A weak topological insulator state in quasi-one-dimensional superconductor TaSe$_3$

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A well-established way to find novel Majorana particles in a solid-state system is to have superconductivity arising from the topological electronic structure. To this end, the heterostructures that consist of normal superconductor and topological material have been actively explored in the past decade. However, a search for the single material system that simultaneously exhibits intrinsic superconductivity and topological phase has been largely limited, although such a system is far more favorable especially for the quantum device applications. Here, we report the electronic structure study of a quasi-one-dimensional (q1D) superconductor TaSe$_3$. Our results of angle-resolved photoemission spectroscopy (ARPES) and first-principles calculation clearly show that TaSe$_3$ is a topological superconductor. The characteristic bulk inversion gap, in-gap state and its shape of non-Dirac dispersion concurrently point to the topologically nontrivial nature of this material. The further investigations of the $Z_2$ indices and the topologically distinctive surface band crossings disclose that it belongs to the weak topological insulator (WTI) class. Hereby, TaSe$_3$ becomes the first verified example of an intrinsic 1D topological superconductor. It hopefully provides a promising platform for future applications utilizing Majorana bound states localized at the end of 1D intrinsic topological superconductors.

Topological superconductors are recently attracting great research attention due to their fundamental importance and potential application to the quantum computation based on the Majorana bound states. A popular approach is to make use of superconducting proximity effect with which Cooper pairs are injected into the topological surface state as theoretically suggested by Fu and Kane. Following this idea, several heterostructures have been fabricated and indeed shown to host the Majorana quasiparticle. From the point of view of the application, however, the heterostructure systems can likely bring technical difficulties in the device fabrication. It is therefore highly desirable to have a material that intrinsically hosts topological superconductivity, although not many systems have been experimentally confirmed to be the case. One promising candidate is Fe(Se,Te) where superconductivity is encoded into the topological surface state on the two-dimensional (2D) surface. In this material, the topological superconductivity can certainly be intrinsic. However, the location of the Majorana state can be at the center of the vortex or anywhere on the edge of 2D surface; namely, it is not pre-determined locally.

In this regard, an important research direction, that has not been well explored so far, is to search for intrinsic topological superconductivity in one-dimensional (1D) systems. Different from the 2D or three-dimensional (3D) case, Majorana states exist at both ends of the 1D topological superconductor thus making its manipulation much easier. It would be useful for circuit device applications by securing the Majorana states at certain desired positions. Here we note a recent theoretical study suggesting that TaSe$_3$ is a strong topological insulator (STI). Also, the previous experiment seem to indicate the unusual superconductivity in this material by showing the absence of a diamagnetic response below the critical temperature $T_c$. To be the first confirmed example of 1D intrinsic topological superconductivity, however, further investigations of its topological properties are strongly requested from both experimental and theoretical sides.

In this Communication, we report our detailed study of the electronic structure of TaSe$_3$ by using angle-resolved photoemission spectroscopy (ARPES) and first-principles density functional theory (DFT) calculation. The band inversion by spin-orbit coupling (SOC) and the additional in-gap surface state with non-Dirac dispersion clearly indicate the nontrivial topology of its band structure. Further analyses of $Z_2$ indices and the number of Dirac points depending on the surface type clearly show that TaSe$_3$ is a weak topological insulator (WTI). Our results establish TaSe$_3$ as the first experimentally verified example of an intrinsic 1D topological superconductor.

**Results**

Crystal and electronic structure. TaSe$_3$ consists of elongated prismatic chains with monoclinic P2$_1$/m space group symmetry (Fig. 1a), which gives rise to the quasi-one-dimensional (q1D) electronic properties. The chains are stacked via van-der-Waals interaction and the weak potential modulation along with the chain-perpendicular direction results in the finite band dis-
FIG. 1. Crystal structure and calculated bulk band structure of TaSe$_3$. a, Crystal structure of TaSe$_3$. Ta and Se atoms are depicted by yellow and violet spheres, respectively. The black solid parallelogram and blue dashed line shows the unit cell and the practical cleavage plane (101). b, 3D and surface-projected BZs. The planes colored by blue and yellow represent the surface BZs of (101) and (010) planes, respectively. $k_\parallel$ and $k_\perp$ indicate projected reciprocal lattice vectors on the (101) surface BZ. c, Calculated bulk band structure (left) and projected density of states (PDOS, right). The projected weights of Ta-5d and Se-4p orbitals are presented in red and blue colors, respectively. The band inversions are clearly observed around the high-symmetry points of B, Z, and D. For the calculated band structure without SOC, see Supplementary Fig. 2.

