Mapping the unconventional orbital texture in topological crystalline insulators

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The newly discovered topological crystalline insulators feature a complex band structure involving multiple Dirac cones1–6, and are potentially highly tunable by external electric field, temperature or strain. Theoretically, it has been predicted that the various Dirac cones, which are offset in energy and momentum, might harbour vastly different orbital character7. However, their orbital texture, which is of immense importance in determining a variety of a material’s properties8–10 remains elusive. Here, we unveil the orbital texture of Pb1−xSnxSe, a prototypical topological crystalline insulator. By using Fourier-transform scanning tunnelling spectroscopy we measure the interference patterns produced by the scattering of surface-state electrons. We discover that the intensity and energy dependences of the Fourier transforms show distinct characteristics, which can be directly attributed to orbital effects. Our experiments reveal a complex band topology involving two Lifshitz transitions11 and establish the orbital nature of the Dirac bands, which could provide an alternative pathway towards future quantum applications.

A counterpart to charge and spin, electron orbitals are of great importance in the underlying physical processes of a variety of systems. The orbital degrees of freedom, for example, play a crucial role in the colossal magnetoresistance effect in manganese oxides, and contribute to the anisotropic electronic and magnetic properties in many other transition-metal oxide systems5. More recently, orbital ordering within the superconducting FeAs layer has been thought to govern structural phase transitions and ‘stripe’-like antiferromagnetism in Fe-based high-temperature superconductors8,10. Similarly, topological materials host complex orbital arrangements often strongly coupled to other electronic degrees of freedom12–14. Topological crystalline insulators (TCIs) in particular are predicted to exhibit intricate band, spin and orbital textures, potentially relevant for interactions in the quantum Hall regime. Although previous experiments provided a glimpse into the complex band topology present in TCIs (refs 3–5,19,20), these experimental efforts have not been able to shed light onto its orbital texture. Here we use Fourier-transform (FT) scanning tunnelling spectroscopy (STS) to reveal the distinct orbital nature of the Dirac bands in the TCI, Pb1−xSnxSe.

In its stoichiometric state, Pb1−xSnxSe with x = 0 is a trivial insulator under the Z2 topological classification of materials owing to the absence of band inversion. The process of adding Sn, which substitutes for Pb, leads to band inversion at an even number of time-reversal points, and the solutions remain Z2 trivial. However, for a particular region of the composition–temperature parameter space, such as x > ~0.23 and room temperature, or x > ~0.18 and 4 K, topologically protected surface states emerge owing to the non-trivial band topology classified by crystalline symmetries15. Pb1−xSnxSe crystals cleave along the (001) crystal direction based on the square lattice observed in scanning tunnelling microscope (STM) topographs (Fig. 1a,b) and 3 Å step heights (Supplementary Section II). We therefore consider the band structure at the (001) face. From theory, the SS band structure consists of two ‘parent’ Dirac cones centred at X and vertically offset in energy2–21. When they intersect, the hybridization between the electron branch of the lower parent Dirac cone and the hole branch of the upper parent Dirac cone opens a gap at all points except along the mirror line, leading to the formation of a pair of ‘child’ Dirac points shifted away from the Dirac point and the overall spectra remain homogeneous to X, one large hole pocket and a smaller electron pocket, both centred at X (Fig. 1c,d; refs 2,19). In its stoichiometric state, Pb1−xSnxSe with x = 0 is a trivial insulator under the Z2 topological classification of materials owing to the absence of band inversion. The process of adding Sn, which substitutes for Pb, leads to band inversion at an even number of time-reversal points, and the solutions remain Z2 trivial. However, for a particular region of the composition–temperature parameter space, such as x > ~0.23 and room temperature, or x > ~0.18 and 4 K, topologically protected surface states emerge owing to the non-trivial band topology classified by crystalline symmetries15. Pb1−xSnxSe crystals cleave along the (001) crystal direction based on the square lattice observed in scanning tunnelling microscope (STM) topographs (Fig. 1a,b) and 3 Å step heights (Supplementary Section II). We therefore consider the band structure at the (001) face. From theory, the SS band structure consists of two ‘parent’ Dirac cones centred at X and vertically offset in energy2–21. When they intersect, the hybridization between the electron branch of the lower parent Dirac cone and the hole branch of the upper parent Dirac cone opens a gap at all points except along the mirror line, leading to the formation of a pair of ‘child’ Dirac points shifted away from the Dirac point and the overall spectra remain homogeneous to X, one large hole pocket and a smaller electron pocket, both centred at X (Fig. 1c,d; refs 2,19).

