Tunable Dirac Fermion Dynamics in Topological Insulators

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Three-dimensional topological insulators are characterized by insulating bulk state and metallic surface state involving relativistic Dirac fermions which are responsible for exotic quantum phenomena and potential applications in spintronics and quantum computations. It is essential to understand how the Dirac fermions interact with other electrons, phonons and disorders. Here we report super-high resolution angle-resolved photoemission studies on the Dirac fermion dynamics in the prototypical Bi2(Te,Se)3 topological insulators. We have directly revealed signatures of the electron-phonon coupling and found that the electron-disorder interaction dominates the scattering process. The Dirac fermion dynamics in Bi2(Te3–xSex) topological insulators can be tuned by varying the composition, x, or by controlling the charge carriers. Our findings provide crucial information in understanding and engineering the electron dynamics of the Dirac fermions for fundamental studies and potential applications.

Topological insulators represent a new state of matter that has an insulating bulk state but a conducting surface/edge state which hosts an unique spin texture and a linear dispersion relation compatible with massless Dirac fermions1–3. In addition to their potential applications in spintronics and quantum computing4,5, the topological insulators also provide a promising platform for realizing a number of novel and exotic quantum phenomena including quantum spin Hall effect6, fractional charge and quantized current7, dynamical axion field8, magnetic monopole9, Majorana fermions10,11 and quantum anomalous Hall effect12. Some of the phenomena have been observed experimentally in two-dimensional topological insulators, such as quantum spin Hall effect13 and quantum Hall effect14 in HgTe quantum wells. However, in the case of three-dimensional topological insulators like Bi2(Te, Se)3 family15–18, in spite of intensive efforts from transport experiments19–23, attempts to observe electronic transport signatures of the surface Dirac fermions have been hampered by a couple of obstacles. First, contrary to usual expectation that the bulk is insulating in topological insulators, the presence of defects in Bi2(Te, Se)3 system makes the bulk conducting24 and the transport properties are then dominated by the bulk state instead of the desired surface state20. Second, in several cases where the surface transport signatures are identified, more than one frequency or one type of charge carriers contribute to the quantum oscillation21–23, indicating the complexity of sample surface exposed to atmosphere or protection layers25. Third, the transport mobility of charge carriers in the Bi2(Te, Se)3 topological insulators, \( \mu \approx 10^4 \text{ cm}^2\text{V}^{-1}\text{s}^{-1} \) (Refs. 19,23,26) is not sufficiently high compared to those from HgTe quantum wells \((1.5 \times 10^6 \text{ cm}^2\text{V}^{-1}\text{s}^{-1})^{13,14} \) and graphene \((2.3 \times 10^3 \text{ cm}^2\text{V}^{-1}\text{s}^{-1})^{27,28} \). Although strict electron backscattering is forbidden in topological insulators due to time reversal symmetry invariance, it is important to uncover other scattering channels that limit the transport mobility of the two-dimensional Dirac fermions29.

The charge transport properties are dictated by the electron dynamics in materials. In particular, the scattering of electrons by other entities such as other electrons, phonons and disorder, determines the lifetime of the quasiparticles, and thus the electron mobility and mean free path in the transport process30. Angle-resolved photoemission spectroscopy (ARPES) has become a powerful tool to directly probe such many-body effects31,32. Under the sudden approximation, ARPES measures a single particle spectral function

\[
A(k,\omega) = \frac{1}{\pi} \frac{|\text{Im}\Sigma(k,\omega)|}{|\omega - \epsilon_k - \text{Re}\Sigma(k,\omega)|^2 + |\text{Im}\Sigma(k,\omega)|^2} \quad (1)
\]
with \( k \) and \( \omega \) representing electron momentum and energy, respectively. The real and imaginary parts of electron self-energy, \( R\Sigma(k, \omega) \) and \( Im\Sigma(k, \omega) \), carry all the information about the band renormalization and quasiparticle lifetime in an interacting system\(^{31,32} \).

