

A Topological Superconductor Tuned by Electronic Correlations

Corresponding Author: Dr Shuolong Yang

This manuscript has been previously reviewed at another journal. This document only contains information relating to versions considered at Nature Communications.

This file contains all reviewer reports in order by version, followed by all author rebuttals in order by version.

Version 0:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

From the revision, the authors significantly advanced their discussion on the relation between topological superconductivity and orbitally selective electronic correlation in the $\text{FeTe}_x\text{Se}_{1-x}$ material, 10 uc thin film on the SrTiO_3 substrate.

However, I have a critical concern about the changed point of the revised manuscript as follows. From the ARPES data on FeTe , the data now show a strongly photon energy-dependent electron pocket around the Gamma point, as shown in Fig.2. [a(vi) and b(vi)]. The authors speculated that this electron pocket is a bulk band having a d_{xz} origin [line 141-142]. The authors interpreted that this band is not a surface band, and changed their conclusion that $\text{FeTe}_x\text{Se}_{1-x}$ at the Te end experiences the quantum topological transition.

The origin of the electron pocket around the Gamma point should be clearly resolved to support this argument. In the absence of the folded band from the expansion of the unit cell, there is only one electron band in the in-plane direction around the Gamma point, which is the $xy^{\{-}}$ band. Please check the following reference [V. Cvetkovic and O. Vafeek, Phys. Rev. B 88, 134510 (2013)] for this information. One possibility is that the vertical streak is also from the $xy^{\{-}}$ band experience decoherence from electronic correlation and photon energy-dependent layer-to-layer contributions. This appearance of the $xy^{\{-}}$ bulk electron band could be a signature of the topological transition since the uppermost position of the $xy^{\{-}}$ band along the z direction is now positioned far below the Fermi level compared to the surface band energy position. However, the author has to clarify all these issues to make sure of their new argument of the quantum topological transition around the Te end.

Reviewer #3

(Remarks to the Author)

I recommend publication in Nature Comm.

Reviewer #4

(Remarks to the Author)

The authors have made great efforts in their responses to the questions and addressed my concerns in the revised manuscript. I recommend the publication of this manuscript in Nature Communications.

Version 1:

Reviewer comments:

Reviewer #1

(Remarks to the Author)

In the revised manuscript and the reply, the authors appropriately addressed my issues. It is appreciated to show that a topologically trivial electronic structure could have a band inversion between the $xy^{\{-}}$ band and the xz/yz band at the Gamma point, which can be a signature of a finite quasiparticle residue of the $d_{\{xy\}}$ orbital.

I agree to publish this manuscript in the Nature Communications journal.

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Referees' comments:

Referee #1 (Remarks to the Author):

From the revision, the authors significantly advanced their discussion on the relation between topological superconductivity and orbitally selective electronic correlation in the $\text{FeTe}_x\text{Se}_{1-x}$ material, 10 uc thin film on the SrTiO_3 substrate.

We thank the referee for acknowledging the significant advance in our revised discussion.

However, I have a critical concern about the changed point of the revised manuscript as follows. From the ARPES data on FeTe, the data now show a strongly photon energy-dependent electron pocket around the Gamma point, as shown in Fig.2.[a(vi) and b(vi)]. The authors speculated that this electron pocket is a bulk band having a d_{xz} origin [line 141-142]. The authors interpreted that this band is not a surface band, and changed their conclusion that $\text{FeTe}_x\text{Se}_{1-x}$ at the Te end experiences the quantum topological transition.

We appreciate the referee for recognizing the updates made in our revised manuscript. Indeed, after a systematic de-contamination effort to minimize the Se contamination during FeTe growth, we were able to obtain better FeTe films and correspondingly clearer ARPES data to reveal the change of band topology. These data clearly show the absence of the TSS. The photon energy-dependent measurement further demonstrated that the new electron pocket around the Γ point for FeTe exhibits k_z dispersion and thus could only be a bulk band. This observation supports our conclusion that at the Te end, the system undergoes a topological transition from a non-trivial to a trivial phase.