Figure 1e shows position-dependent dI/dV spectra for this material. In general, the dI/dV spectra are V-shaped, with two well-defined peaks (Fig. 1e). From our previous studies, the minimum in the density of states of the V-shaped feature denotes the energy of the Dirac point, and the two peaks on either side represent Van Hove singularities (EVHS+/EVHS−) associated with the Lifshitz transition22. Interestingly, despite the disordered nature of our samples, with greater than 30% of the Pb sites replaced by Sn, we find that the Dirac point and the overall spectra remain homogeneous to within a few meV (Fig. 1e and also see Supplementary Section III). Having identified the important energy scales, we proceed to the application of the Fourier-transform quasiparticle interference (FT-QPI) of this system.

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FT-QPI imaging has been successfully applied to extract the band structure of many complex systems, such as high-temperature superconductors, heavy-fermion compounds, and Z₂ topological materials, but this technique has yet to be used fully for spin- and orbital-texture mapping of extracted bands. Representative FTs of dI/dV conductance maps acquired over a 1300 Å square region of Pb₁₋ₓSnₓSe are shown in Fig. 2c.f. To analyse the FTs, as a starting point, the expected QPI pattern can be obtained using a simple autocorrelation of the CECs, not including any matrix element effects (Fig. 2b,e). The pattern produced can be understood by studying the Fermi surface topology shown in Fig. 2a.d. From this, we see that two main sets of scattering wavevectors (Q₁/Q₂ and Q₃/Q₄) are expected for the (001) SS. Above the Lifshitz transition, for example (Fig. 2d,e), the QPI pattern around the momentum positions Q₁/Q₂ originates from scattering between two (110) mirror-symmetric pairs of Dirac cones, and is expected to be quasi-elliptical. In contrast, the pattern at Q₃/Q₄ marks the scattering between two pairs of parent Dirac cones rotated 90 degrees with respect to one other, and should be nearly circular (Supplementary Section IV).

By studying the experimental FTs, our first observation is that there is a marked difference between the QPI patterns above E_{VHS+} (Fig. 3a), where clear, nearly circular dispersing features appear, at E_{VHS}, (Fig. 3b), and below E_{VHS+}, where a (non-dispersing) ‘clover’-like set of four dots appear (Fig. 3c). This remarkable switch in the QPI pattern is directly connected to the change in Fermi surface topology across the Lifshitz transition (Fig. 3a–f). The simple autocorrelation of the CECs shown in Fig. 2b,e confirms this picture; the disconnected CECs below E_{VHS}, result in a non-dispersing clover pattern near Q₃/Q₄, whereas the continuous oval-shaped CECs above E_{VHS}, result in a quasi-circular dispersing pattern.

The main challenge of any QPI study is extracting the band structure in k-space from the observed dispersive modes in q-space. An important piece of information is provided in the set of scattering wavevectors Q close to the Dirac point where the clover is observed (Figs 2a–c and 3c). As the clover is a product of quasiparticle scattering between four child Dirac cones in the first Brillouin zone, the average distance in q-space between the centres of neighbouring clover leaves allows us to precisely extract the k-space distance between two Dirac cones symmetric around X to be 0.060 ± 0.006 Å⁻¹. Interestingly, as the k-space location of the Dirac cones in TCI is directly connected to doping, and nominal doping is often quite different from the actual and can
also vary within the sample, this method presents an alternative way to locally determine and compare the doping level in this class of materials.

To extract SS dispersion anisotropy in \( k \)-space, we need to use the \( q \)-space dispersions of the sets of wavevectors at both \( Q_1 \) and \( Q_2 \). Remarkably, although \( Q_3 \) is expected to be weak, the high
levels in Pb1.

the arithmetic mean of dispersion velocities

spectrum obtained on the same sample (Fig. 3h). Furthermore, with the energy of the corresponding feature in the average d

I

d − 93 meV

d + 97 meV

d + 7 meV

Ed + 97 meV

Ed − 93 meV

Ed + 7 meV

Ed + 122 meV

Ed + 2 meV

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expected to have different orbital character and are associated with orbitals with opposite signs of mirror eigenvalues. Let us now consider the two sets of parent/child Dirac cones in $k$-space, one around $X (\pi/a_{x}, 0)$ and the other around $Y (0, \pi/a_{y})$ (Fig. 1c). Theoretically, whereas the upper branch of the lower Sn parent Dirac cone is associated with $p_{x}$ orbital character for both $X$- and $Y$-momenta, the lower branch of the upper Se parent Dirac cone has a different orbital wavefunction—that is, Se $p_{y}$ around $X$ and Se $p_{x}$ around $Y$ (Fig. 1c). The different orbital textures below $E_{d}$ at $(\pi/a_{x}, 0)$ and $(0, \pi/a_{y})$ positions should suppress scattering between them and therefore result in diminished QPI patterns below $E_{d}$, exactly as seen in Fig. 4. Our data thus provide direct experimental confirmation of the proposed orbital alignment of the TCI SS.