There have been a few investigations on the dynamics of Dirac fermions in Bi\(_2\)(Te, Se)\(_3\) topological insulators using ARPES\(^{33,34} \) or pairs of silicon surface scattering probes\(^3\). However, the results obtained so far do not consist with each other. While the electron-phonon coupling strength (\( \lambda \)) was reported to be 0.25 in Bi\(_2\)Se\(_3\) from temperature-dependent photoemission measurements\(^{35} \), it was claimed that the electron-phonon coupling is exceptionally weak in Bi\(_2\)Se\(_3\) in another measurement\(^{36} \). The inconsistency comes presumably from two issues related to the ARPES study on many-body effects in topological insulators. First, the coexistence of bulk bands and surface bands near the Fermi level, due to the bulk doping by defects, complicates the identification of weak band renormalization effects in the surface bands. Second, the small momentum space and energy window of the Dirac fermions and the weak many-body effects require high precision ARPES measurements. The momentum space occupied by the surface state, \( \sim \pm 0.1 \) \( \AA^{-1} \) for Bi\(_2\)Se\(_3\) (Fig. 1a) and \( \sim \pm 0.04 \) \( \AA^{-1} \) for p-type Bi\(_2\)Te\(_3\) (Fig. 2a), is rather small\(^{17,18,25} \). To overcome these problems, it is ideal to first suppress the interference of the bulk bands in the measurements. Since the as-grown Bi\(_2\)Se\(_3\) is electron-doped (n-type) while Bi\(_2\)Te\(_3\) can be hole-doped (p-type), balancing the proportion of selenium and tellurium in Bi\(_2\)(Se,Te)\(_3\) system may neutralize the bulk carrier doping and reduce the effect of the bulk bands on the surface state. Furthermore, vacuum ultraviolet (VUV) laser-based ARPES\(^{37}\) can help suppress the bulk bands\(^2\), in addition to its super-high-energy and momentum resolutions (Supplementary Fig. S2) that are desirable in studying many-body effects in topological insulators.

**Results**

**Signatures of electron-phonon coupling in Bi\(_2\)Se\(_3\).** The Bi\(_2\)Se\(_3\) sample shows sharp surface state band (Fig. 1a) that facilitates the investigation of the Dirac fermion dynamics in the material. The momentum distribution curves (MDCs) at different binding energies (Fig. 1b) show well-defined sharp peaks that can be fitted by Lorentzian lineshape to obtain peak position and peak width (Full-Width-at-Half-Maximum, \( \Gamma_{\text{MDC}} \)) (Supplementary Fig. S3). The obtained MDC peak position as a function of the binding energy gives the measured dispersion relation (left panels of Fig. 1c and 1d). We assume a bare band as a linear line that connects the two points on the measured dispersion, one at the Fermi level and the other at 100 meV binding energy. This is reasonable within such a small energy window and particularly for the Dirac fermions that shows a linear dispersion over a large energy range. The effective real part of the electron self-energy (Re\(\Sigma\)) is obtained by subtracting the measured dispersion with the bare band, as shown in right panels of Fig. 1c and Fig. 1d (black empty circles). The imaginary part of the electron self-energy (Im\(\Sigma\)) is obtained from the fitted MDC width: \( \text{Im}\Sigma = \Gamma_{\text{MDC}} \times v_0/2 \) with \( v_0 \) being the Fermi velocity of the bare band (right panels of Fig. 1c and Fig. 1d, blue empty squares)\(^{32} \).

The extraction of the electron self-energy provides full information for understanding the electron dynamics in a material where the electron scattering usually involves three main processes: electron-electron interaction, electron-phonon coupling, and electron-disorder interaction\(^{39,40} \). The electron-disorder interaction is characterized by the imaginary part of the electron self-energy at zero energy (Fermi level) since the contribution from the electron-electron interaction and electron-phonon coupling approaches zero at zero energy at very low temperature. The electron-phonon interaction is characterized by the band renormalization (usually a kink in dispersion, corresponding to a peak in the real part of the electron self-energy) and a drop in the imaginary part of the electron self-energy. As exemplified in Fig. 1 for Bi\(_2\)Se\(_3\), there are clear indications of electron-phonon coupling revealed, such as the kink in dispersions (left panels of Fig. 1c and Fig. 1d), peak in the real part of the electron self-energy and a drop in the imaginary part of the electron self-energy (right panels of Fig. 1c and Fig. 1d). The characteristic energy scale of the phonons involved (\( \omega_0 \)) is determined by the peak position of the real part of the electron self-energy; in the case of Bi\(_2\)Se\(_3\), it is \( \sim 18 \) meV as marked by arrows in Fig. 1c and 1d. The electron-phonon coupling strength, \( \lambda \), can be determined by the