The origin of the electron pocket around the Gamma point should be clearly resolved to support this argument. In the absence of the folded band from the expansion of the unit cell, there is only one electron band in the in-plane direction around the Gamma point, which is the $xy^{\{-}}$ band. Please check the following reference [V. Cvetkovic and O. Vafek, Phys. Rev. B 88, 134510 (2013)] for this information. One possibility is that the vertical streak is also from the $xy^{\{-}}$ band experience decoherence from electronic correlation and photon energy-dependent layer-to-layer contributions. This appearance of the $xy^{\{-}}$ bulk electron band could be a signature of the topological transition since the uppermost position of the $xy^{\{-}}$ band along the z direction is now positioned far below the Fermi level compared to the surface band energy position. However, the author has to clarify all these issues to make sure of their new argument of the quantum topological transition around the Te end.

We fully agree with the referee that identifying the orbital origin of the electron pocket at Γ is essential to supporting our argument of a topological trivial phase in 10 UC FeTe films. As shown in Fig.4 of the suggested reference (Ref.¹), the only electron band in the in-plane direction around the Γ point is indeed the xy band and this band is also above E_F . However, in the strongly correlated FeTe limit, the xy band shifts to a much lower

energy, resulting in a complete band inversion between the xy and the d_{xz} bands along $\Gamma - Z$. This scenario is also captured by our tight-binding model calculation in Fig. S12 (reproduced below), where band hybridization gives rise to a small electron pocket right above the “vertical streak” feature. This pocket is what we observed experimentally by ARPES and carries hybrid orbital characters ($xy + d_{xz} + d_{yz}$). Moreover, our DFT+eDMFT calculations also show that the small electron pocket is mainly of $d_{xz/yz}$ characters, consistent with our polarization-dependent ARPES measurements (see main text Lines 141-145). We also agree with the referee that the appearance of the bulk electron pocket is a signature of the topological transition. We would just like to point out that the *observed* electron pocket has hybrid orbital characters, due to the hybridization between the xy and the $d_{xz/yz}$ bands. This is precisely the scenario we illustrate in Fig. S24 (reproduced below).

To clarify this understanding, we have added the following statement in main text Lines 145-148, incorporating the suggested reference: “*Theoretically, in the topologically non-trivial phase the only electron pocket near Γ in the vicinity of E_F is the xy band above E_F [PRB 88, 134510 (2013); PRB 92, 115119 (2015); PRB 93, 115129 (2016)]. However, the observed electron pocket in the correlation-driven trivial phase is a hybridized band of both $d_{xz/yz}$ and xy orbital characters (Fig. S12 and S13c).*”

