Observation of topological superconductivity in a stoichiometric transition metal dichalcogenide 2M-WS$_2$

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Topological superconductors (TSCs) are unconventional superconductors with bulk superconducting gap and in-gap Majorana states on the boundary that may be used as topological qubits for quantum computation. Despite their importance in both fundamental research and applications, natural TSCs are very rare. Here, combining state of the art synchrotron and laser-based angle-resolved photoemission spectroscopy, we investigated a stoichiometric transition metal dichalcogenide (TMD), 2M-WS$_2$ with a superconducting transition temperature of 8.8 K (the highest among all TMDs in the natural form up to date) and observed distinctive topological surface states (TSSs). Furthermore, in the superconducting state, we found that the TSSs acquired a nodeless superconducting gap with similar magnitude as that of the bulk states. These discoveries not only evidence 2M-WS$_2$ as an intrinsic TSC without the need of sensitive composition tuning or sophisticated heterostructures fabrication, but also provide an ideal platform for device applications thanks to its van der Waals layered structure.

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Superconductors and topological matter are two classes of quantum materials with fascinating physical properties and application potentials, and their combination may further give rise to a unique quantum phase, the topological superconductor (TSC). A TSC can host exotic emergent particles such as the Majorana fermion, a particle of its own anti-particle that can not only show remarkable phenomena such as thermal quantum Hall effect, but also serve as a key ingredient for the realization of topological quantum computation—a promising approach to realize the fault-tolerant quantum computation.

However, TSCs are rare in nature. Up to date, intrinsic TSC candidates are only found in a few materials, which are either controversial (e.g., Sr$_2$RuO$_4$, FeTe) or nonstoichiometric compounds (e.g., Cu$_x$Bi$_2$Se$_3$, FeTeSe, Li(Fe,Co)As) that require fine tuning in composition and inevitably consist of defects that are unfavorable for device applications. On the other hand, artificial structures combining conventional superconductors and nondegenerate spin states (e.g., topological insulators, quantum anomalous hall insulators, semiconductors with strong spin-orbit coupling, or ferromagnetic thin film and atomic chains) have been explored, but the need to construct sophisticated heterostructures and the requirement of long superconducting coherence length make this approach material selective.

Recently, a stoichiometric transition metal dichalcogenide (TMD), 2M-WS$_2$ was proposed as an intrinsic TSC with the highest intrinsic superconducting transition temperature ($T_C$ = 8.8 K) among all TMDs in the natural form. The stoichiometry makes the synthesis of high-quality crystals possible, and the layered structure with van der Waals coupling makes it ideal for device fabrication for potential applications.

The superconductivity of 2M-WS$_2$ was recently confirmed by transport and scanning tunneling microscopy/spectroscopy (STM/STS) measurements, and zero energy peaks in the STS spectra were observed in magnetic vortex cores, suggesting the possible existence of Majorana bound states. However, compelling evidence for the topological electronic structure, including the direct observation of topological surface states (TSSs), the superconducting gap of the TSSs and its temperature evolution, are yet to be found.

In this work, combining the state of the art synchrotron- and laser-based angle-resolved photoemission spectroscopy (ARPES), we not only systematically investigated the band structure of 2M-WS$_2$ across the full three-dimensional Brillouin zone, but also successfully observed the TSSs thanks to the large photon energy range of the synchrotron light source. Furthermore, by carrying out detailed temperature-dependent measurements, the superconducting gap from the TSSs (as well as the bulk states, BSs) was clearly observed below $T_C$, establishing the TSC nature of 2M-WS$_2$.

The mechanism of the topological superconductivity in 2M-WS$_2$ can be briefly shown in Fig. 1a. Above the superconducting transition ($T > T_C$), 2M-WS$_2$ is a bulk topological semimetal...
(TSM) with nontrivial TSSs; while in the superconducting state \((T < T_c)\), the bulk s-wave superconductivity can induce superconductivity to the TSSs by internal proximity effect, thus realizing a topologically nontrivial superconducting state of spin-helical electrons on the surface, as illustrated in Fig. 1a. In addition, if a magnetic domain is formed, the domain boundary (green line in the right panel of Fig. 1a) can host the MBS, which, if bound with a magnetic flux quantum, obey non-abelian statistics\(^\text{1-4,6}\) and can be used to form the quantum-bit for logical quantum computation\(^1\).

**Results**

**Sample characterization.** Like many TMD compounds, 2M-WS\(_2\) is a van der Waals layered crystal (space group \(C_{2/m}\) No. 12). As shown in Fig. 1b, within each WS\(_2\) layer, the distorted octahedral structure forms quasi-1D chains of W and S atoms along \(Y\) direction (see Fig. 1b); and the adjacent WS\(_2\) layers are offset to each other along \(X\)- and \(Y\)-directions by \(\Delta X = 2.86\ \text{Å} \) and \(\Delta Y = 1.62\ \text{Å}\), respectively (Fig. 1b, and more details on the crystal structure are shown in Supplementary Fig. 1). The high quality of the crystals used in this work was confirmed by X-ray diffraction (Supplementary Fig. 2), the large residual-resistivity ratio (RRR = 152, Fig. 1c, right inset), and the sharp superconducting transition \((\Delta T = 0.2\ \text{K}, \text{Fig. 1c})\). In fact, the \(T_c\) (8.8 K) of 2M-WS\(_2\) is the highest among all intrinsic TMD compounds under ambient conditions up to date\(^{26}\).

