protected by the (110) mirror symmetry \(^{2-7} \). Recent experiments and theoretical calculations \(^{3,22,33} \) suggest that \( E_{\Gamma}^{\text{DP}} \) is close to \( E_{F} \) of the bulk states for \( \text{Pb}_{1-x}\text{Sn}_{x}\text{Se} \). In a magnetic field perpendicular to the surface, it is shown \(^{32} \) that the LLs of the SS near \( E_{F} \) are well approximated by LLs of two independent Dirac cones at \( E_{\Gamma}^{\text{DP}} \) and \( E_{\Gamma}^{\text{DP}} \) (Fig. 4b), which have energies of \( E_{n} = \text{sign}(n) \sqrt{2 \hbar v_{F}/\mu_{n}c} \) with respect to each Dirac point and \( P_{n} \sim 0.4 \times 10^{4} \text{m s}^{-1} \). Within this picture, a CR mode due to the intra-band LL transition associated with the Dirac cone at \( E_{\Gamma}^{0} \) is expected at low energy (Fig. 4b). The surface Fermi energy with respect to \( E_{\Gamma}^{0} \) is estimated to be \( E_{\Gamma}^{0} \sim 136 \pm 14 \text{ meV} \) based on STM and ARPES experiments \(^{3,32,33} \), from which the energy of the surface CR mode is expected to be \( E_{n} = \text{sign}(n) \sqrt{2 \hbar v_{F}/\mu_{n}c} \) with \( m_{n} = E_{n}^{0} / (v_{F}^{0})^{2} = 0.15 \pm 0.015 m_{e} \). LL transitions of SS associated with the Dirac cone at \( E_{\Gamma}^{0} \) can also contribute to the \( R(\omega, B) \) spectra at \( \sqrt{2 \hbar v_{F}^{0} / B} \) or higher energy, but it is very difficult to separate them from bulk LL transitions that are in the same energy range. It is shown \(^{32} \) that LLs associated with the Dirac cone along the \( \Gamma\Delta \) line are restricted in the energy range between \( E_{\Gamma\Delta} \) and \( E_{\Gamma\Delta} \) in Fig. 4a, b, so these LLs below \( E_{F} \) cannot be probed from optical measurements, which requires optical transitions from a LL below \( E_{F} \) to one above \( E_{F} \). To examine the IR signatures of the surface CR mode, the magneto-Drude-Lorentz model is used to simulate the \( R(\omega, B) \) spectra (see “Methods”). The real and imaginary parts of \( \sigma_{ex}(\omega) \), \( \sigma_{xy}(\omega) \), \( \sigma_{y\omega}(\omega) \), and \( \sigma_{z\omega}(\omega) \) in representative models are displayed and discussed in Supplementary Note 2. We find that the principle features of \( R(\omega, B) \) can be reproduced by model optical conductivity spectra that are based on the expected behaviors of the surface CR mode \(^{32} \): remarkably, a strong resonance peak at \( \omega_{r} \) in \( \sigma_{ex}(\omega, B) \) that changes linearly with \( B \) field (Fig. 4c) can capture the spectral feature in \( R(\omega, B) \) below 25 meV and its evolution with \( B \) field. In particular, the analysis on our data yields \( m_{n} \) values in the range of 0.15–0.19 \( m_{e} \) for the CR mode, which are consistent with those estimated from STM and ARPES experiments \(^{3,33} \) within 15% as shown in Fig. 4d (see Supplementary Note 2). The small deviation between our results and STM and ARPES measurements may arise from the difference in Fermi energy in different samples. Based on LL...
Fig. 4 IR conductivity of SS in magnetic fields. a A schematic of the SS band structure (dark blue) and that of the bulk (red) along two high-symmetry directions for one of the four Dirac cones inside the surface Brillouin zone. \(E_{\text{DP}}\), \(E_{\text{VHS1}}\), and \(E_{\text{VHS2}}\) are energies of the Dirac points associated with the two Dirac cones located at the X points and the Dirac nodes along the \(\Gamma X\) line, respectively. \(E_{\text{VHS1}}\) and \(E_{\text{VHS2}}\) are energies of the two Van Hove singularities in the band structure. b A schematic of LLs of the SS in the same energy scale as that in a. While the LLs between \(E_{\text{VHS1}}\) and \(E_{\text{VHS2}}\) in the gray-shaded area have nontrivial dispersions (not shown here), the LLs near \(E_{\text{F}}\) are well approximated by LLs of two independent Dirac cones at \(E_{\text{DP}}\) and \(E_{\text{DP}}\) as illustrated by the dashed lines in the shaded area. The CR of the SS associated with the Dirac cone at \(E_{\text{DP}}\) is shown by the vertical arrows. c The real part of the model optical conductivity \(\sigma_{xx}(\omega)\) in several magnetic fields that are used to simulate the \(R(\omega, B)\) spectra below 70 meV. The strong peak below 15 meV in \(\sigma_{xx}(\omega)\) arises from CR of the SS, which represents the spatially averaged 3D optical conductivity of the SS within the IR penetration depth. d The CR energy of SS \(\omega_{c}^n\) as a function of magnetic field. The error bars represent the range of \(\omega_{c}^n\) values that can be used in our model to produce excellent fit of the \(R(\omega, B)\) data within experimental uncertainties as discussed in Supplementary Note 2. The shaded area indicates the range of \(\omega_{c}^n\) values estimated from previous STM and ARPES experiments.