ARPES electronic structure and an in-gap state.
In order to examine the electronic structure and its topological nature, we carried out ARPES measurements on high-quality single crystals of TaSe$_3$. Figure 2 presents the ARPES spectra measured on the cleaved (101) surface. A constant energy contour plot at Fermi level ($E_F$) (Fig. 2a) shows an elongated Fermi surface along $k_\perp$, reflecting the overall 1D character of this material ($x$- and $y$-axis corresponds to $k_\parallel$ and $k_\perp$ direction in Fig. 1b, respectively). The band dispersions were measured along two projected high-symmetry lines, namely, $\Gamma$–$\Upsilon$ (cut 1) and $\Xi$–$\Sigma$ (cut 2). These two lines are particularly interesting because the characteristic band inversions are observed (see Fig. 1b, c) and the Dirac cone, a hallmark of a topological insulator, has been predicted by the previous calculation[13].

In taking the ARPES spectra, we utilized both linear vertically (LV) and horizontally (LH) polarized incident light to avoid missing of the spectral weight caused by matrix element effect. Note that each spectrum taken with different polarizations highlights different bands and is therefore complementary to each other. For instance, two hole bands with different band-top energies near $E_F$ are well identified in the cut 1 taken with LV (Fig. 2b) while only the lower one is seen by LH (Fig. 2c). Along cut 2, LV clearly reveals a hole band near $E_F$ (Fig. 2d) whereas LH proves the relatively large electron-like band together with additional two tiny electron bands which barely cross the $E_F$ (Fig. 2e). To analyze the complicated band dispersion of TaSe$_3$ more clearly, the curvature method[17] is applied (Figs. 2f–i) which enables us to make more direct comparisons with calculation results.

As shown in Fig. 2b–i, the overall band structure of ARPES spectra is in good agreement with our theoretical result. The calculated bulk bands along $\Gamma$–$\Upsilon$ (Z–C)
The spectrum of this band is large enough in ARPES but not seen in DFT result. Understanding the discrepancy between theory and experiment, particularly for the case of this second band, is of key importance because it locates in the band inversion gap and thus is likely to be involved with the topological nature of this material. In the below, we will show that this 'in-gap state' has in fact the surface origin and its dispersion feature can be regarded as an indication of WTI state rather than STI\cite{13}.

**Surface electronic structure and band topology: DFT calculation.** To further elucidate the nature of band dispersion including the in-gap state, we performed the slab calculation by using iterative Green’s function method\cite{19,20} based on maximally localized Wannier functions (MLWFs)\cite{11,19} generated from DFT bands (see Methods for more details). In Fig. 3, the results are presented and compared with experimental spectra where two ARPES spectra taken from both polarizations are added up. Besides the overall good agreement in between calculation and experiment, we first take a special note on the broad continuum states (indicated by white arrows in Figs. 3a, e). This continuum is well identified also in the calculation being attributed to the surface projection of bulk states at a range of $k_z$ values (see Fig. 1b). In ARPES measurement, on the other hand, the broadening of photoelectron momentum along $k_z$ direction is due to the finite probing depth of incident light in real space.
Therefore, this consistency between theory and experiment confirms that our slab calculation sufficiently well describes the states near the surface.

Interestingly, Fig. 3 clearly shows that the two aforementioned bands (which are present in ARPES but not observed in the bulk calculation) are originated from surface states. For clearer comparisons, see Fig. 3b and 3f for the case of hole-like (in cut 1; $\bar{\Gamma}$–$\bar{Y}$ line) and electron-like band (in cut 2; $\bar{X}$–$\bar{S}$ line), respectively, as denoted by the black-dashed lines and the red arrows. From the fact that these bands are only reproduced by surface band calculations (and not found in 3D bulk calculations), we conclude that they are the surface states. Another fact that these bands are well separated from the nearby bulk spectra also supports this conclusion.

