

Supplementary Information for

A Topological Superconductor Tuned by Electronic Correlations

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Supplementary Notes

1. Determining the Te content

According to Vegard's law, the Te content (x) in $\text{FeTe}_x\text{Se}_{1-x}$ (FTS) thin films grown on SrTiO_3 (STO) substrates should scale roughly linearly with the c -axis lattice constant. However, for films with very dilute Se doping ($T_{\text{Se}} < 115^\circ\text{C}$), the lattice constants tend to saturate and do not scale linearly with x . Here, for films grown with Se temperatures greater than 115°C , we compare our XRD measurements with those in the literature¹. We extract a polynomial relationship between the c -axis lattice constant and the Te content from Ref.¹, and use this relationship with a modified overall scaling according to the end values at $x = 1$ and $x = 0$ to determine the Te contents of our samples. For films with Se evaporation temperature lower than 115°C , since the c -axis lattice constant saturates to the value for FeTe, we estimate their Te contents using the theoretical fluxes for Se and Te:

$$P = 10^{A - \frac{B}{T}},$$
$$F \propto \frac{P}{\sqrt{mT}},$$

where P is the vapor pressure in mbar at source temperature T in units of K; F is the theoretical flux; and m is the molecular mass. $A_{\text{Se}} = 9.325$, $A_{\text{Te}} = 10.014$, $B_{\text{Se}} = 5779$, $B_{\text{Te}} = 7692$ are constants calibrated by Dr. Eberl MBE-Komponenten GmbH - the manufacturer of our effusion cells².

To further verify the Te:Se ratios in our FTS thin films, we performed additional inductively coupled plasma mass spectrometry (ICP-MS) measurements (fig. S2) for various samples. We found that the results from ICP-MS in general agree with our polynomial fitting results. Notably, in the nominal 10 UC FeTe sample, we find the Te content to be $x = 0.98$, indicating that any incidental Se doping remains very small. We emphasize that even for low Se temperatures (*e.g.*, 110°C), a few percent doping of Se makes the electronic structure substantially different, as can be seen in Fig. 2 and 3 in the main text.

2. Photoemission matrix elements

ARPES measures the product of the one-particle matrix element $|M_{f,i}^k|$, the Fermi-Dirac distribution $f(\omega)$, and the spectral function $A(k, \omega)$ ³. Assuming a free-electron final state, the matrix element can be calculated as:

$$|M_{f,i}^k| \propto |\langle e^{ikr} | \epsilon \cdot \mathbf{x} | \Psi_i^k \rangle|^2,$$

where ϵ is a unit vector along the polarization of the incident photons. The calculated matrix element magnitude along the $\bar{\Gamma} - \bar{X}$ direction for all Fe t_{2g} orbitals and chalcogen p_z orbital is presented in fig. S3. For a p -polarized incident beam, the d_{yz} and d_{xz} bands are visible throughout the $\bar{\Gamma} - \bar{X}$ momentum path, with the d_{yz} band being more prominent. The d_{xy} and p_z band show suppressed intensities near $\bar{\Gamma}$ but become more noticeable away from $\bar{\Gamma}$. In contrast, the s -polarization enhances the d_{xz} band while suppressing the d_{yz} band. The d_{xy} and p_z bands are also heavily suppressed over a broad momentum range. The results for matrix element calculations are summarized in table. S1. These calculated features are consistent with the experimentally observed spectra shown in Fig. 2 of the main text.

Fig. S7 shows spectra taken with an s -polarized 6 eV laser. At $x = 0.79$, a crossing feature is resolved near -10 meV. A linearly dispersive band is seen at $\bar{\Gamma}$ in fig. S7b(ii), corresponding to the topological surface state (TSS). This observation demonstrates that 10 UC $\text{FeTe}_{0.79}\text{Se}_{0.21}$ is in the

topologically nontrivial phase. In addition, the d_{xz} band becomes increasingly invisible as x approaches 1, reflecting the inverted character of xy^* on the d_{xz} band. The xy^* band has a mixture of the p_z and d_{xy} characters, neither of which can lead to detectable intensities using the s -polarized light.

3. Non-trivial FTS in the superconducting state probed by the p -polarized photons

We perform ARPES measurements on 10 UC $\text{FeTe}_{0.93}\text{Se}_{0.07}$ using p -polarized light below and above the superconducting transition temperature. This polarization allows us to observe the TSS as well as the d_{xy} and d_{yz} orbitals (fig. S5). Fig. S5b shows that both the upper TSS and the hybridized d_{xy}/d_{yz} band connect with a bulk band continuum in the normal state, as predicted by established theory⁴. However, when lowering the temperature below the superconducting T_c (fig. S5a), we observe a quasi-flat dispersion contributed by the bulk d_{xy}/d_{yz} band. Since the smallest superconducting gap observed on the quasi-flat dispersion is still a factor of two larger than that on the TSS (main text Fig. 1e), we conclude that the spectral weight of the TSS near E_F is completely dominated by that of the bulk d_{xy}/d_{yz} band using the p -polarized light. This result is consistent with the picture reported by⁵ on bulk FTS crystals.

4. Superconducting gap fitting

We extract the superconducting gap magnitudes shown in Fig. 1 of the main text by fitting the symmetrized energy distribution curves (EDCs) to the empirical model proposed by Norman⁶. Although this model can yield a finite gap at temperatures above 14 K, the raw symmetrized EDCs do not show the characteristic two-peak structure expected for a gapped state. To verify this, we compared single- and double-Lorentzian fits of the symmetrized EDCs. These data were taken from the spectra using s -polarized light. As illustrated in fig. S6, at temperatures below 14 K, the residuals from double-Lorentzian fits are clearly lower than those from single-Lorentzian fits, consistent with a gapped spectral shape. In contrast, at 14 K, the double-Lorentzian approach offers no obvious improvement, indicating the absence of a discernible gap. Consequently, we identify 14 K as the superconducting gap closing temperature in the main text.

5. Non-trivial topology in thin film FTS

To theoretically establish the non-trivial topology in thin film FTS, we perform DFT and tight-binding calculations. The spinful system with time reversal symmetry ($\mathcal{T}^2 = -1$) belongs to the AII symmetry class and the corresponding topological invariant in 3D is $\mathbb{Z}_2$ ⁷. The topological invariant can be obtained from the product of the parity eigenvalues of occupied Kramers pairs at the time-reversal-invariant momenta⁸. Without the inversion symmetry, as in the case of our thin film FTS, the topological invariant can be obtained by calculating the Wilson loop⁹, where "partner switching" of the Wannier function center during time-reversal pumping implies a topologically nontrivial $\mathbb{Z}_2$. The surface Dirac band in FTS emerges from the band inversion at the Z point, which induces a topological invariant $\mathbb{Z}_2$. The inversion symmetry breaking in FTS induces Rashba spin-orbit coupling but will not change the topological invariant. The DFT band structure with asymmetric Te/Se height is shown in fig. S8a and the corresponding wannier centers at $k_z = 0$ and $k_z = \pi$ planes are shown in fig. S8c. The wannier bands exhibit "partner switching" in the $k_z = 0$ plane but not in the $k_z = \pi$ plane, suggesting a nontrivial $\mathbb{Z}_2$ topology.

From the tight-binding calculation, the Γ_6^+ and Γ_6^- bands with opposite parities have a band inversion at the Z point (as shown in fig. S8b), leading to a strong topological phase according to the calculation of topological invariant. This topology will generate nontrivial surface states. The parities of the relevant states along the $M - \Gamma - Z$ line near E_F are given in fig. S8b. The bands

around X and M points do not show parity exchange along the Z direction and will not affect topology, and hence are not shown in the Figure.

6. Electron pocket near the zone center in FeTe

Previous DMFT results predicted that strong correlations in FeTe can lead to a complete band inversion between the xy^- and d_{xz} bands throughout the $\Gamma - Z$ trajectory¹⁰. We adjust our phenomenological tight-binding model to realize such a "complete band inversion" and plot the band structure at $k_z = 0$ projected to different orbitals (fig. S12). There is a small electron pocket right above the innermost hole pocket, and it is of d_{xz}/d_{yz} orbital characters. Meanwhile, the innermost hole pocket, which was termed the d_{xz} band in a band-uninverted regime, is primarily composed of xy^- orbitals according to Ref.¹⁰. This is fully consistent with our ARPES data taken with s -polarized light. The s -polarized light has a relatively large nonzero photoemission cross-section for d_{xz} orbitals, but almost negligible cross-section for p_z , d_{xy} , and d_{yz} orbitals (fig. S3). Hence, ARPES with s -polarized light will detect the small electron pocket near Γ , but neither the d_{yz} -derived hole-like dispersion connecting to the electron pocket, nor the innermost hole pocket primarily of xy^- characters (hybridized p_z and d_{xy}). We note that such a bulk-derived electron pocket can appear in the vicinity of the TSS, which makes it rather difficult to distinguish between these two spectral features. However, the complete disappearance of the valence band using s -polarized light can only occur in the complete band inversion regime. This leads to a dominant xy^- character in the valence band and a vanishing cross-section using the s -polarized photons, hence supporting our assignment of the d_{xz}/d_{yz} -driven electron pocket in 10 UC FeTe.

This small electron pocket is also seen in both DFT+eDMFT calculations and ARPES data taken with p -polarized light. Main text Fig. 3c(v) already shows that there is an electron pocket of d_{xz}/d_{yz} orbital characters (re-plotted in fig. S13c). For the ARPES data taken with p -polarized light, the momentum distribution curve (MDC) plot (fig. S13b) clearly shows an electron pocket. Therefore, DMFT, tight-binding model, and ARPES data using both p - and s -polarized light yield a consistent picture about the electron pocket observed in 10 UC FeTe.

7. Loss of quasiparticle coherence

EDCs taken from the laser-ARPES data in main text Fig. 2a are fit to the model in Eqn. 1. These EDCs are normalized by the intensities at -130 meV. A_i , E_i , and w_i represent the amplitude, central energy, and peak width of each Lorentzian peak. C stands for an incoherent background due to lattice disorder, secondary electrons, and other extrinsic effects. The sum of these three terms is multiplied by a Fermi-Dirac function with Fermi level E_F and width w_T . Note that w_T represents a combined broadening due to the finite temperature and energy resolution. y_0 is an overall background.

$$I(E) = y_0 + \left[\frac{A_1 w_1^2}{(E - E_1)^2 + w_1^2} + \frac{A_2 w_2^2}{(E - E_2)^2 + w_2^2} + C \right] \frac{1}{1 + \exp\left(\frac{E - E_F}{w_T}\right)} \quad (1)$$

This model fits the EDCs for a broad range of Te content from $x = 0.60$ to $x = 1.00$. The spectral weight of each peak is calculated by $A_i w_i$. The spectral weight of the lower Dirac cone likely has contributions from the valence band right below the TSS. This spectral weight manifests the band inversion, as the xy^- character gradually enters the valence band and consequently makes it incoherent. Eventually, the dominant xy^- character toward the $x = 1$ limit causes the lower Dirac

cone to completely lose coherence (fig. S14; main text Fig. 2d). The error bars of the spectral weights (s_i) are calculated as $\Delta s_i/s_i = \sqrt{(\Delta A_i/A_i)^2 + (\Delta w_i/w_i)^2}$.

8. Fitting the effective mass

To extract the mass enhancement factor from ARPES spectra, we obtain the 2D curvature plots using the method described in Ref.¹¹. Fig. S17a shows the 2D curvature plot near $\bar{\Gamma}$ for 10 UC FeTe_{0.6}Se_{0.4}. The dispersion for the d_{xy} band is extracted by tracking the local minima of the EDCs of the 2D curvature plot; those of the d_{xz} and d_{yz} bands are extracted by tracking the local minima of the momentum distribution curves (MDCs) (fig. S17b). These dispersions are subsequently fitted to a parabolic function:

$$E = \frac{3.82}{m^*} k^2 + ak + b,$$

where E is the energy of the band in units of eV; k is the momentum of the band in units of $\text{\AA}^{-1}$; and m^* is the effective mass of the fitted band in units of the free electron mass.