energies calculated for \(\Delta\) and \(v_F\) for the bulk and Eq. (1), the peak between 30 and 50 meV in \(\sigma_{xx}(\omega)\) spectra can be assigned to the LL\(_{0} \rightarrow\) LL\(_{1}\) transition (intra-band LL transition or CR) from the bulk states for \(B > 6\) T, which is responsible for the feature around 37 meV in \(R(\omega, B)\). Because \(E_F\) is between LL\(_{0}\) and LL\(_{1}\) above \(B > 6\) T, the bulk CR LL\(_{0} \rightarrow\) LL\(_{1}\) around 37 meV has the lowest energy among all allowed LL transitions for the bulk for \(B > 6\) T (Fig. 3c). Therefore, the spectral feature in \(R(\omega, B)\) below 25 meV can only be assigned to the SS, because it is well below the energy range of all LL transitions (including CR) from the bulk states for \(B > 6\) T. Moreover, from the area under the resonance at \(\omega_{c}^n\) in \(\sigma_{xx}(\omega)\), the SW of this resonance is found to be \(\text{SW}_{\text{SS}} = (4.4 \pm 0.9) \times 10^{-4} \, \Omega^{-1} \, \text{cm}^{-2}\), which is in agreement with the extra Drude SW in zero field besides the bulk contribution \((6.3 \pm 2.1) \times 10^{-4} \, \Omega^{-1} \, \text{cm}^{-2}\). This agreement provides further support for our identification of the surface CR mode. Therefore, our data in zero and high magnetic fields taken together provide robust evidence for the IR signatures of SS in Pb\(_{1-x}\)Sn\(_x\).Se.

**SS dominated far-IR conductivity.** Strikingly, the far-IR conductivity of (001)-oriented Pb\(_{1-x}\)Sn\(_x\)Se is dominated by the SS in the energy range of the Drude absorption in zero field and that of the surface CR mode in magnetic field, which is around 7–25 meV or 2–6 THz. We now show that this is a consequence of the unique band structure of the (001) SS. The SS of our n-doped samples has two Fermi surfaces associated with the two Dirac cones at \(E_{\text{DP}}\) and \(E_{\text{DP}}\) (Fig. 4a). In particular, the latter Dirac cone gives rise to a large Fermi surface with \(k_F \sim\)
$E_{SS}^2/(h\nu_{SS}^2) \sim 0.052 \pm 0.005 \text{ A}^{-1}$. Neglecting the small contribution from the Fermi surface associated with the Dirac cones at $E_{DP}^{\text{DP}}$ and taking into account $g_{\text{e}} = 1$ and $g_{\text{h}} = 2$ for the SS, we estimate that the large SS Fermi surface leads to a very high surface carrier density of $n_{SS}^2 \sim (4.2 \pm 0.8) \times 10^{12} \text{ cm}^{-2}$, which is more than one order of magnitude higher than that of the (111) surface in TCI's. The large value of $n_{SS}^2$ of the (001) surface is critical for achieving surface dominated far-IR conductivity in Pb$_1$\textsubscript{−}SnSe despite the presence of bulk carriers. Our measurements only probe the region of the sample within the IR penetration depth $\delta$. An estimate for the length scale of $\delta$ for our samples between 7 and 25 meV can be obtained from a SW analysis. The Drude weight of the (001) SS can be estimated as $\sigma_{\text{DP}}^2 \tilde{\gamma}_{\text{DP}}/(8h^2)$, which only includes contributions from the massless Dirac fermions associated with the Dirac cone at $E_{DP}^{\text{DP}}$. The same surface Drude weight can be obtained from $\nu_{SS}^2 \delta$ based on our IR measurements, so the averaged length scale of $\delta$ between 7 and 25 meV is estimated to be $13 \pm 4$ nm for our samples. It can be shown that $\delta$ is comparable to the penetration depth of the SS (see “Methods”). This suggests that the high-carrier density of the (001) SS leads to strong IR absorption and attenuates the IR light within a very small penetration depth, which effectively reduces the bulk contribution to the far-IR conductivity. It is instructive to compare our results with a recent IR study of (111)-oriented TCI thin films, which reported substantially lower surface SW compared to the bulk contribution. It is found that the (111) surface carrier density is ten times lower and the IR penetration depth is much larger than those in our samples, owing to a much smaller $k_s$ for the (111) SS. These results are consistent with our findings that the high surface carrier density reduces the IR penetration depth and thus the bulk contribution to the overall far-IR conductivity in (001)-oriented Pb$_1$\textsubscript{−}SnSe. Therefore, the unique band structure of (001) SS plays a crucial role in achieving surface-dominated far-IR conductivity observed in our study. The large difference in the IR properties of (001) and (111) surfaces is a new aspect in TCIs compared to those in Z$_2$ topological insulators.