From the point of view of band topology, important is the unambiguous identification of the second band (i.e., the electron-like band indicated by ‘SS’ in Figs. 3f, h) as being originated from the surface. As is well known, a hallmark of topological insulator is the characteristic surface state with Dirac crossing, which is located inside the inversion gap $g_{\text{inv}}$. The absence of Dirac cone at (10$\bar{1}$) surface leads us to conclude that TaSe$_3$ is not STI contrary to the previous theoretical suggestion[13], and to require further investigation regarding its topological nature.

Discussion

$Z_2$ topological indices and weak topological insulator. According to Fu et al[22], STI has surface Dirac cones on its surface. On the other hand, WTI has Dirac surface state only on a certain type of surfaces. The other type of surface is called topologically ‘dark’. Here it is important to note that the cleaving surface of TaSe$_3$ is in fact a dark surface. Therefore, our observation of no Dirac point in ARPES spectra does not exclude the intriguing possibility for TaSe$_3$ to be the first confirmed example of 1D topological superconductor. In order to examine the possibility of WTI, we calculated $Z_2$ topological indices, and found that TaSe$_3$ is indeed a WTI: $(\nu_0; \nu_1, \nu_2, \nu_3) = (0; 101)$. Figure 4a shows the products of the occupied band parities at the eight TRIM points from which $(\nu_0; \nu_1, \nu_2, \nu_3)$ are defined[22]. As is well established, $\nu_0 = 0$ and $\nu_{1,3} \neq 0$ are a clear signature of WTI[22].

In order to further confirm the WTI nature of TaSe$_3$, we explored the other surfaces than (10$\bar{1}$), namely the ‘non-dark’ surfaces on which the surface Dirac cones are expected to appear. The dark surface of WTI is defined by the Miller indices of the given surfaces. That is, the surfaces defined by $(\nu_1, \nu_2, \nu_3)$ and $(\nu_1, \nu_2, \nu_3) + 2\bar{G}$ ($\bar{G}$: the reciprocal lattice vector) are topologically dark[22]. As is already mentioned, our cleaving surface is therefore a
FIG. 4. The calculated $Z_2$ topological invariants and the surface Dirac points. a, The products of band parities for all occupied bands ($\delta$) at eight bulk TRIM points. The red and blue dots represent $\delta = +1$ and $-1$, respectively. b, Schematic picture for the TaSe$_3$ surfaces and their topological nature. According to the $Z_2$ classification, the (101) surface should be topologically dark while two other side surfaces, (010) and (100), are expected to have two Dirac points on each plane and to host topologically protected helical surface states (depicted by blue and red arrow). c, The calculated (010) surface band structure. The inset shows the surface-projected BZ and the relevant high-symmetry points. Two Dirac points are found as expected at B and Z points. The topological surface states are indicated by the black (connecting to the conduction band) and the green dashed lines (connecting to the valence band) with arrows. The color bar represents the intensity strength. d The same as in (c), for the (101) surface. As expected, no Dirac cone is found.

dark surface, and no Dirac cone found in our ARPES data is consistent with the WTI nature. The calculation result of the other type (010) surface is presented in Fig. 4c, and two Dirac points are clearly noticed; see the points indicated by ‘DP’. The dashed lines with arrows demonstrate that both Dirac points are indeed the points made by the crossings of the surface bands connecting valence and conduction bands, and the band inversion changes the band parity. For another allowed (i.e., non-dark) surface of (100), we found Dirac points as well; see Supplementary Fig. 6. While some Dirac points are embedded in the bulk bands, the Dirac nature of their crossings can always be checked straightforwardly. Hereby, we establish that TaSe$_3$ has a WTI nature.

It should be noted that the experimental verification of WTI nature can be challenging due to the two distinctive types of topological surfaces and the limited capability of choosing cleaving surface. Further, additional subtleties are also involved in the computation side arising from the notorious issue of exchange-correlation functionals. Our effort of resolving this issue for the current case of TaSe$_3$ can be found in Supplementary Information. To the best of our knowledge, the only example of the experimentally reported 1D WTI is $\beta$-Bi$_2$I$_4$ and its WTI nature has still been debated until very recently. Our case of TaSe$_3$ is the second, if not the first, example of 1D WTI, and it is the first example of 1D topological superconductor.