The nontrivial topological electronic structure of 2M-WS\(_2\) was theoretically proposed in a recent work\(^{26,29}\) and is confirmed by our ab initio calculations: 2M-WS\(_2\) is a TSM in its normal state with the TSSs located at the center of the surface BZ (sketched in the left panel of Fig. 1d); in the superconducting state (right panel of Fig. 1d), both the BSs and TSSs are expected to be gapped (as observed in our measurements which will be discussed in details later).

Due to the weak van der Waals interaction between WS\(_2\) layers, the cleaved surfaces for the ARPES study in this work are of high quality. As illustrated in Fig. 1e, a topography map from our STM measurement (left panel of Fig. 1e) reveals a defect-free region of the topmost S-atom layer with clear quasi-1D chains, in excellent agreement with the ab initio simulation (right panel of Fig. 1e and more details in Supplementary Fig. 3).

**General electronic structure of 2M-WS\(_2\).** To investigate the overall electronic structure of 2M-WS\(_2\), we first carried out synchrotron-based ARPES measurements, and Fig. 2b shows a typical set of experimental band structure, containing rich details that can be successfully reproduced by ab initio calculations (see Fig. 2c and Supplementary Fig. 4 for more details). Thanks to the
Fig. 3 Band structure of the topological surface states. a Comparison between slab calculation (left) and photoemission (right) band dispersions along $\bar{\Gamma} - \bar{\Gamma} - \bar{\Gamma}$ direction. The TSSs are highlighted by red color (left) and indicated by the red dashed lines (right). The bulk dispersions around $\bar{\Gamma}$ point (red shaded area, $k_y$ integration window is 0.06 $\AA^{-1}$) are overlapped to better illustrate the TSSs (right). b Bottom left: Zoomed in dispersion of the TSS in the pristine sample covering the yellow box area in the right panel of a. The red dashed lines show the TSS dispersions. Top left: Schematic of the calculated dispersions for comparison and the $E_F$ is marked as a black dashed line. Bottom right: Full dispersion of the TSS band observed after in situ potassium (K) doping (illustrated as the middle inset). Top right: Calculated dispersions with the new $E_F$ position indicated after the K-dosing, showing a clear upshift $\Delta E_F$. c Side-by-side comparison of TSS dispersions before (left panel) and after (right panel) K-dosing, shows a $\Delta E_F$ of ~45 meV. d Stacking plots of the experimental constant energy contours (CECs) at different energy close to the Dirac point, showing rapid evolution. e, f Comparison between experimental (e) and ab initio calculated (f) CECs evolution near the Dirac point showing overall agreement. For clarity, the CECs of the TSSs are highlighted by red color. In e, the faded red dashed lines represent the less visible TSSs due to the hybridization to the bulk states.

Wide photon energy range of the synchrotron light source, we were able to do detailed photon-energy-dependent measurements to examine the electronic structures across multiple 3D BZs along the $k_x$ direction, as illustrated in Fig. 2d (see Supplementary Fig. 5). With the $k_z$ electronic structures identified, we can study band dispersions at different $k_y$ momenta using specific photon energies. As two examples, the dispersions cut across the bulk $\Gamma$ ($k_x = 0$) and $Z$ ($k_x = \pi/a$, where $a$ is the out-of-plane lattice constant) points can be accessed by 30 and 44 eV photons, respectively (see Fig. 2d); and the comparison between experiments and ab initio calculations (Fig. 2e, f) shows nice overall consistency (see Supplementary Fig. 6).

Topological surface states of 2M-WS$_2$. According to our calculation and the previous theory work$^{26}$, the TSSs in 2M-WS$_2$ reside only in a close vicinity around the center ($\bar{\Gamma}$ point) of the surface BZ, thus very high energy and momentum resolution is needed to resolve the TSS band structures. Therefore, we adopted the laser-based ARPES ($h\nu = 6.994$ eV, $\Delta E = 0.9$ meV and $\Delta k = 0.003$ $\AA^{-1}$) in the search for TSSs.

Indeed, as presented in Fig. 3a, the TSSs near the $\bar{\Gamma}$ in the calculation (left panel) were clearly seen in measurement (right panel). However, in the pristine sample, the upper part of the TSSs was above the Fermi-level ($E_F$) and could not be seen experimentally, and slight n-type doping was needed to lift the $E_F$ to reveal the whole TSS dispersions. As expected, after in situ potassium doping, we successfully raised the $E_F$ by ~45 meV and the whole TSS dispersions could be observed (Fig. 3b, c), in excellent agreement with the calculation. Besides the dispersions, the existence of the TSSs can also be seen in the constant energy contours of the band structures (Fig. 3d–f), again in good agreement with the calculations.