We have also extracted the Fermi momentum (k_F) and the band minimum (E_{Band}) for the d_{xy}/d_{xz} hybridized electron band near $\bar{M}$, and plot the quantity k_F^2/E_{Band} (fig. S17d) which serves as a general measure of how "flat" a band is. From both analyses, the strong renormalization of the d_{xy} band, particularly near the $x = 1$ limit, is evident.

9. Potential spectral weight transfer indicating Mott physics

As we substitute Se with Te, we observe a general reduction in the spectral weight of the Fe $3d$ bands near E_F , consistent with the picture of quasiparticle weight reduction. In 10 UC FeSe (fig. S19a) the incoherent feature at the binding energy of approximately 1-3 eV has been identified as the lower Hubbard band (LHB)¹²⁻¹⁴. Similar incoherent features also appear in both FeTe_{0.6}Se_{0.4} and FeTe, and the intensity seems to grow as the Te content increases. This observation is also consistent with theoretical calculations based on a five-orbital Hubbard model using the U(1) slave-spin method¹⁴. Nevertheless, we caution that these samples may have slight differences in the overall quality and photoemission yield, making a direct quantitative comparison of subtle changes difficult.

Although our EDC results confirm that the quasiparticle spectral weight near E_F diminishes with higher Te content, it remains difficult to pinpoint precisely where that missing spectral weight is transferred to. This is because one cannot easily cover the entire Brillouin zone for spectral weight analysis. Even if this is accomplished, different band features can have different matrix elements, making a quantitative evaluation of the spectral weight transfer very challenging. Hence, we refrain from distinguishing the orbital-selective Mott phase transition from a coherent-to-incoherent transition.

10. Quasi-particle weights extracted by eDMFT

Quasi-particle weights with respect to DFT band masses are extracted using a statistical average of the self-energy obtained directly from continuous time quantum Monte Carlo (CTQMC) calculations on the Matsubara axis. We then extrapolate the imaginary part of the Matsubara Green's function at zero frequency and take the derivative i.e. $Z^{-1} = 1 - \frac{\delta \Sigma(iw)}{\delta(iw)}|_{w=0}$. These inverse quasiparticle weights represent the mass enhancement factors with respect to the DFT-calculated masses, and are plotted in fig. S20 for FeTe as a function of the Te height. The results

for $\text{FeTe}_{0.5}\text{Se}_{0.5}$, as well as the bulk phase of FeSe and FeTe are also included in fig. S20. The mass enhancement factors we calculated for bulk FeSe and bulk FeTe are consistent with literature results such as Nature Mater. 10, 932 (2011)¹⁵.

11. Spin-orbit coupling

Effect of spin-orbit coupling (SOC) on $\text{FeTe}_{0.5}\text{Se}_{0.5}$: Figure S21a-d show DFT+eDMFT calculations without SOC for $\text{FeTe}_{0.5}\text{Se}_{0.5}$ where the spectral functions are along $\bar{X} - \bar{\Gamma} - \bar{X}$ at different $\mathbf{k}_z$'s. The d_{xy} - d_{yz} hybridization gaps are calculated by removing the broadening from the spectral functions and obtaining an energy difference between the d_{xy} and d_{yz} band tops. The same spectral functions with SOC are shown in fig. S21e-h, where we notice a significant opening of the gap between the two outermost hole bands. The increase of this gap due to SOC is 0.004 eV at $\mathbf{k}_z = 0$ and 0.016 eV at $\mathbf{k}_z = 0.5 \pi/c$. The maximum SOC gap of 0.016 eV is consistent with the experimental value of 0.02 eV extracted from 10 UC $\text{FeTe}_{0.6}\text{Se}_{0.4}$ (Fig. 2). We further notice that after incorporating the SOC the innermost d_{xz} band splits. This splitting comes from the asymmetrical chalcogen placement in our calculations, where all Se atoms lie above the Fe plane and all Te atoms below it. This structural configuration leads to an enhanced spin-orbit splitting, which is not observed experimentally. In light of this theory-experiment disagreement, which is not fully understood, along with the fact that our main scope is on how strong correlation impacts topology and superconductivity, we find it appropriate to show the simpler DFT+eDMFT calculations without SOC in the main text.

Effect of SOC on FeTe: We compare the spectral functions with and without SOC for FeTe in fig. S22. The d_{xy} band becomes more incoherent and weaker near $\bar{\Gamma}$ when including the SOC, but no hybridization-induced effects are visible, matching our experimental results in main text Fig. 2.

12. Challenges in determining $\mathbf{k}_z$

It is a complex issue at which $\mathbf{k}_z$ we compare the ARPES data and DFT+eDMFT results. This issue is compounded by three difficulties. First it is established in prior ARPES studies^{16,17} that the weak experimental $\mathbf{k}_z$ dependence of the FTS band structure, especially for the top valence band below the TSS, is substantially different from first principles calculations. Second, with our thin film materials, one cannot trust the inner potentials used in the prior ARPES studies to determine $\mathbf{k}_z$. We notice that different prior ARPES studies on bulk FTS also used different inner potentials^{16,18}. Third, when the Te:Se ratio is varied, the inner potentials may also change as a function of the composition, making it impossible to use a single best $\mathbf{k}_z$ for the theory-experiment comparison.

These challenges are illustrated in fig. S23, where we compare DFT+eDMFT results for $\mathbf{k}_z = 0$, $0.25\pi/c$, and $0.5\pi/c$. The key experimental observations we look for in the calculations are: i) the spectral weights of the d_{xy} band near $\bar{\Gamma}$ and of the d_{xy}/d_{xz} hybridized band near $\bar{M}$ are suppressed in the strong correlation limit (FeTe with $h_{\text{Te}} = 1.85 \text{ \AA}$); ii) the innermost hole band near $\bar{\Gamma}$ becomes a "vertical streak" with xy characters in the strong correlation limit; iii) there is a small electron pocket of d_{xz}/d_{yz} characters near $\bar{\Gamma}$ in the strong correlation limit; iv) the d_{xy} band top is slightly lower than the d_{yz} band top near $\bar{\Gamma}$ according to fig. S17a. The calculation results for $\mathbf{k}_z = 0$ and $0.25\pi/c$ reproduce *Observations i)-iii)*, but not *iv)*. The results for $\mathbf{k}_z = 0.5\pi/c$ reproduce *Observations i)* and *iv)*, but not *ii)* or *iii)*. This reflects the aforementioned challenges of comparing theory and experiment at a single best $\mathbf{k}_z$. Since our study focuses on how strong electronic correlations impact topology and superconductivity, and both our Helium-lamp-based and laser-

based ARPES resolve *Observations i)* and *ii)*, we choose to present the results for $\mathbf{k}_z = 0$ in the main text. We do not rule out the possibility that perhaps $\mathbf{k}_z = 0.5\pi/c$ could be more appropriate for the theory-experiment comparison for the less correlated FTS compounds according to the *Observation iv)*. Such a d_{xy} - d_{yz} band crossing near $\bar{\Gamma}$ in $\text{FeTe}_{0.5}\text{Se}_{0.5}$ will also lead to a sizable hybridization gap when incorporating the spin-orbit coupling effect (supplementary text section 11).

13. Mismatch between the theoretical and experimental doping levels

Theoretical calculations for strongly correlated materials remain inherently challenging, and achieving quantitative agreement with experiments at the meV scale is not yet expected at this stage. Below we provide some plausible explanations for the theory-experiment discrepancy in doping levels:

- 1) As can be seen in Fig. 3c(i) and 3c(iii), stronger correlation generally pushes the band top of the d_{xy} band downward. As the DFT+eDMFT calculation for $\text{FeTe}_{0.5}\text{Se}_{0.5}$ still significantly underestimates the d_{xy} renormalization in 10 UC $\text{FeTe}_{0.6}\text{Se}_{0.4}$, it is plausible that the calculated band top appears at a higher energy.
- 2) As explained in Supplementary note 12 above, there is no single $\mathbf{k}_z$ at which the DFT+eDMFT calculation can reproduce all key features from our experiments using either 6 eV or 21.2 eV. In particular, for lighter Te content (e.g., $x \sim 0.5$), we do not rule out the possibility that $\mathbf{k}_z = 0.5 \pi/c$ can allow a more appropriate comparison with the experimental data. In this case, the band top of the d_{xy} band is also shifted below the band top of the d_{xz}/d_{yz} bands, resulting in band hybridization. We chose to present the DFT+eDMFT calculation at $\mathbf{k}_z = 0$ in the main text Fig. 3, with the consideration to focus more on the Te heavy side, where the calculated spectrum reproduces the experimental data very well (fig. S13).

14. Electrical transport measurements

As shown in fig. S25, superconducting transitions are observed in Te-heavy FTS thin films, even up to $x = 1$, which contrasts with bulk single crystals where superconductivity is suppressed by a bi-collinear antiferromagnetic order^{14,19} near the FeTe limit. The evolution of the R-vs-T curves when sweeping the out-of-plane magnetic field is consistent with the suppression of superconductivity by magnetism (fig. S9). The broad humps in the R-vs-T curves (fig. S25), which occur near 42 K, 63 K, and 76 K for $x = 1$, 0.95, and 0.93 respectively, were previously identified as signatures of a suppressed antiferromagnetic transition in FTS thin films¹. However, the doping dependence of the characteristic temperature is inconsistent with the antiferromagnetic phase boundary of the bulk FTS phase diagram²⁰. More investigations are needed to elucidate the nature of this hump feature.

15. Structural measurement of the FTS thin films

Atomic resolution Scanning Transmission Electron Microscopy (STEM) is employed to extract the lattice parameters of FTS thin films. The as-grown FTS films are capped with polycrystalline Te to protect the films. Fig. S26a presents a typical cross-sectional STEM image of a Te-capped $\text{FeTe}_{0.93}\text{Se}_{0.07}/\text{STO}$ heterostructure with a single layer of TiO_x between the STO and FTS. The film thicknesses of capped $\text{FeTe}_{0.93}\text{Se}_{0.07}$ and FeTe, as observed in STEM, range from approximately 9 to 13 unit cells within a single sample, with a nominal thickness reported as 10 unit cells. The averaged in-plane lattice constant for each layer is depicted in fig. S26b. Each measured in-plane lattice constant is determined from the average unit cell spacing across 30 unit cells of FTS. Pixel

size is calibrated for each image from the STO substrate. Across layers, we find an average in-plane lattice constant of $3.88 \text{ \AA}$ (st. err. $0.001 \text{ \AA}$), which indicate significant lattice strain compared to the bulk in-plane FeTe lattice constant of $3.82 \text{ \AA}$ ²¹, though it remains slightly smaller than the STO lattice spacing of $3.905 \text{ \AA}$. The out-of-plane chalcogen heights are also extracted from the STEM images, as shown in fig. S27. The average chalcogen heights above and below the Fe sheet in $\text{FeTe}_{0.93}\text{Se}_{0.07}$ are 1.83 and $1.66 \text{ \AA}$, respectively. These heights for FeTe are 1.81 and $1.67 \text{ \AA}$. Given that the statistical uncertainties in these extracted chalcogen heights are usually $\sim 0.02 \text{ \AA}$, we do not identify any significant structural difference between 10 UC $\text{FeTe}_{0.93}\text{Se}_{0.07}$ and FeTe films. Such a structure breaks the glide-mirror symmetry²² and should in principle result in a splitting of the electron pocket near the $\bar{\text{M}}$ point. However, previous studies on monolayer FeSe ²², which also breaks the glide-mirror symmetry, suggested that such a splitting can be negligible ($< 5 \text{ meV}$) near the electron pocket bottom.

16. Additional temperature-dependent ARPES measurements

Additional temperature-dependent ARPES measurements have been performed. Figure S28 shows the measurements on a 10 UC $\text{FeTe}_{0.6}\text{Se}_{0.4}$ film. This sample is topologically trivial and displays no clear gap opening down to 8 K , consistent with our transport measurements showing an onset temperature of 7.3 K . Fig. S29 shows similar measurements on a 10 UC FeTe film. A possible gap opening can be identified around 8 K ; however, the evidence remains inconclusive due to the limited energy resolution and the weak, incoherent spectral weight near E_F . Further high-resolution measurements will be required to establish whether a superconducting gap emerges in FeTe.