In summary, we provide the convincing evidence of WTI nature in TaSe$_3$ by means of ARPES experiments and first-principles calculations. Topological features of band inversion and surface in-gap state are clearly identified. The calculated $Z_2$ indices as well as the absence and the presence of Dirac cones respectively on the dark and the non-dark surfaces confirm our conclusion of WTI nature. With this, this material is suggested to be the first experimentally confirmed example of 1D topological superconductor in an intrinsic as-is form. Unveiling the relation between the superconductivity and topology can also be an exciting future direction.

Methods

Sample growth and characterization. Single crystals of TaSe$_3$ were grown via the chemical vapor transport (CVT) method. Ta (99.99%) and Se (99.999%) powder of molar ratio 1 : 3.3 were loaded into one end-side (source zone) of the quartz tube. The additional amount of Se is intended to prevent Se deficiency, and to work as a transport agent. Then the quartz tube was evacuated, sealed and loaded into the two-zone furnace with the temperature of the zones set by 720°C and...
680°C, and maintained for 14 days. Whisker-like single crystals of typical dimensions of 10×1.5×0.05 mm³ were achieved. The structural and electrical properties of TaSe₃ single crystals were characterized using energy dispersive spectroscopy (EDS), X-ray diffraction (XRD), and electrical resistivity measurement (see Supplementary Fig. 1).

**ARPES measurements.** ARPES measurements were performed at beamline 5-4 of Stanford Synchrotron Radiation Lightsource (SSRL), SLAC National Laboratory and beamline 4.0.3 of Advanced Light Source (ALS), Lawrence Berkeley National Laboratory. ARPES spectra were acquired with Scienta R4000 (R8000) electron analyzer at SSRL (ALS). A clean surface of the sample was obtained by in-situ cleaving of single crystals of TaSe₃. Samples were cleaved to reveal the natural cleavage plane of (10¯1) at 10K under ultra-high vacuum pressure better than 5×10⁻¹¹ torr. Measurements were carried out maintaining sample temperature at 10K at both beamlines. At SSRL, linearly polarized light of photon energy \( h\nu = 20 \text{ eV} \) was used for the measurements. At ALS, linearly polarized light of photon energy \( h\nu = 56 \text{ eV} \) was used. The total energy resolution and angular resolution was set to be better than 10 meV and 0.3°, respectively, for the measurements at both beamlines.

**DFT band structure calculations.** Our DFT calculations were performed with ‘Vienna Ab initio Simulation Package (VASP)’ based on the projector augmented-wave pseudopotential³⁰,³¹. We used the experimental lattice parameters³² and the internal atomic coordinates were optimized with a force criterion of 0.001 eV/Å. The 7 × 15 × 7 k-points and the energy cutoff of 600 eV were adopted. SOC was also taken into account. All of the presented results were obtained within so-called ‘PBE’ generalized gradient approximation (GGA) for exchange-correlational functional.³³ Our main results were double-checked by using GGA-PBEsol³⁴ and local density approximation (LDA) as parameterized by Perdew and Zunger³⁵,³⁶. We also double-checked the results by using all-electron full-potential code, Wien2k³⁷ for which \( R_{\text{mt}}K_{\text{max}} = 7 \) and 7 × 19 × 6 k-points were adopted in the irreducible Brillouin zone. The augmented planewave sphere radii of Ta and Se were 2.48 and 2.36 a.u., respectively. We used ‘Wannier90’³⁸ code to extract MLWFs for Ta \( d \) and Se \( p \) orbitals,³⁹ and ‘WannierTools’ code to analyze the topological property and surface state. The parities at TRIM points were calculated directly from DFT wave-functions by using ‘vasp2trace’. For comparison with ARPES data, the \( E_F \) was shifted by 39 meV in the calculated band structure for both bulk and slab case. As reported in previous studies,³⁹ it likely reflects the presence of a certain amount of defects in crystals.

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**Author contributions** Y.K. and M.J.H. conceived the work. J.H. synthesized and characterized the TaSe₃ single crystals. J.H., S.K., Y.L., C.L., J.C., G.L., Y.A., and Y.K. performed the ARPES measurements with support from M.H., D.L. and J.D.D. J.H., S.K. and Y.K. analyzed the ARPES data. M.Y.J., M.-C.J. and M.J.H. carried out theoretical calculations. All authors discussed the results. J.H., M.Y.J., M.J.H. and Y.K. wrote the manuscript with contribution from all authors.
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