**Discussion**

Our study sheds new light on the mobility of the (001) SS of Pb$_1$\textsubscript{−}SnSe. From half width of the surface CR mode in Re $\sigma_s(\omega)$, the scattering rate of the (001) SS is estimated to be $1/\tau_{SS} = 1.2 \pm 0.6$ meV, which clearly manifest itself as the very narrow dip feature around 32 meV in $R(\omega, B)$ (Supplementary Note 4). Therefore, the mobility of the SS can be estimated: $\mu_{SS} = \epsilon_{SS} \tau_{SS}^2 / (h^2) = \epsilon_{SS}/m_{\text{eff}} \sim 40,000 \text{ cm}^2 \text{V}^{-1} \text{s}^{-1}$ with an uncertainty of about 50%. Such a high surface mobility is very promising for studying the intrinsic physics of TCIs. Detailed analysis on the scattering rate and mobility from previous IR study of LL transitions in graphene support our estimation of surface mobility in TCIs since both systems feature massless Dirac fermions (Supplementary Note 4). In contrast, previous studies on TCI thin films reported surface mobility values in the range of $100$–$2000 \text{ cm}^2 \text{V}^{-1} \text{s}^{-1}$ and a broad surface IR absorption feature that is ten times wider than that in our data. The low mobility values of these TCI thin films are very likely limited by sample quality. We note that the small scattering rate and high mobility of the (001) SS also play an important role in leading to strong surface absorption in a narrow energy range in far-IR and thus a small IR penetration depth, which is responsible for the observed surface-dominated IR conductivity.

A long-standing challenge in the research of TCIs as well as $Z_2$ topological insulators is to realize surface-dominated transport. Our study elucidates the surface and bulk contributions to electronic transport. From the effective mass of bulk states $m_{\text{bulk}}$ obtained from zero-field data and the scattering rate estimated from bulk LL transitions $1/\tau_{\text{bulk}} = 10 \text{ meV}$, the mobility of our samples can be estimated: $\mu_{\text{bulk}} \sim 13,000 \text{ cm}^2 \text{V}^{-1} \text{s}^{-1}$. Similarly, high mobility has been reported by previous transport experiments. For a sample with thickness $d$, the fraction of surface contribution to the total conductance can be estimated as $n_{SS}^2 \mu_{SS} = n_{SS}^2 \mu_{\text{SS}} + n_{\text{bulk}} \mu_{\text{bulk}}$. The mobility and carrier density values obtained in our study suggest that the SS in our samples will contribute to more than 50% of the total conductance in crystals with $d < 0.4 \mu$m. Surface-dominated transport can be achieved in even thicker crystals if the bulk carrier density is reduced further in future.

In conclusion, our study has revealed several IR signatures of SS in Pb$_1$\textsubscript{−}SnSe, including their contribution to the Drude absorption in zero field and their CR mode in magnetic field, whose SWs are further found to be consistent. Moreover, the frequency of the surface CR mode $\omega_{\text{SS}}$ inferred from our data is in accord with those estimated from theoretical studies and STM and ARPES measurements. Finally, we show that the resonance at $\omega_{\text{SS}}$ in field can only be assigned to the SS, because it is well below the energy range of all bulk LL transitions. Our study demonstrates the viability of surface-dominated IR conductivity and transport, as well as an ultrahigh surface mobility of $\sim 40,000 \text{ cm}^2 \text{V}^{-1} \text{s}^{-1}$ in Pb$_1$\textsubscript{−}SnSe with (001) surface. We expect that these results can be found in a broad class of IV–VI semiconductor TCIs Pb$_1$\textsubscript{−}SnSe(Te) because of the similar properties of their Se$_{1-x}$. These findings establish Pb$_1$\textsubscript{−}SnSe(Te) thin crystals (potentially with electrostatic gating) as a fertile system for accessing the optical and transport properties of the (001) SS, which is a crucial step for exploring many novel topological phenomena in TCIs and their potential electronic, spintronic, and valleytronic applications.