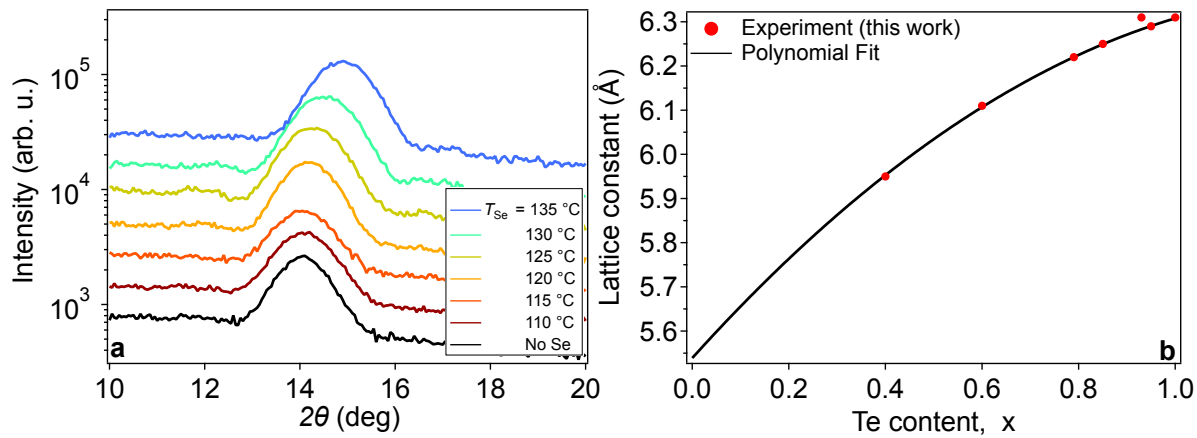


Fig. S1 **Structural characterization of $\text{FeTe}_x\text{Se}_{1-x}$.** (a) X-ray diffraction (XRD) scans for 10 UC $\text{FeTe}_x\text{Se}_{1-x}$ samples grown at different Se temperatures. The diffraction patterns are vertically shifted for clarity. (b) c -axis lattice constant as a function of the Te content. The black curve represents the polynomial relationship between c -axis lattice constant and Te content in the literature¹, while the red dots are experimental data points from this study.

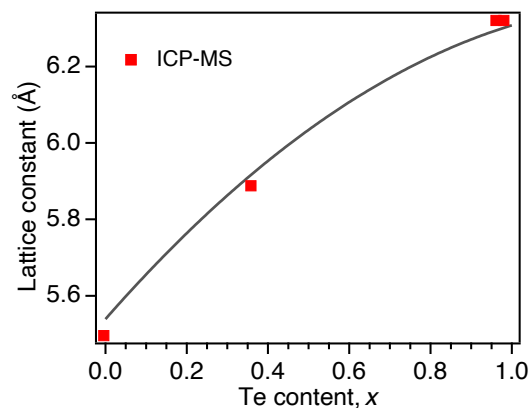


Fig. S2 **Comparison of two methods to determine Te:Se ratios in $\text{FeTe}_x\text{Se}_{1-x}$ thin films.** Black curve: the polynomial relationship between c -axis lattice constant and Te content in the literature¹. Red squares: Te content determined independently by inductively coupled plasma mass spectrometry (ICP-MS).

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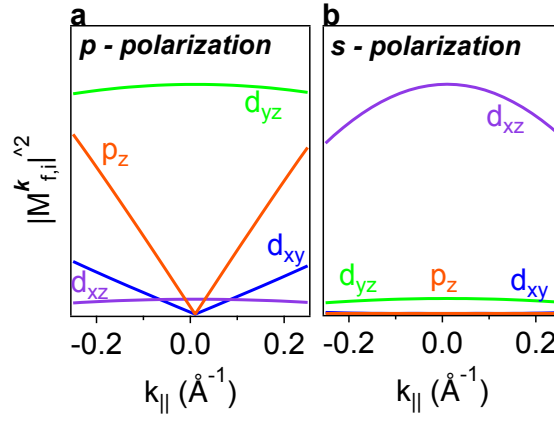


Fig. S3 **Matrix element magnitude.** Matrix element magnitude along the $\bar{\Gamma} - \bar{X}$ direction for Fe d_{xy} , d_{xz} , d_{yz} and chalcogen p_z orbitals. Results for p - and s -polarized light are shown in panels (a) and (b), respectively.

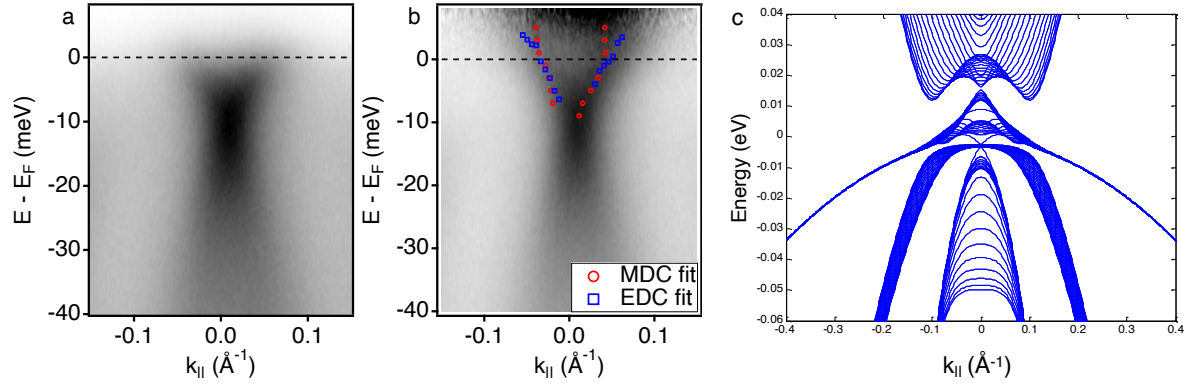


Fig. S4 The band dispersion of the TSS for 10 UC $\text{FeTe}_{0.93}\text{Se}_{0.07}$ films at 25 K. (a) Raw spectrum of $\text{FeTe}_{0.93}\text{Se}_{0.07}$. **(b)** Spectrum after division by the Fermi-Dirac function convolved with a Gaussian resolution function. Red circles indicate the TSS band positions obtained from MDC fitting, while blue squares represent the band positions obtained from EDC fitting. **(c)** Slab calculations for a 21-layer $\text{FeTe}_{0.5}\text{Se}_{0.5}$ showing how the TSS connects to the bulk band.

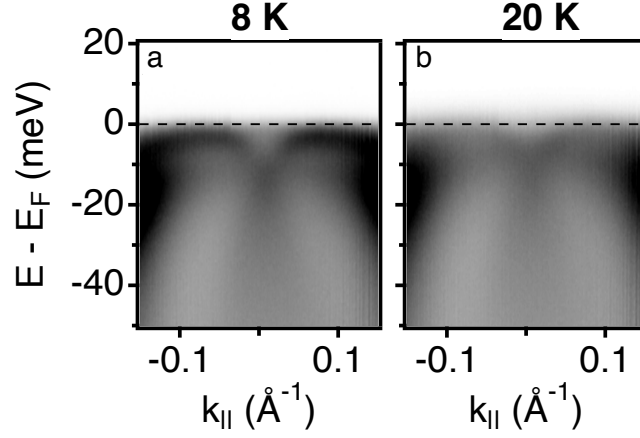


Fig. S5 **Comparison between the spectra in the superconducting and normal states for 10 UC $\text{FeTe}_{0.93}\text{Se}_{0.07}$ film.** (a) ARPES data taken with p -polarized light at 8 K. (b) Same as A but taken at 20 K. The “flat band” near E_F observed at 8 K disappears in the normal state at 20 K. It originates from the superconducting gap opening of the bulk d_{xy}/d_{yz} band. The spectral weight of this bulk-driven flat band dominates the ARPES spectrum at 8 K, making it challenging to observe the intrinsic superconducting gap of the TSS.

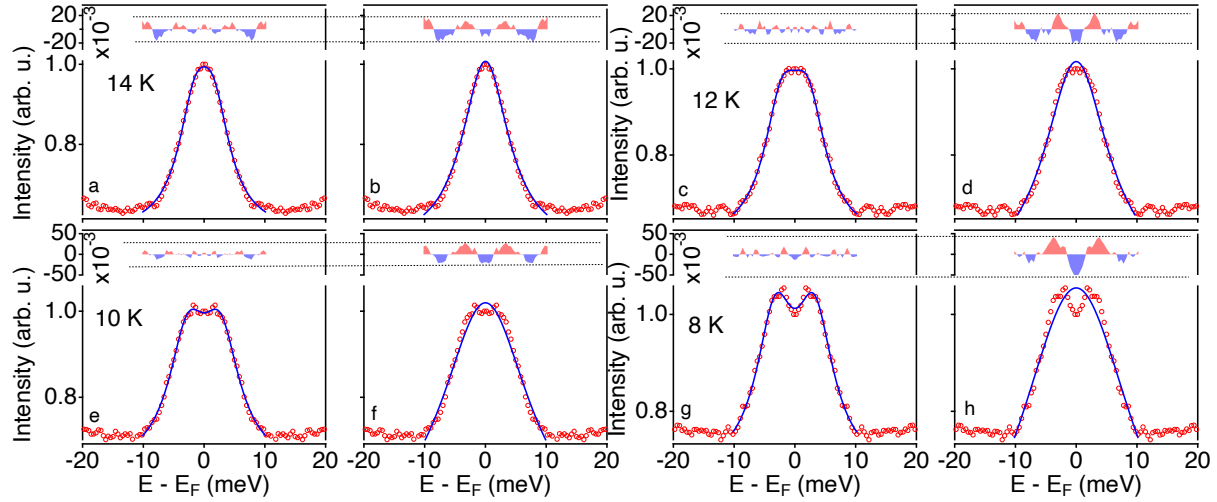


Fig. S6 Checking the superconducting gap closing temperature using symmetrized EDCs extracted from the data taken with *s*-polarized photons. (a) Symmetrized EDCs at 14 K fitted by a double-Lorentzian function. Top curve shows the fitting residual. **(b)**, Same as A, but fitted with a single Lorentzian. **(c, e, g)** Same as A, but for 12 K, 10 K, and 8 K, respectively. **(d, f, h)**, Same as B, but for 12 K, 10 K, and 8 K, respectively.

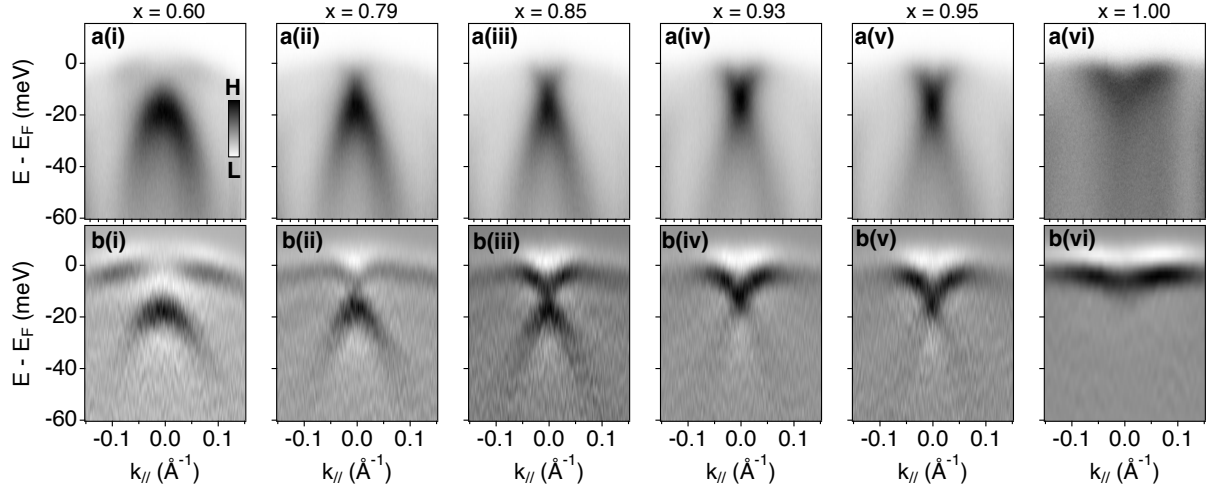


Fig. S7 Probing the band topology in 10 UC $\text{FeT}_x\text{Se}_{1-x}$ films using s -polarized 6 eV laser-based ARPES. (a) ARPES spectra near the $\bar{\Gamma}$ point for 10 UC films with the different tellurium content (x). (b) Corresponding second derivative plots of the raw data in A. Due to the Fermi edge, second derivative spectra near the Fermi level may not correctly reflect the band dispersions. To extract the Fermi crossings, one should turn to the EDC or MDC analysis in Fig. S4.