![Figure 1](image-url)
Extremely weak electron-phonon coupling in p-type Bi$_2$Te$_3$. The electron-phonon coupling is extremely weak in the p-type Bi$_2$Te$_3$ sample, as seen in Fig. 2. This sample was prepared under similar condition as the above Bi$_2$Se$_3$ sample and it is p-type because the Fermi level intersects with the bulk valence band. Even measured with such a super-high instrumental resolution (Supplementary Fig. S2), we do not resolve any clear signature of electron-phonon coupling. The measured dispersion is basically a straight line, corresponding to a real part of electron self-energy that is nearly zero (black empty circles in right panels of Fig. 2c and Fig. 2d).

There is no clear drop in the imaginary part of the electron self-energy (blue empty squares in right panels of Fig. 2c and Fig. 2d). All these observations point to an extremely weak electron-phonon coupling in the p-type Bi$_2$Te$_3$ sample. In the mean time, the p-type Bi$_2$Te$_3$ also exhibits a rather weak electron-disorder scattering when compared with Bi$_2$Se$_3$. As seen from the right panel of Fig. 2c, the imaginary part of the electron self-energy at zero energy is only 5.6 meV along the $\Gamma$–$K$ direction. This is significantly lower than 37 meV along the same direction in Bi$_2$Se$_3$, a topological insulator (right panel of Fig. 1c).

Tunability of Dirac fermion dynamics in the Bi$_2$(Te$_{3-x}$Se$_x$) series. The dramatically different behaviors between Bi$_2$Se$_3$ (Fig. 1) and p-type Bi$_2$Te$_3$ (Fig. 2) motivated us to further investigate the composition dependence of the Dirac fermion dynamics in the Bi$_2$(Te$_{3-x}$Se$_x$) series. We observe a systematic evolution of band structure, electron-phonon coupling, and electron-disorder scattering, with the change of composition $x$ in Bi$_2$(Te$_{3-x}$Se$_x$) (Fig. 3). Note that all these samples are prepared under similar conditions. First, the location of the Dirac point ($E_D$) moves monotonically to higher binding energy with increasing $x$. For $0 \leq x \leq 0.05$, the samples are p-type because the Fermi level intersects with the bulk valence band. For $0.15 \leq x < 1.5$, the Fermi level intersects with the surface state bands only. For $1.5 \leq x \leq 3$, the samples become n-type because the Fermi level intersects with the bulk conduction band. Note that the Dirac point exhibits a big jump when $x$ changes from 0.05 to 0.15. Second, there is an overall increase of electron-disorder interaction with increasing $x$ in Bi$_2$(Te$_{3-x}$Se$_x$).

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The electron-phonon coupling strength increases from nearly zero for p-type Bi$_2$Te$_3$ to \( \lambda \approx 0.17 \) for Bi$_2$Se$_3$.

It is interesting that for the same Bi$_2$Te$_3$ nominal composition, when prepared under different conditions, samples can become either p-type or n-type that exhibit dramatic difference in their Dirac fermion dynamics. As shown in Fig. 3b, the Dirac point of the n-type Bi$_2$Te$_3$ lies nearly 310 meV below the Fermi level which differs significantly from the 85 meV location in the p-type Bi$_2$Te$_3$ (Fig. 3a1). Particularly, the n-type Bi$_2$Te$_3$ exhibits a pronounced electron-phonon coupling, as seen from the obvious peak in the real part of the electron self-energy (black circles in Fig. 3d) and imaginary part of the electron self-energy (blue squares) for n-type Bi$_2$Te$_3$.