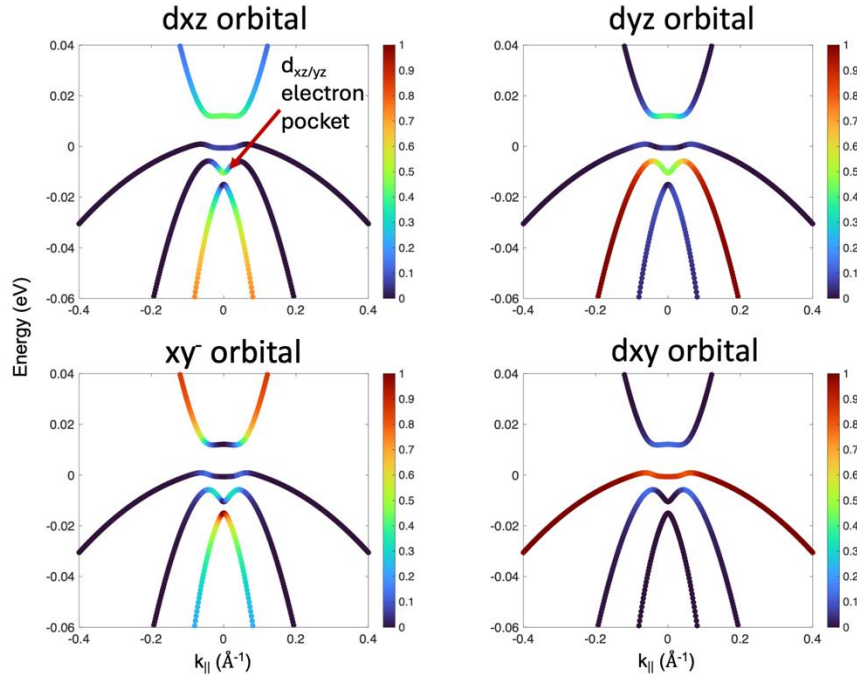


Fig. S12 Orbital projections of a tight-binding model for the complete band inversion between the xy and d_{xz} orbitals.

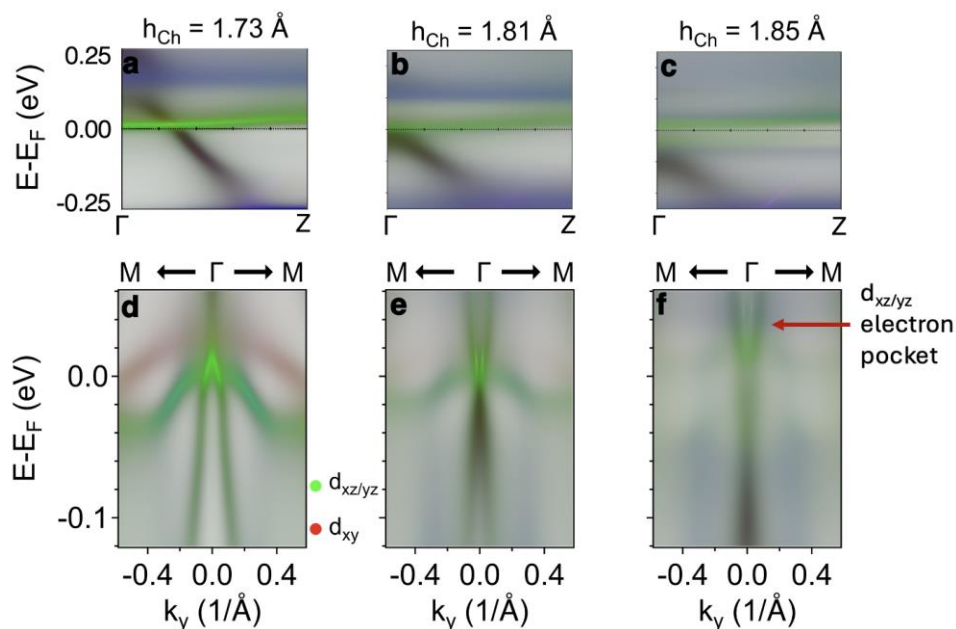


Fig. S24 **Tuning electronic correlations by adjusting the Te height.** (a-c) Orbital-resolved spectral functions calculated by DFT+eDMFT for FeTe as a function of different Te heights along the $\Gamma - Z$ direction. (d-f) Same as a-c, but along the $M - \Gamma - M$ direction.

Reviewer #3 (Remarks to the Author):

I recommend publication in Nature Comm.

[We thank the referee for the recommendation for publication.](#)

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The authors have made great efforts in their responses to the questions and addressed my concerns in the revised manuscript. I recommend the publication of this manuscript in Nature Communications.

[We appreciate the referee's comments and recommendation for publication.](#)

1. Cvetkovic, V. & Vafek, O. Space group symmetry, spin-orbit coupling, and the low-energy effective Hamiltonian for iron-based superconductors. *Phys. Rev. B* **88**, 134510 (2013).
2. Wu, X., Qin, S., Liang, Y., Fan, H. & Hu, J. Topological characters in $\text{FeTe}_{1-x}\text{Se}_x$ thin films. *Phys. Rev. B* **93**, 115129 (2016).

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I agree to publish this manuscript in the Nature Communications journal.

We thank the Referee #1's comment and the recommendation for publication. We agree that the band inversion between the xy band and the $d_{xz/yz}$ band at Gamma may indicate a finite quasiparticle weight of the d_{xy} orbital.