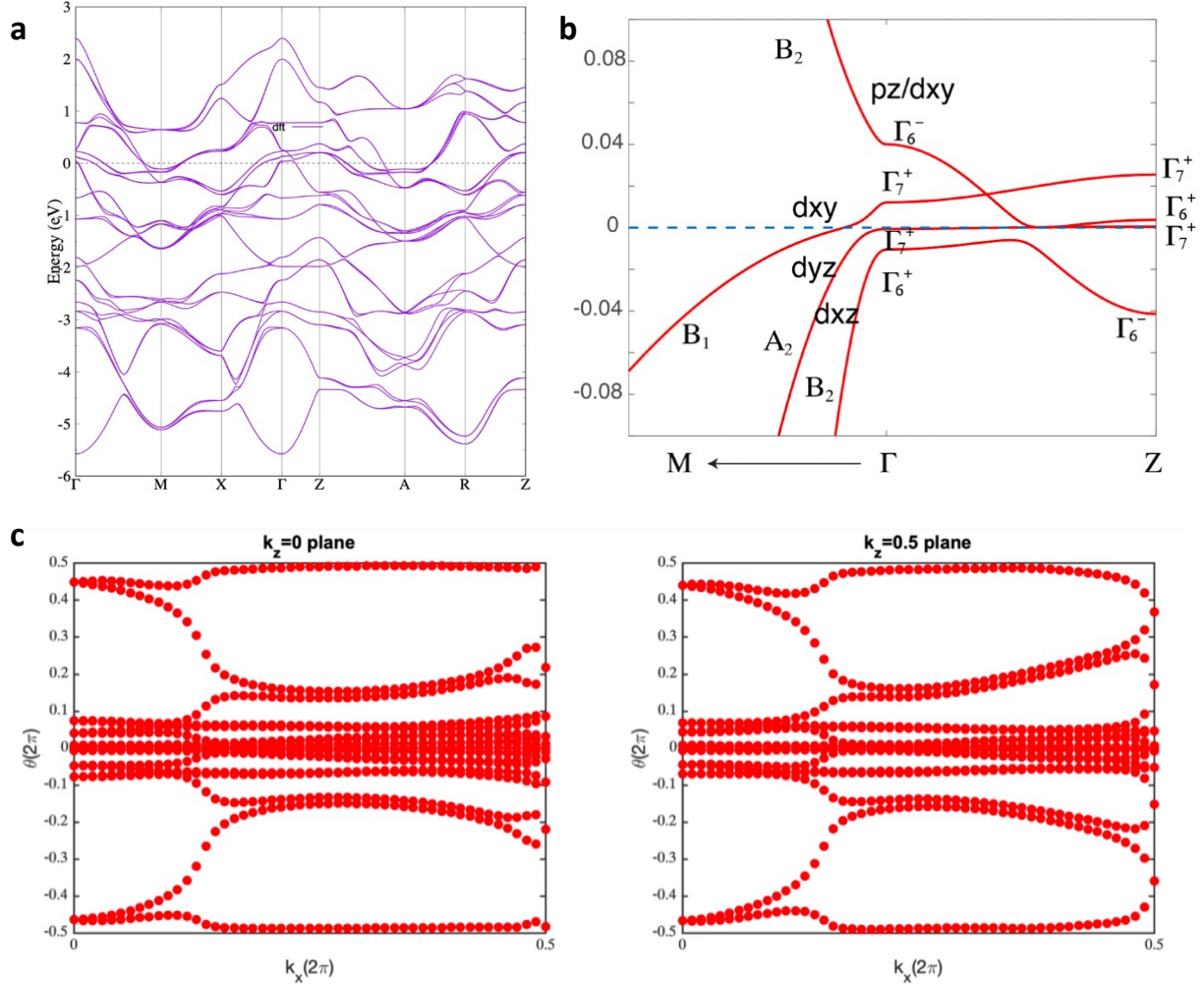


Fig. S8 **Nontrivial topology revealed by theory for FTS films.** (a) DFT band structure with asymmetric Se/Te height. (b) Band dispersion along the M- Γ -Z line for FTS. The orbital characters, parities and irreducible representations at the high-symmetry points and path are indicated. (c) Wannier centers evolution as a function of k_x in $k_z = 0$ (left) and $k_z = \pi$ (right) planes.

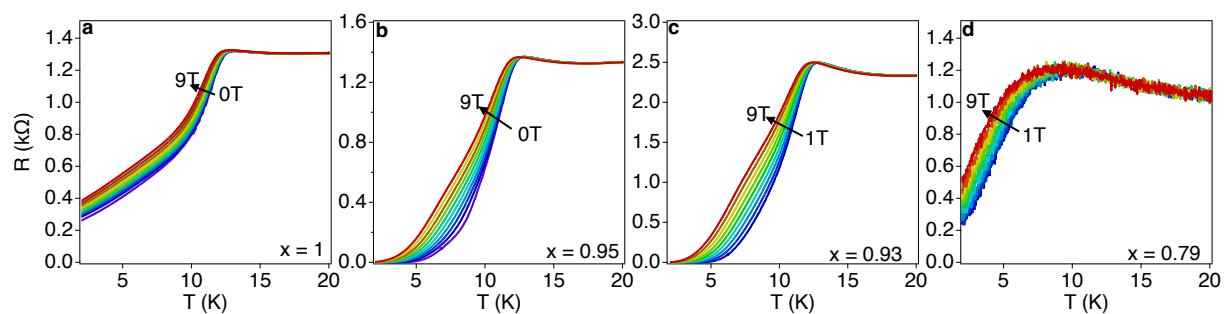


Fig. S9 **Magnetic field dependence of the resistances for FTS films.** R-vs-T curves under different magnetic fields for FTS films with Te contents of (a) 1, (b) 0.95, (c) 0.93, and (d) 0.79, respectively.

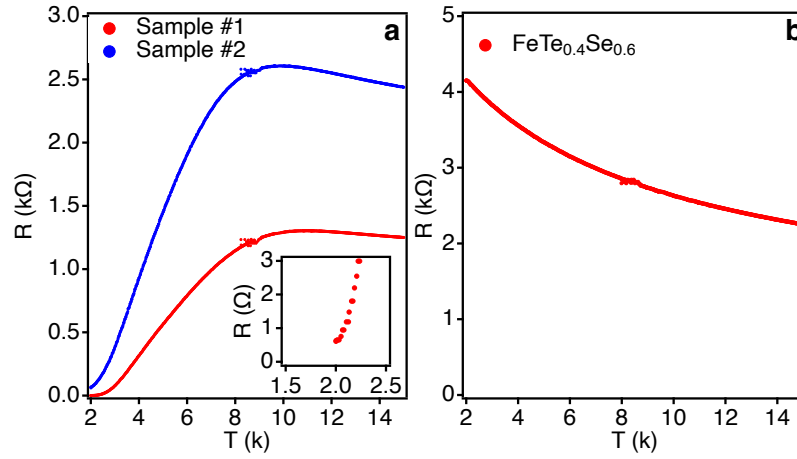


Fig. S10 **Resistance-vs-Temperature (R-vs-T) curves for the trivial FeTe_{0.6}Se_{0.4} (a) and FeTe_{0.4}Se_{0.6} (b).** It appears that there is an overall factor-of-two scaling between the two data sets for $x = 0.6$. This is due to the limited controllability of our Indium dot fabrication, which results in varied geometries for the resistance measurements.

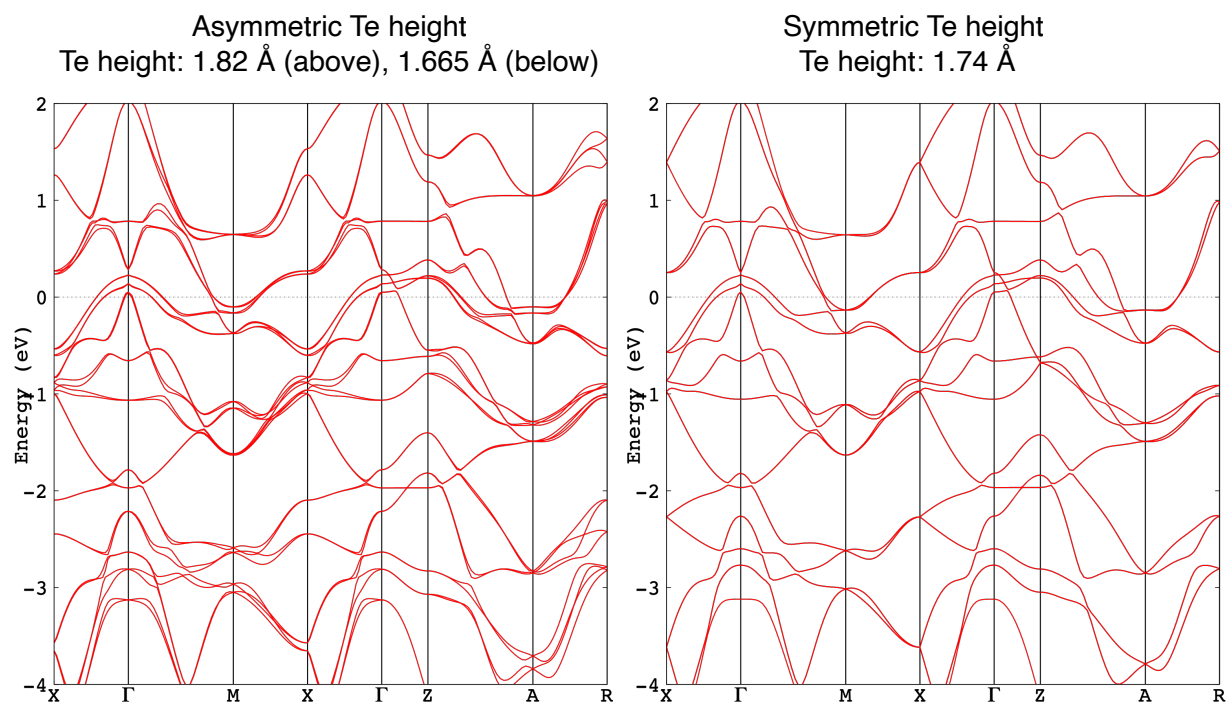


Fig. S11 **DFT band structure with symmetric Te height for FeTe.** DFT predicts a topologically non-trivial phase for FeTe.