**Methods**

**IR measurements and data analysis.** Pb$_1$\textsubscript{−}SnSe ($x = 0.23\text{–}0.25$) single crystals with centimeter size were grown by the Bridgman growth method. As Pb$_1$\textsubscript{−}SnSe crystals have cubic symmetry, the samples in our study have been chosen from naturally cleaved cubic crystals with easy low indexing (00L) cleavage planes. The carrier density in the crystals is batch (growth)-dependent even for the same nominal Pb/Sn ratio because of the self-doping effect of Se vacancy defects (see Supplementary Note 1). The crystals used in our IR study are from a batch with low-carrier (defect) density. The samples were cleaved in ambient conditions before the first IR measurement, and their exposure time in air was minimized between subsequent measurements. The zero-field reflectance spectrum $R(\omega)$ was measured in the energy range of 7.5–900 meV at a temperature of $T = 8$ K. The magneto-reflectance ratio spectra $R(\omega, B)/R(\omega, B = 0)$ were measured at $T = 4.5$ K in a superconducting magnet in the Faraday geometry (magnetic field perpendicular to the sample surface). The $R(\omega, B)$ spectra were obtained by multiplying the magneto-reflectance ratio spectra by $R(\omega)$ at zero field (Supplementary Note 1). We estimate the experimental uncertainty of the absolute values of $R(\omega, B)$ is about 1–1.5%, but the relative change (evolution) of $R(\omega, B)$ with magnetic field has a smaller uncertainty owing to the better accuracy of the magneto-reflectance ratio data.

As an approximation for exploring spatially averaged properties of the SS, our data analysis employs a model with uniform carrier density $n_{\text{SS}}$ for the SS without a sharp surface/bulk interface, which will be elaborated below. In such a model, the IR data can be analyzed using formulas for bulk materials and the resulting optical conductivity contains contributions from both the SS and bulk, with all quantities of the SS being spatially averaged along the direction perpendicular to the surface within the IR penetration depth. The complex optical conductivity $\sigma(\omega) = \sigma(\omega) + i\sigma_{\text{SS}}(\omega)$ in zero field at $8$ K was obtained from analysis of $R(\omega)$ using Drude-Lorentz model combined with Kramers-Kronig (KK) constrained variational dielectric functions. Alternatively, $\sigma(\omega)$ in zero field can be obtained from KK transformation of $R(\omega)$ data, in which Hagen–Rubens formula was employed in the low-energy region and the $R(\omega)$ data at $8$ K was extended to $5.765 \text{ eV}$ using room temperature reflectance data. The two approaches above yield consistent $\sigma(\omega)$ spectra with identical spectral features. In magnetic field, the $R(\omega, B)$ spectra were analyzed using magneto-Drude-Lorentz model, in which the optical conductivity for right and left circularly polarized light (denoted + and −, respectively) is given as a function of frequency $\omega$ and magnetic field $B$. The resulting $\sigma(\omega, B)$ spectra were used to determine the scattering rates of the SS and bulk carriers, as well as the surface conductivity of Pb$_1$\textsubscript{−}SnSe.

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always yield quite similar Re

which the transition LL

with distinct surface and bulk layers45, 47. NATURE COMMUNICATIONS

values from

Pb1

can be analyzed using the methods detailed above. Regarding SS penetration depth,

simulate the

spectral features in

0.23

λ

SS is expected to be ~12.5 nm. Moreover, theoretical calculations12 have shown

similar energies, which allows us to determine Re

σ

from the corresponding author upon reasonable request.

Data availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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Author contributions

L.F. and Z.L. conceived the research and supervised the project together with F.C.; R.S. and F.C. contributed to the single-crystal growth; Z.L. and N.-L.W. supervised the experiments; Y.W. and Z.L. analyzed the data, interpreted the experimental results, and wrote the paper; N.-L.W., L.F., G.L., and J.L. contributed to the data analysis. All authors discussed the results and commented on the paper.

Additional information

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