To further substantiate this picture and elucidate the origin of the specific orbital-selective scattering processes responsible for the subtle features in the QPI patterns, we use the proposed spin and orbital textures in model simulations of the QPI data. We find that the resulting simulations capture many features of our data, including changes in intensity with angle in the set of scattering wavevectors $Q_{i}$ (Supplementary Section VII) that were clearly lacking in the simple autocorrelations. Take the QPI pattern at $E_{d} + 122$ meV as an example (Fig. 4d–f). The outermost high-intensity ring is due to scattering between the two Sn $p_{z}$ parent cones across $X$- and $Y$-neighbourhood. The inner ring comes from the scattering between the Se parent cone at one Brillouin zone corner neighbourhood and the Sn cone at the other. Without the spin- and orbital-matrix element effect (Fig. 4e), little intensity variation around the rings was found in the autocorrelation map. When the matrix element effect is turned on (Fig. 4f), the inner ring is greatly suppressed because it represents scattering between different kinds of orbitals, once again consistent with the postulated orbital texture. The intensity variation on the outer ring is in part due to the spin texture on the oval-shaped CEC and partly a consequence of the chosen impurity potential for matching the experimental data (Supplementary Section VII). We note that orbital-selective ARPES experiments might be able to provide further information regarding this orbital arrangement, especially within the narrow energy range between the two Lifshitz transitions ($E_{\text{VIS}} < E < E_{\text{VIS}}$) where a weaker QPI signature is observed.

Finally, our data show evidence for asymmetry in the dispersion velocities between the hole and electron branches of the outer Dirac cones. The dashed grey lines in Fig. 2g represent linear fits to the experimental data points and show a slight asymmetry in the slope above and below $E_{d}$. This asymmetry is also noticeable in the Landau level dispersion in the same material and the positions of the upper and lower Lifshitz transitions with respect to the Dirac point (Supplementary Section VI). This phenomenon could be attributed to the fact that bulk conduction and valence bands are not necessarily particle–hole symmetric. Our observations provide strong evidence that, in addition to asymmetric nature of the orbital wavefunction, a particle–hole symmetric model with identical parent Dirac cones dispersions may not be enough to completely encompass the underlying physics present in this class of materials and that non-identical parent Dirac cones need to be taken into account.

Methods

The $\text{Pb}_{1-x}\text{Sn}_{x}\text{Se}$ single crystals used for QPI imaging were grown by the self-selecting vapour growth method, cleaved at 77 K, and immediately inserted into the STM head. Doping concentration was checked using energy-dispersive X-ray spectroscopy (EDS) and electron microprobe analysis (see Supplementary Section I for more details). All $dI/dV$ measurements were acquired at 6 K using a standard lock-in technique with $\sim 5$ meV peak-to-peak modulation at a frequency of 1,488 Hz. We use the Lawler–Pujita drift-correction algorithm on all acquired data to remove the effects of slow thermal and piezoelectric drift. The quasiparticle interference (QPI) imaging technique uses the interference of elastically scattered quasiparticles (which are just electrons in this case) with different momenta $k_{1}$ and $k_{2}$, resulting in a standing wave ‘ripples’ of wavevector $q = k_{1} - k_{2}$ which can be detected in STM $dI/dV$ conductance maps.

Two-dimensional fits of $dI/dV$ maps (FF-QPI; ref. 23) were used to extract all the scattering wavevectors.

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Author contributions

Y.O., I.Z. and V.M. designed the experiments. Samples were obtained from R.S., F.C. and M.Z.H. Theoretical analysis and calculations were done by C-Y.H., M.S., W-F.T., H.L., A.B. and L.F. STM experiments were carried out by Y.O., I.Z., D.W. and W.Z., I.Z., Y.O., H.L., L.F. and V.M. analysed the data and wrote the paper.

Additional information

Supplementary information is available in the online version of the paper. Reprints and permissions information is available online at www.nature.com/reprints. Correspondence and requests for materials should be addressed to V.M.

Competing financial interests

The authors declare no competing financial interests.