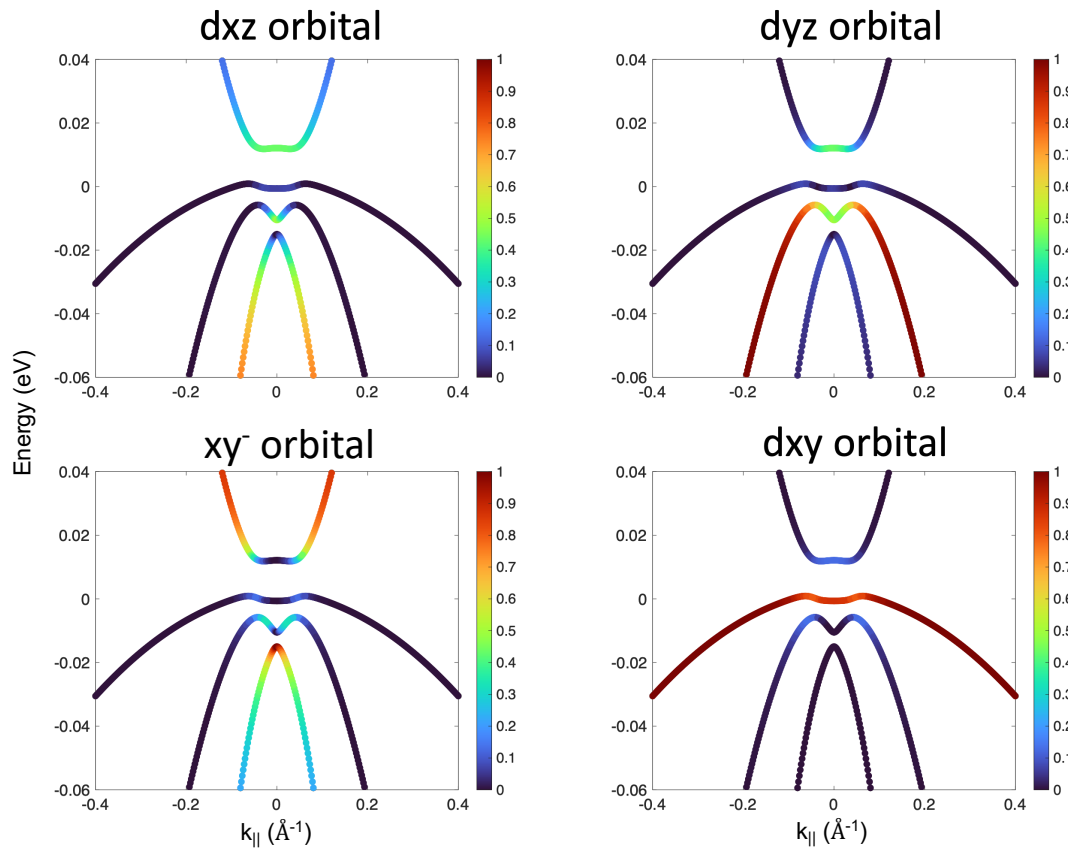


Fig. S12 **Orbital projections of a tight-binding model for the complete band inversion between the xy and d_{xz} orbitals.**

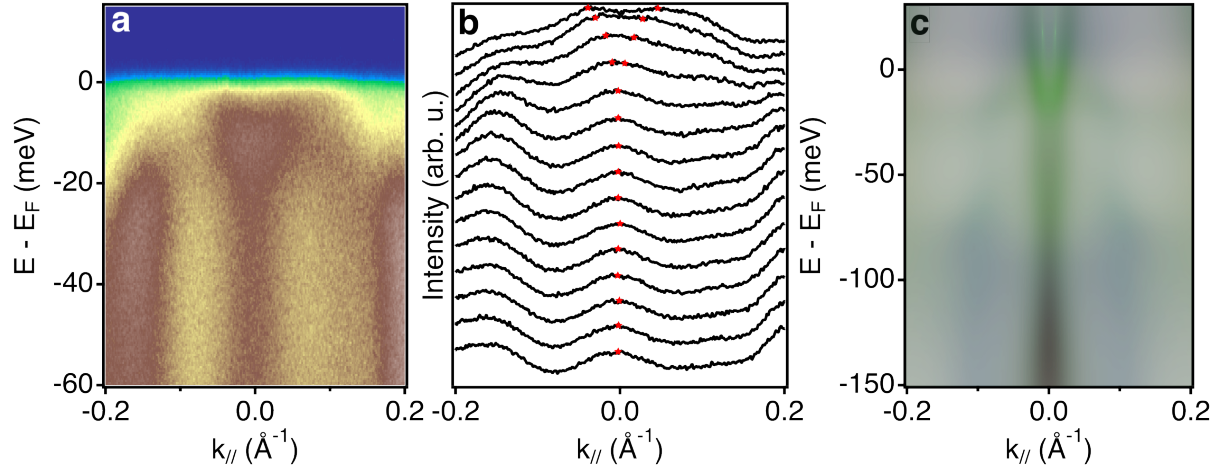


Fig. S13 **ARPES and DMFT results on FeTe thin films.** (A) ARPES data taken with p -polarized light. (B) Corresponding MDCs extracted from A. Red stars indicate the dispersion of the electron pocket, serving as guides to the eye. (C) DFT+eDMFT calculations for FeTe with a Te height of 1.85 $\text{\AA}$. The calculated spectrum captures the electron pocket and the “vertical streak” feature.

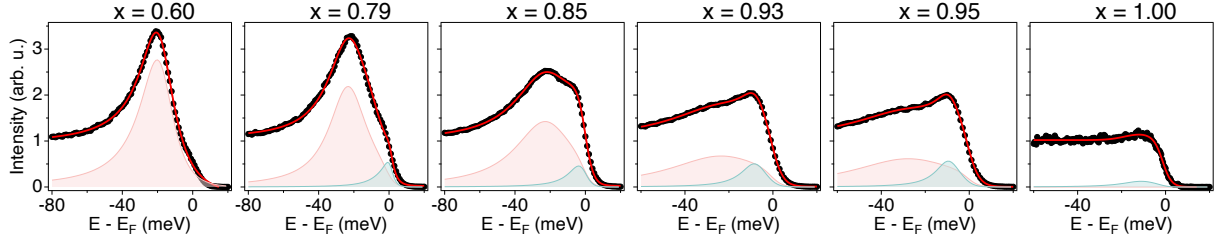


Fig. S14 Fitting the EDCs cutting through the Dirac cones. EDCs (black dots) for films with different Te content (x) taken at $-0.025 \text{ \AA}^{-1}$ of the laser-ARPES data from main text Fig. 2a, fit by summing two Lorentzian functions (red and green shades) and a constant background, and multiplying the sum with a Fermi-Dirac function. The composite fitting functions are shown as red solid curves.

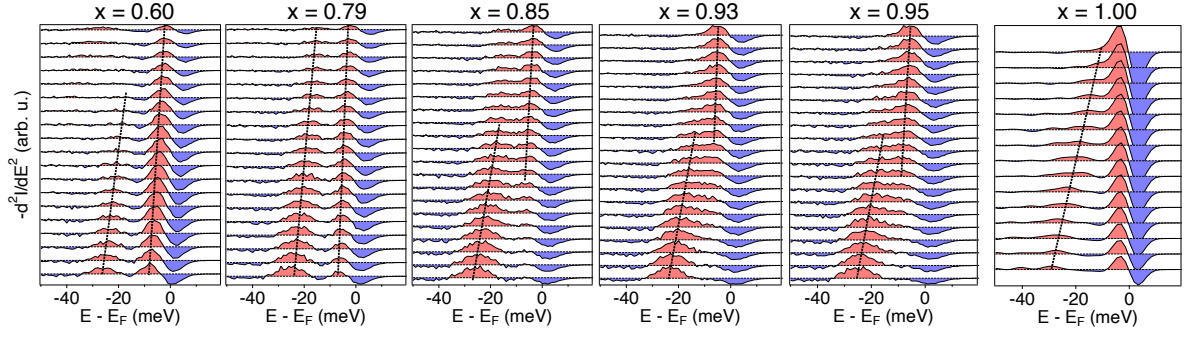


Fig. S15 **Hybridization gap magnitude between the d_{xy} and d_{yz} bands near $\bar{\Gamma}$ as a function of the Te content (x).** Inverted second derivative curves corresponding to momentum-dependent EDCs in the range between -0.065 and $-0.165 \text{ \AA}^{-1}$ are plotted. The hybridization gap (main text Fig. 2e) was determined by fitting two Lorentzian peaks to the second derivative of the EDC integrated over the momentum range of -0.11 to $-0.12 \text{ \AA}^{-1}$, except for FeTe, where only a single peak could be identified. Dashed lines are guides to the eye indicating the band dispersion.

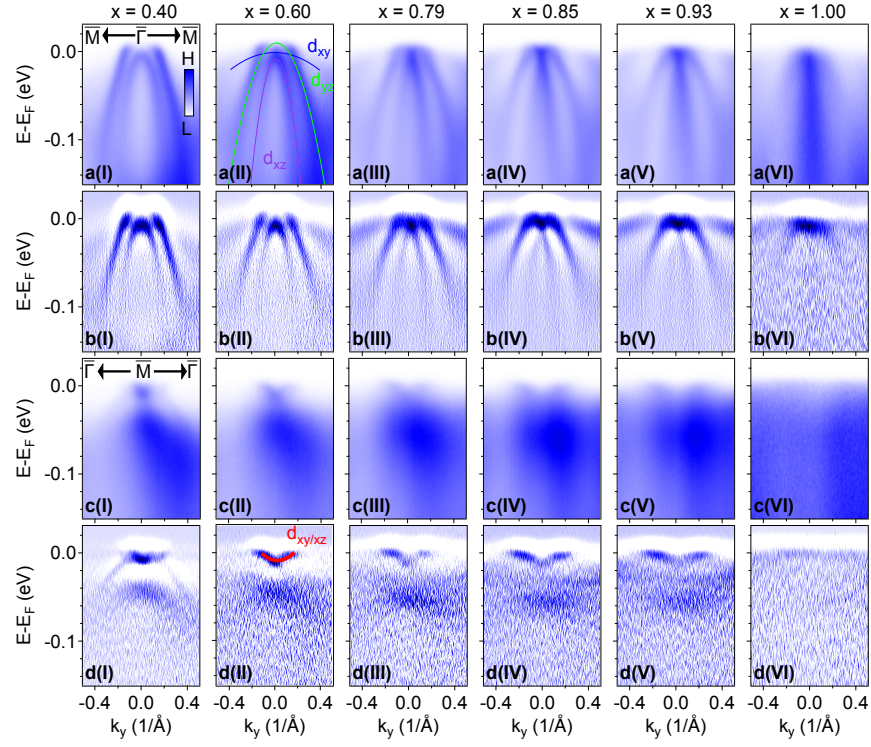


Fig. S16 **Experimental spectral functions of 10 UC FeTe_xSe_{1-x} films measured by ARPES using 21.2 eV photons.** (a) Evolution of the ARPES spectra as a function of the Te content near the $\bar{\Gamma}$ point. (b) Corresponding 2D curvature plots of the raw data in a, extracted using the protocol reported by Ref.¹¹. (c, d) Same as a and b, but near the $\bar{M}$ point. All ARPES measurements were done at a sample temperature of 20 K.

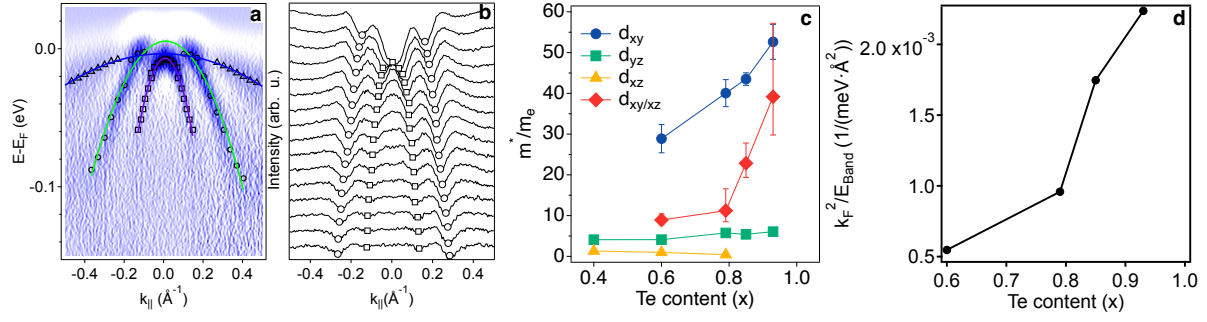


Fig. S17 **Extracting the band dispersion and effective mass.** (a) 2D curvature plot for $\text{FeTe}_{0.6}\text{Se}_{0.4}$. (b) MDCs of the 2D curvature plots. (c) Extracted effective masses as a function of Te content (x) normalized by the free-electron mass for the d_{xz} , d_{yz} and d_{xy} bands near $\bar{\Gamma}$, as well as the hybridized $d_{xy/xz}$ band near $\bar{M}$. Error bars indicate one-standard-deviation (1σ) uncertainties of the fitting results. (d) k_F^2/E_{Band} for the $d_{xy/xz}$ band as a function of Te content (x).