Superconducting gap of 2M-WS$_2$. Having identified the TSSs, we then investigate the superconducting state properties of the TSSs and BSs. The superconducting gaps of both the TSSs and BSs from a pristine sample are clearly seen in the left panel of Fig. 4a (spectra were symmetrized with respect to $E_F$ to get rid of the Fermi-Dirac function cutoff), which disappear when $T > T_C$ (right panel of Fig. 4a). The evolution of the superconducting gap...
with temperature can be seen in the energy distribution curves (EDCs, see Fig. 4b) or directly from the spectra plot (Fig. 4c). The superconducting gaps from both the TSSs and BSs are consistent with a mean field temperature dependence, as indicated by the black dashed line. Horizontal error bars represent the temperature variance during the measurements. Vertical error bars represent the confidence intervals of the fitting under 95% confidence level. E constant energy contour plot (intensity integrated between $E_F$ and $E = 10$ meV) shows the loci of the TSS and BS ($T = 6$ K). $\theta$ defined the angle used in f, g, f EDCs from the loci of different $\theta$ angles at the superconducting state ($T = 6$ K) show that the superconducting gap magnitude of both TSS (left) and BS (right) are nearly isotropic. g Plot of the superconducting gap size as a function of $\theta$ angle for both TSS (red symbols) and BS (blue symbols), showing similar magnitude. Error bars represent the energy resolution determined by ARPES spectra on multi-crystalline gold.

Discussion

Our unambiguous observation of superconducting gap developed on the TSSs of 2M-WS$_2$ provide the key ingredient for TSC based...
on the Fu-Kane proposal. The helical spin texture of the TSS is confirmed by previous first principle calculation and should survive possible surfacebulk hybridization since it is well separated from the BS in both energy and momentum space. Moreover, spin polarization persists in the TSSs of n-doped topological insulators, topological metals, and even the topological surface resonance states of Dirac semimetals, which exhibits robustness of spin texture in TSSs against surfacebulk hybridization. Therefore, the spin-momentum locked surface states could result in effective p-wave superconductivity in 2M-WS2, namely a TSC state.

As shown in Fig. 3a–c, 2M-WS2 hosts a Rashba-like TSS, which is a reminiscence of semiconductor–superconductor TSC scheme. As the upper and lower branches of the TSS must eventually merge in the spin unpolarized BSs, the Ep position is critical: (1) when only one pair of spin-polarized bands crosses Ep, 2M-WS2 falls to the superconducting topological insulator scenario, as proposed in Cu6Bi4Se11; (2) when two pairs of spin-polarized bands cross Ep, 2M-WS2 falls to the strong spin–orbit coupling semiconductor–superconductor scenario, as proposed in Al-InAs superlattice; in the latter situation, the topology of the system can be tuned by the Zeeman coupling Vg, which is due to the external magnetic field as shown in Fig. 1a, chemical potential μ, and the self-proximity-induced pairing gap Δ2214. The compatibility and versatility of the TSS configuration in 2M-WS2 with the appearance of Majorana states require further investigation.

The formation of the TSC state in stoichiometric 2M-WS2, together with its layered structure with van der Waals coupling, makes it ideal for device fabrication and thus a promising platform to explore the phenomena of MBSs and their application in topological quantum computation. Moreover, the discovery of the TSC state in 2M-WS2 further enriches the interesting phenomena hosted by TSSs (e.g., gate and pressure-tunable superconductivity, charge density waves, Mott insulators, gyrotropic electronic orders, moiré-trapped valley excitons, type-II Dirac and Weyl semimetals, etc.), thus enabling the study of these properties and their interplay, as well as the design of novel devices for new applications. We note that, after the acceptance of this manuscript, self-proximity induced TSC was also theoretically proposed in RRRu2B2 (R=Y, Lu).
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Author contributions

Y.L.C. conceived the experiments. Y.W.L. and H.J. Zheng carried out ARPES measurements with the assistance of Z.K.L., L.X.Y., Y.I.C., C.Chen, A.J.L., D.P., L.X.X., D.H.L., M.H., A.B., S.W.J., C.Cacho, and Y.H.; Y.W.L. and H.J. Zheng performed the data analysis on the ARPES results. Y.Q.F., J.P., and F.Q.H. synthesized and characterized the bulk single crystals. D.Q.Z., W.I.S., and H.J.Zhang performed ab initio calculations. H.J.Zheng, S.L., and M.X.W. carried out STM measurements. Y.W.L. wrote the first draft of the paper. Y.L.C., Z.K.L., L.X.Y., H.J.Zhang, Y.H., and L.F. contributed to the revision of the manuscript. All authors contributed to the scientific planning and discussions.

Competing interests

The authors declare no competing interests.

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