Figure 3 | Evolution of surface state band structure and electron self-energy with the composition, \( x \), in Bi$_2$(Te$_{3-x}$Se$_x$) system. (a) The surface state band structure of Bi$_2$(Te$_{3-x}$Se$_x$) with various compositions, \( x \), measured along the \( \Gamma-K \) direction. (b) The surface state band structure of n-type Bi$_2$Te$_3$ along the \( \Gamma-K \) direction. (c) Corresponding effective real part of the electron self-energy (black circles) and imaginary part of the electron self-energy (blue squares) for Bi$_2$(Te$_{3-x}$Se$_x$) with different compositions. (d) Corresponding effective real part of the electron self-energy (black circles) and imaginary part of the electron self-energy (blue squares) for n-type Bi$_2$Te$_3$.

The electron-disorder scattering also increases in the n-type Bi$_2$Te$_3$ (Fig. 3c1). The first principle calculation, which expects a rather weak electron-phonon coupling in n-type Bi$_2$Te$_3$ (\( \lambda \approx 0.05 \)) is hard to explain the strong electron-phonon coupling in n-type Bi$_2$Te$_3$ and particularly the disparity of electron-phonon coupling between the n-type and p-type Bi$_2$Te$_3$.

The different electron dynamics between Bi$_2$Se$_3$ and Bi$_2$Te$_3$, and between n-type Bi$_2$Te$_3$ and p-type Bi$_2$Te$_3$, is intriguing. We note that the native majority defect types are totally different in these samples, i.e., Se vacancies in Bi$_2$Se$_3$, Te-Bi antisites in the n-type Bi$_2$Te$_3$, and Te vacancies in the p-type Bi$_2$Te$_3$. The difference of the electron-disorder interaction can be due to such different types of disorders and their densities in the samples. In addition, as the main difference between the n-type and p-type Bi$_2$Te$_3$ lies mainly in the relative position between the Fermi level and the bulk bands, it is reasonable to deduce that the bulk bands may play a role in the electron dynamics of the Dirac fermions. This is consistent with the observation that, for n-type Bi$_2$Te$_3$ (\( x = 0 \)), although its composition is rather different from Bi$_2$(Te$_{2.85}$Se$_{0.15}$) (\( x = 0.15 \)), their Dirac fermion dynamics is quite similar in both the electron-phonon coupling and electron-disorder interaction (Figs. 3 and 4). The distinctness of the electron-phonon coupling between the n-type and p-type Bi$_2$Te$_3$ indicates that the control of the charge carrier doping can be an alternative way to tune the Dirac fermion dynamics in topological insulators.

One may wonder whether the origin of the observed inelastic scattering in Bi$_2$(Se$_x$Te$_{3-x}$)-based topological insulators is related to the materials themselves (defect, atomic arrangement and etc.) or the intrinsic topological properties associated with the time reversal symmetry (TRS) invariance. The TRS-protection may reduce the scattering strength because strict backscattering is not allowed, but it is difficult to quantitatively analyze this weakening effect. Note that the entire Bi$_2$(Te$_{3-x}$Se$_x$) series are topological insulators. Therefore, we believe the systematic evolution of their electron dynamics is mainly a manifestation of the usual many-body effects, other than a manifestation of the intrinsic TRS-related topological properties.