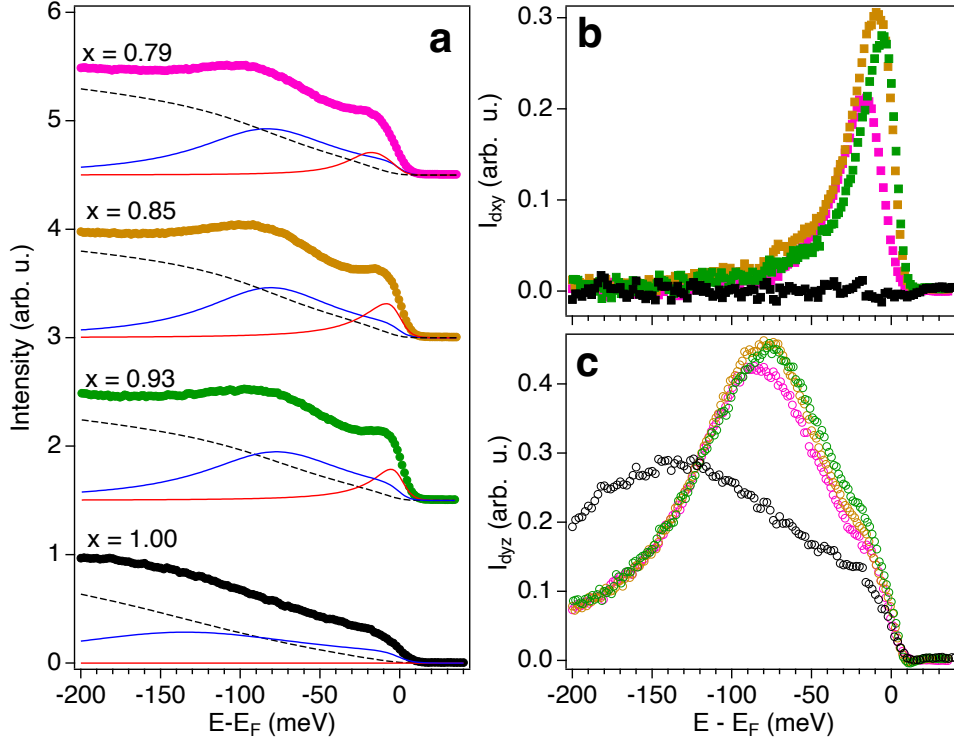


Fig. S18 **Rapid loss of the d_{xy} spectral weight toward the FeTe limit.** (a) EDCs for films with different Te content (x) integrated between 0.375 and 0.425 $\text{\AA}^{-1}$ of the 21.2 eV ARPES data from fig. S16a, fit by summing two Lorentzian functions (blue and red lines) and a constant, multiplied by a Fermi-Dirac function, and then adding a Shirley type background (black dash lines)²³. (b) Fitted d_{xy} band spectral weight as a function of energy. (c) Fitted d_{yz} band spectral weight as a function of energy. The d_{xy} band loses spectral weight in the FeTe limit.

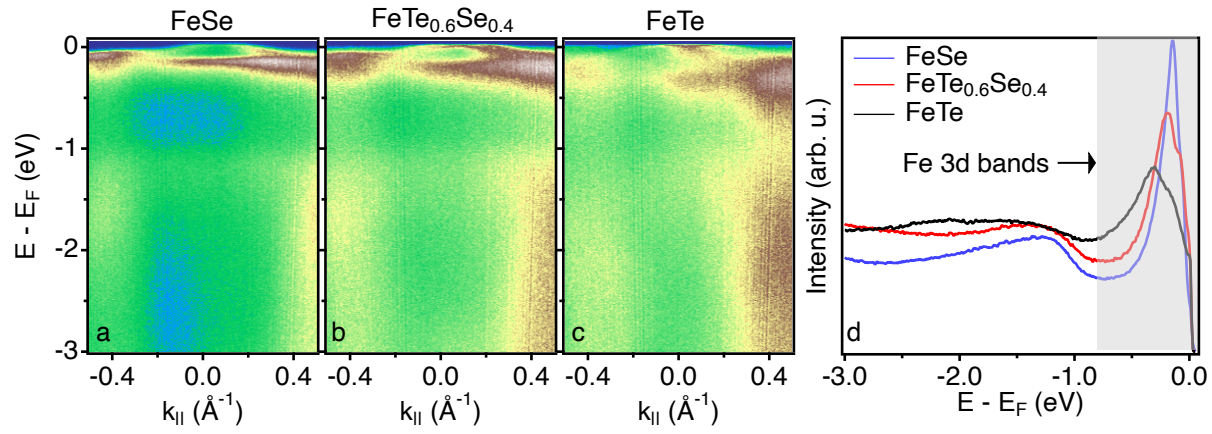


Fig. S19 **Valence band structures for FeTe_xSe_{1-x}**. Valence band structures for 10 UC (a) FeSe, (b) FeTe_{0.6}Se_{0.4}, and (c) FeTe. (d) EDCs for these three samples integrated over a momentum range from -0.5 to 0.5 Å⁻¹.

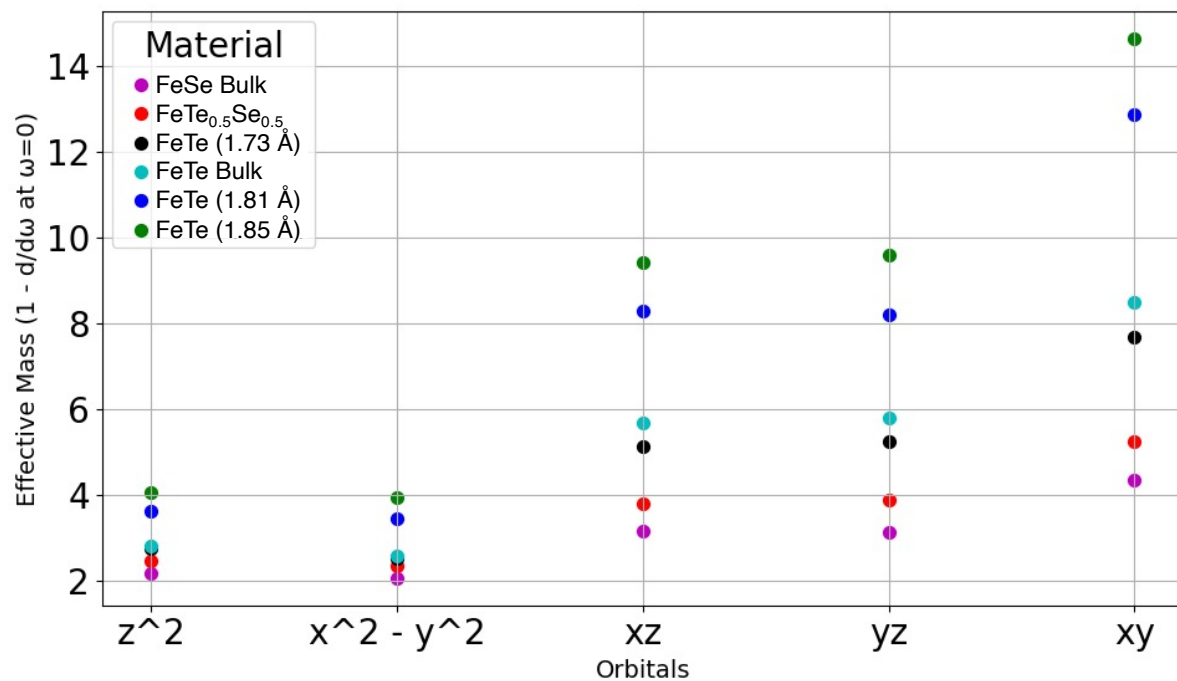


Fig. S20 eDMFT computed effective masses (Z^{-1}) with respect to DFT band masses for FTS as a function of Te-height. The results for FeTe_{0.5}Se_{0.5}, as well as the bulk phase of FeSe and FeTe, are also compared.

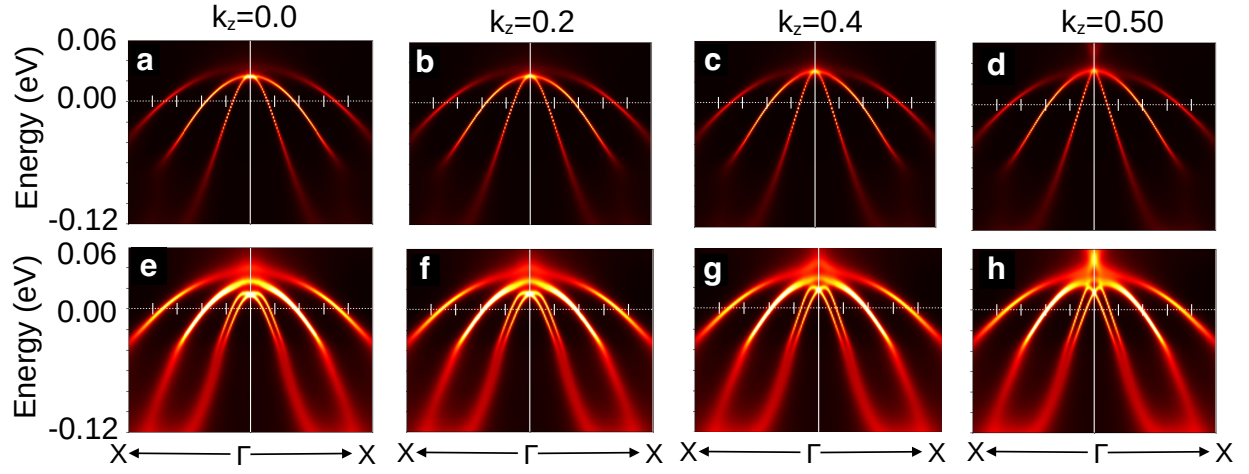


Fig. S21 **Effect of spin-orbit coupling (SOC) on FeTe_{0.5}Se_{0.5}**. DFT+eDMFT spectral functions for FeTe_{0.5}Se_{0.5} along X – Γ – X without SOC (top row) and the same with SOC (bottom row). This comparison shows the d_{xy} - d_{yz} hybridization gap opening with SOC for (a, e) $k_z = 0$, (b, f) $k_z = 0.2\pi/c$, (c, g) $k_z = 0.4\pi/c$, and (d, h) $k_z = 0.5\pi/c$.

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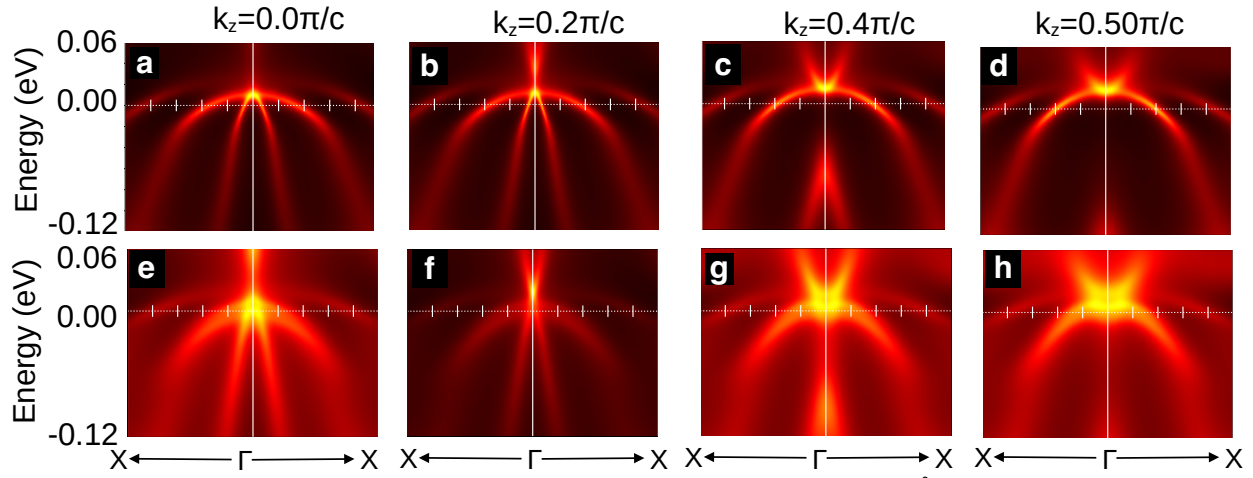


Fig. S22 **Effect of spin-orbit coupling (SOC) on FeTe.** $h_{\text{Te}} = 1.73 \text{ \AA}$. DFT+eDMFT spectral functions for FeTe along $X - \Gamma - X$ without SOC (top row) and the same with SOC (bottom row), shown for different k_z 's: (a, e) $k_z = 0$, (b, f) $k_z = 0.2\pi/c$, (c, g) $k_z = 0.4\pi/c$, and (d, h) $k_z = 0.5\pi/c$.

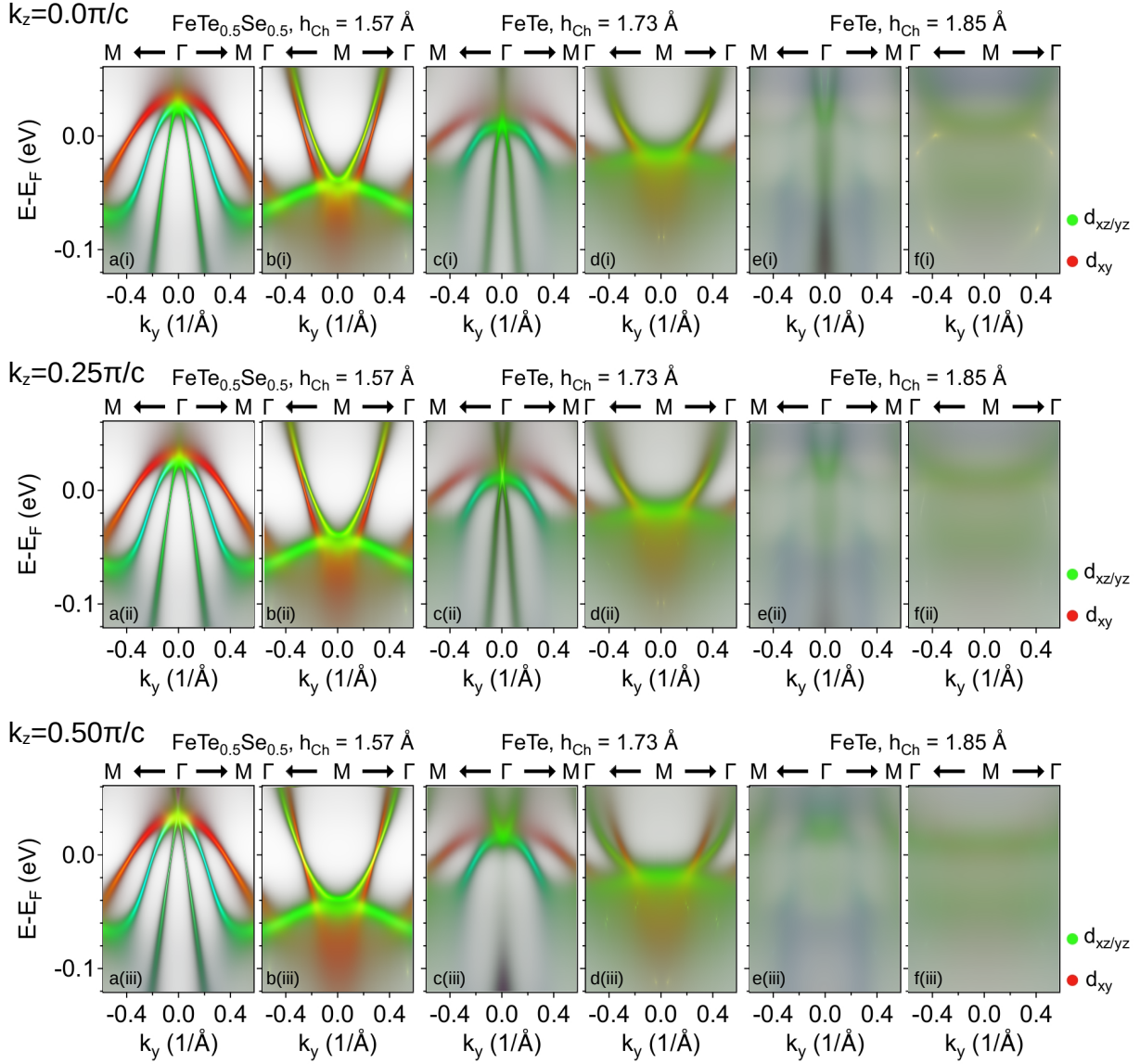


Fig. S23 DFT+eDMFT results for selected k_z 's. (a(i)-f(i)) Calculation results at $k_z = 0$ for (a(i), b(i)) $\text{FeTe}_{0.5}\text{Se}_{0.5}$, (c(i), d(i)) FeTe with an eDMFT-relaxed Te height, and ((e(i), f(i))) FeTe with a fixed Te height of 1.85 Å. (a(ii)-f(ii)) Calculation results at $k_z = 0.25\pi/c$. (a(iii)-f(iii)) Calculation results at $k_z = 0.5\pi/c$.

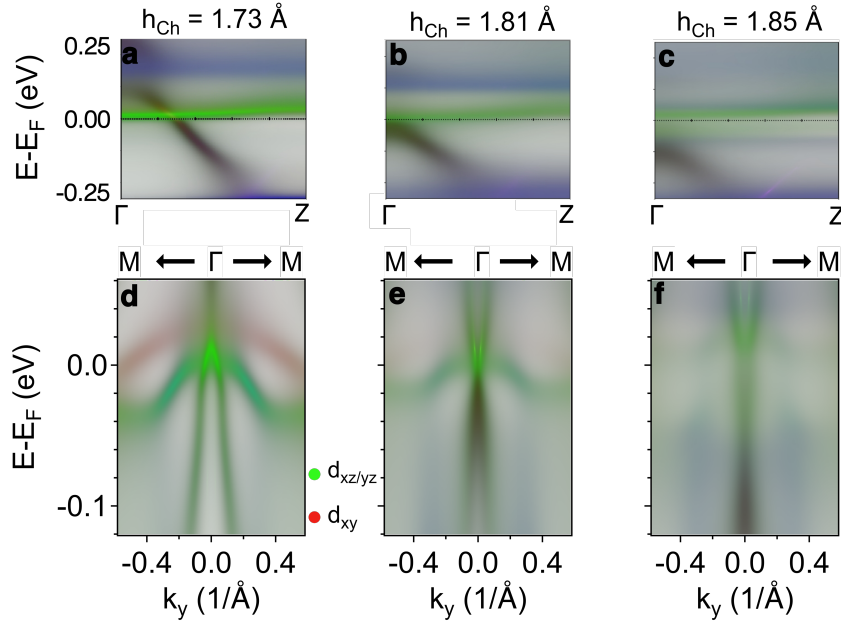


Fig. S24 **Tuning electronic correlations by adjusting the Te height.** (a-c) Orbital-resolved spectral functions calculated by DFT+eDMFT for FeTe as a function of different Te heights along the $\Gamma - Z$ direction. (d-f) Same as a-c, but along the $M - \Gamma - M$ direction.

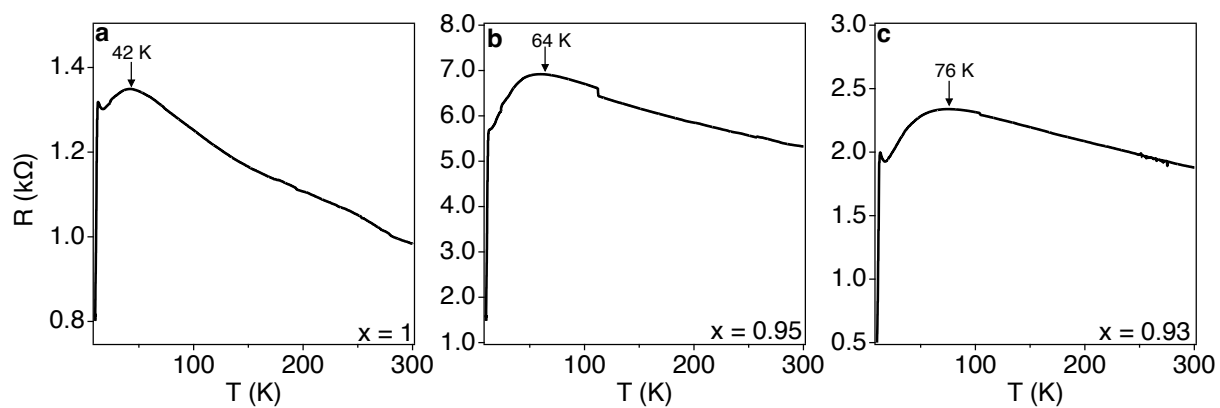


Fig. S25 **R-vs-T curves for the Te-heavy films.** R-vs-T curves are shown for FTS films with Te contents of (a) 1, (b) 0.95, (c) 0.93.

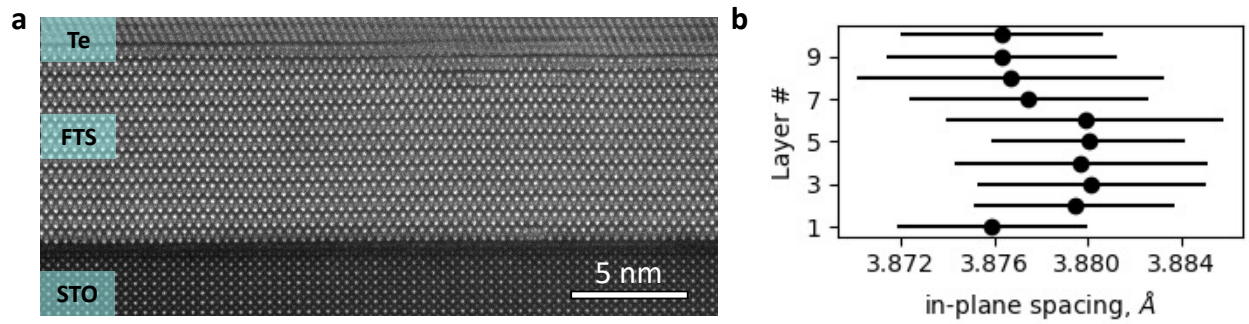


Fig. S26 **Extracting in-plane lattice constants.** (a) STEM image of a 10 UC FeTe_{0.93}Se_{0.07} film imaged along the [100] direction. FTS is grown on STO substrate and capped with Te protection layers. A visible defect to the right of the field of view in the first FTS layer likely accommodates part of the strain. (b) Extracted in-plane lattice constant for each FeTe_{0.93}Se_{0.07} layer. In-plane lattice constants are determined by averaging the lattice spacing across 30 UC of FeTe_{0.93}Se_{0.07}, with the values shown representing the average of measurements taken from three distinct regions of the sample. Pixel size is calibrated based on the STO substrate in each image.