Fig. 4 gives a quantitative summary of the electron-phonon coupling and electron-disorder scattering in the Bi$_2$(Te$_{3-x}$Se$_x$) system. The relative position between the Dirac point and the bulk band is depicted in Fig. 4a for a couple of typical compositions and both p- and n-type Bi$_2$Te$_3$. With increasing x in Bi$_2$(Te$_{3-x}$Se$_x$), there is an abrupt jump in the electron-phonon coupling strength from \( x = 0 \) to \( x = 0.15 \), accompanied by a transition from p-type to n-type. Further increase of x leads to an overall slight increase of electron-phonon coupling (Fig. 4c). Meanwhile, the characteristic energy scale, \( \omega_0 \),
also increases with $x$, which is consistent with the increase of the phonon frequency from Bi$_2$Te$_3$ to Bi$_2$Se$_3$ (Supplementary Fig. S7). This indicates that the (Te$_x$Se$_{1-x}$) vibrations play a major role in dictating the electron-phonon coupling strength in the Bi$_2$(Te$_{3-x}$Se$_x$) system. This is also consistent with the observation that the characteristic energy scale is identical in n-type and p-type Bi$_2$Te$_3$ because their phonon frequencies are quite similar (Supplementary Fig. S7) although they have different electron-phonon coupling strength. It is clear that electron-disorder scattering is dominant in the scattering process. As seen from Fig. 4b, the imaginary part of the electron self-energy at zero energy $\text{Im} \Sigma(E_F)$, which gives a good measure of the electron scattering rate, shows an overall increase from the p-type Bi$_2$Te$_3$ for $x=0$ to Bi$_2$Se$_3$ for $x=3$ in the Bi$_2$(Te$_{3-x}$Se$_x$) system (red circles in Fig. 4b). The quasi-particle mean free path, $\lambda'$, which can be determined from the MDC width $T_{\text{MDC}} = 1/T_{\text{MDC}}'$, first decreases abruptly from the p-type Bi$_2$Te$_3$ ($x=0$) to $x=0.15$, followed by an overall slight decrease with $x$ in Bi$_2$(Te$_{3-x}$Se$_x$) (blue circles in Fig. 3b). The electron mobility of the Dirac fermions ($\mu'$) is directly related to the quasi-particle mean free path, $\mu' = e\lambda'/(nk_F^2)$, with $k_F$ being the Fermi momentum. It is then estimated that the electron mobility in p-type Bi$_2$Te$_3$ is near 1000 cm$^2$V$^{-1}$s$^{-1}$ (Supplementary Fig. S4). By comparison, the electron mobility in Bi$_2$Se$_3$ and n-type Bi$_2$Te$_3$ is $\sim 70$ and $\sim 100$ cm$^2$V$^{-1}$s$^{-1}$, respectively, which are significantly lower than that of p-type Bi$_2$Te$_3$. We note that, although the electron mobility obtained from ARPES is not strictly the same as the electron mobility in the transport properties, they have good correspondence, especially for materials with circular Fermi surface and nearly isotropic electron scattering.

**Discussion**

The present study will provide important information for engineering the Dirac fermion dynamics in Bi$_2$(Te$_{3-x}$Se$_x$) topological insulators. It clearly indicates the presence of electron-phonon coupling and dominant role of the electron-disorder scattering in dictating the Dirac fermion dynamics in the system. The Dirac fermion dynamics can be tuned by varying either the composition or the charge carrier concentration. Specifically for the Bi$_2$(Te$_{3-x}$Se$_x$) system, our results clearly show that Bi$_2$Te$_3$ system is superior over Bi$_2$Se$_3$ in achieving higher electron mobility both due to its weak electron-phonon coupling and its weak electron-defect scattering. To further enhance the electron mobility in searching for novel quantum phenomena, it is necessary to choose a system with weak electron-phonon coupling and reduced disorder scattering.

**Methods**

High quality single crystals of Bi$_2$(Te$_{3-x}$Se$_x$) (0 $\leq x \leq 3$) were grown by the self-flux method. The mixed materials were heated to 1000°C, held for 12 hours and then slowly cooled down to 500°C over 100 hours before cooling to room temperature. Single crystals of nearly one centimeter in size were obtained by cleaving. The angle-resolved photoemission measurements were carried out on our vacuum ultra-violet (VUV) laser-based angle-resolved photoemission system, with an overall energy resolution of $\sim 1$ meV. The momentum resolution is $\sim 0.004$ Å$^{-1}$ for 30° angular mode and $\sim 0.002$ Å$^{-1}$ for 14° angular mode of the R4000 electron energy analyzer. The samples were all cleaved and measured in vacuum with a base pressure better.
than $5 \times 10^{-3}$ Torr and a temperature $T \sim 20$ K. For more details about the experimental methods, see supplementary information.

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Author contributions
C.Y.C. and X.J.Z. conceived and designed the research; C.Y.C. performed measurements; C.Y.C. and X.J.Z. analyzed data and wrote the paper. J.Z., L.Y., X.Y.W., Q.J.P., Z.M.W., S.J.Z., F.Y., C.T.C., Z.Y.X. and X.J.Z. contributed new reagents/analytic tools; C.Y.C. and X.J.Z. analyzed data and wrote the paper.

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