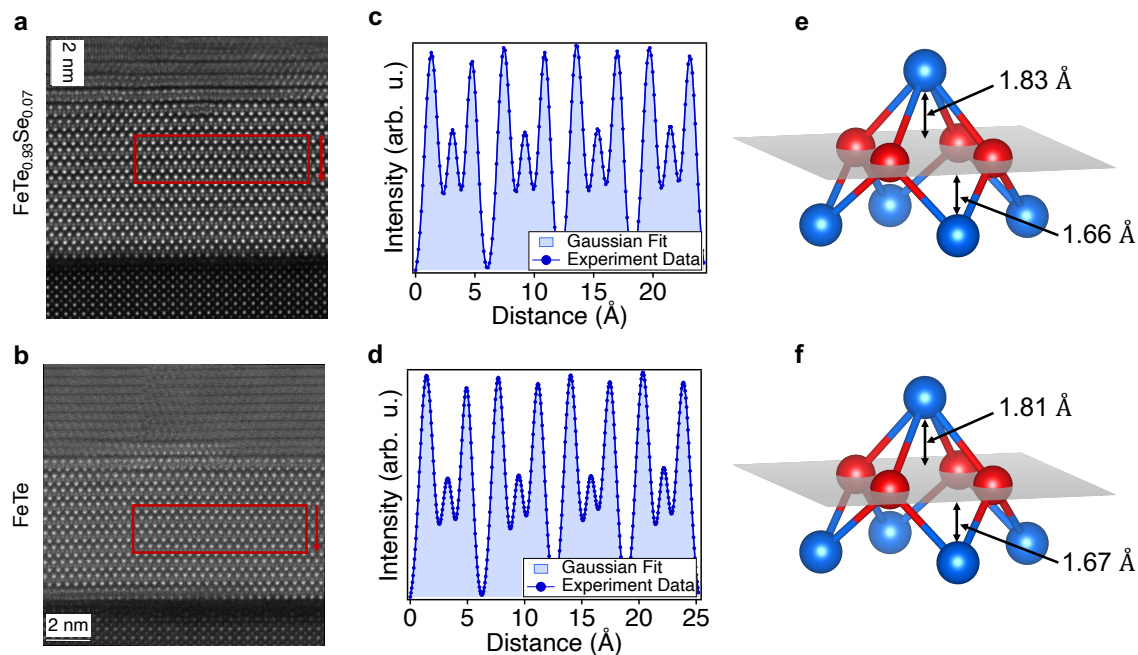


Fig. S27 **Extracting the chalcogen heights from STEM.** (a) Selected STEM image of a 10 UC $\text{FeTe}_{0.93}\text{Se}_{0.07}$ film capped by Te protection layers, projected along the [100] direction. (b) Same as a, but for 10 UC FeTe. (c) Intensity line profiles derived from the region marked in the red box in panel A. (d) Intensity line profiles derived from the region marked in the red box in panel B. (e, f) Extracted averaged chalcogen heights above and below the Fe sheet. Structural models are visualized using VESTA²⁴.

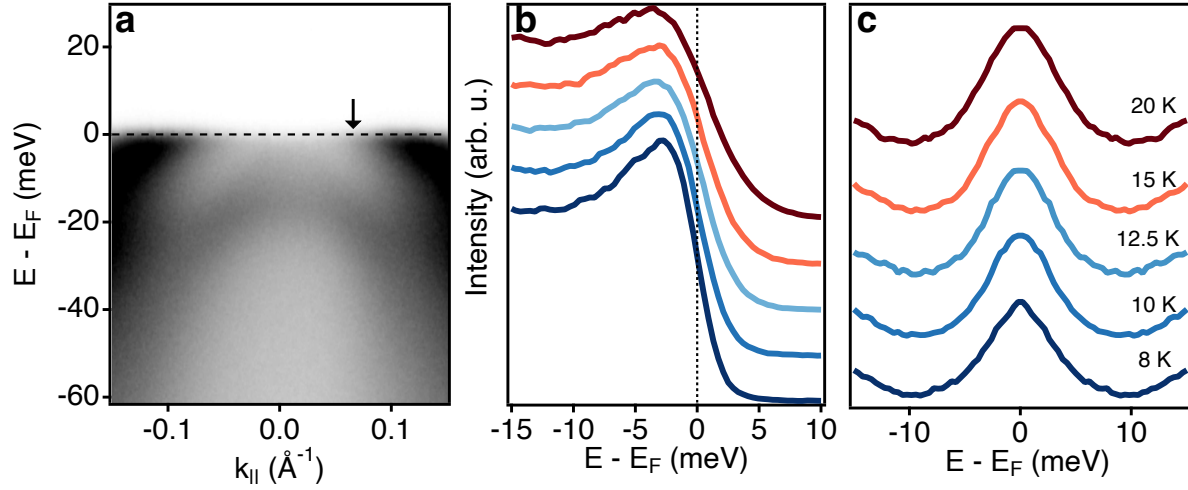


Fig. S28 **Temperature-dependent ARPES on 10 UC FeTe_{0.6}Se_{0.4}**. (a) ARPES spectrum on 10 UC FeTe_{0.6}Se_{0.4} measured at 8 K using *p*-polarized 6 eV photons. The black arrow indicates the momentum at which energy distribution curves (EDCs) are taken. We select the momentum where the energy gap at E_F is minimized. (b) Selected EDCs at various temperatures. (c) EDCs symmetrized around the zero energy.

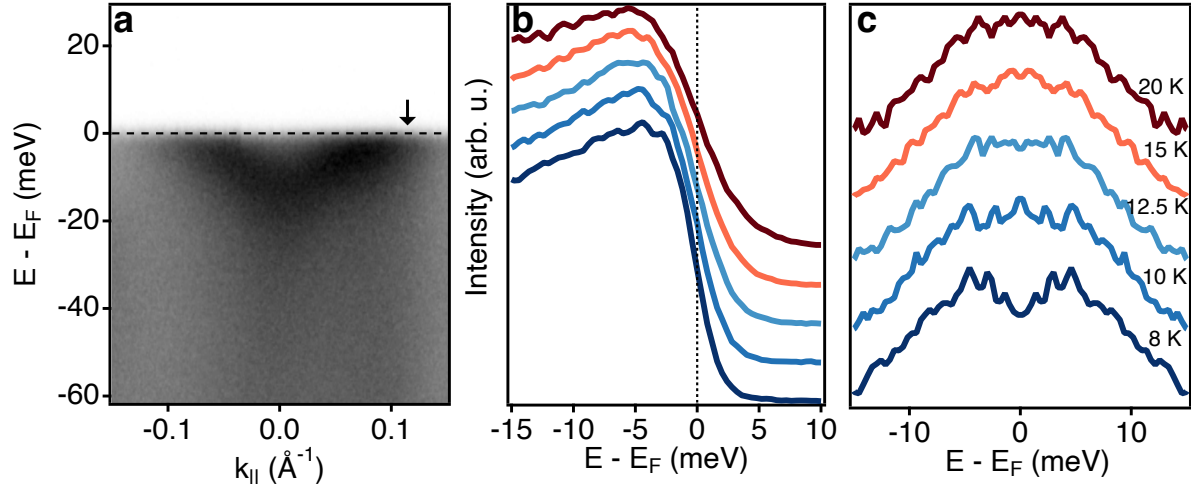


Fig. S29 **Temperature-dependent ARPES on 10 UC FeTe.** (A) ARPES spectrum on 10 UC FeTe measured at 8 K using *s*-polarized 6 eV photons. The black arrow indicates the momentum at which energy distribution curves (EDCs) are taken. We select the momentum where the energy gap at E_F is minimized. (B) Selected EDCs at various temperatures. (C) EDCs symmetrized around the zero energy.

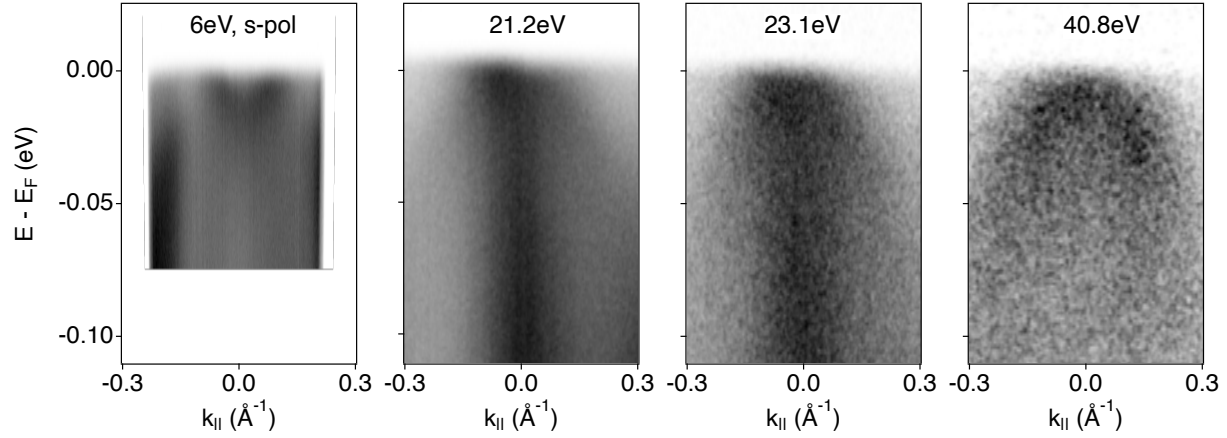


Fig. S30 **Photon energy dependent ARPES on 10 UC FeTe**. ARPES spectra on 10 UC FeTe measured at 8 K with different photon energies. The “vertical streak” feature due to the strong correlation effect is observed by 21.2 eV (He 1 α) and 23.1 eV (He 1 β), as well as 6 eV but with a *p*-polarization (main text Fig. 2a(vi)). It is not seen with the 40.8 eV (He 2 α) photon energy due to the suppressed matrix element for the *p_z* orbital component¹⁷. The shallow electron pocket at Γ is visible with 6 eV photons but not seen using other photon energies. Its k_z dependence indicates that this pocket is likely of bulk rather than surface origin.

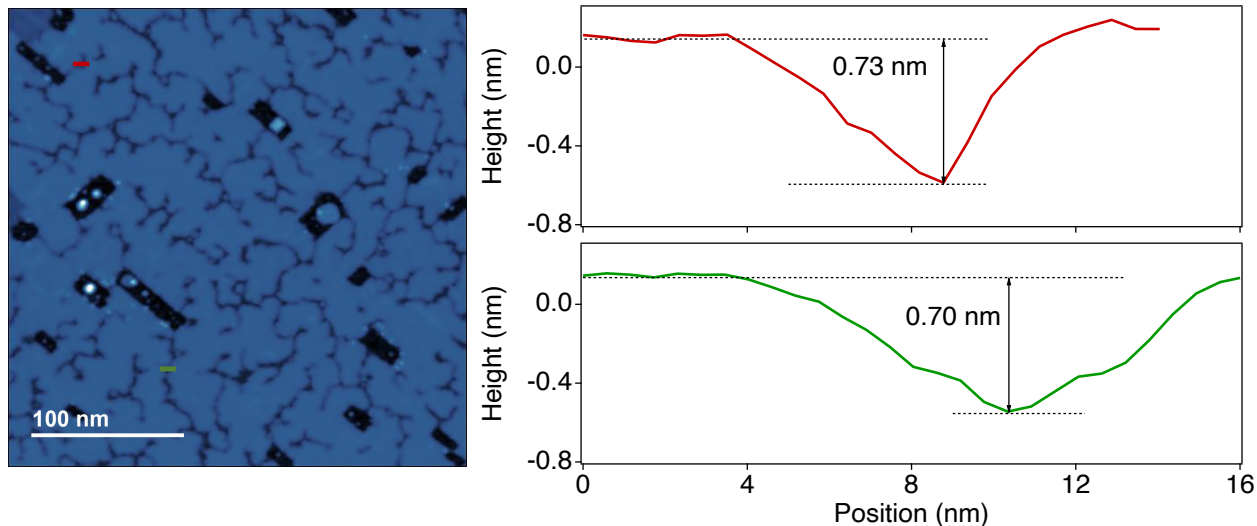


Fig. S31 **Scanning tunneling microscope (STM) characterization of monolayer FTS film.** Topography image of 0.8 UC $\text{FeTe}_{0.79}\text{Se}_{0.21}$, with step heights shown in the right panel. The step height measured by STM across the edge of a monolayer film is higher than the c -axis lattice constant of FTS, because the smaller density of states on the STO substrate requires the STM tip to get closer to the STO surface to maintain the same tunneling current.

Table S1. **Matrix element magnitude under different polarizations.**

	<i>p</i> -polarization	<i>s</i> -polarization
d_{xz}	S	E
d_{yz}	E	S
d_{xy}	E	S
p_z	E	S

Note: E (S) stands for enhanced (suppressed) matrix element magnitude.

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