Dual topological superconducting states in the layered titanium-based oxypnictide superconductor BaTi$_2$Sb$_2$O

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Topological superconductors have long been predicted to host Majorana zero modes which obey non-Abelian statistics and have potential for realizing non-decoherence topological quantum computation. However, material realization of topological superconductors is still a challenge in condensed matter physics. Utilizing high-resolution angle-resolved photoemission spectroscopy and first-principles calculations, we predict and then unveil the coexistence of topological Dirac semimetal and topological insulator states in the vicinity of Fermi energy ($E_F$) in the titanium-based oxypnictide superconductor BaTi$_2$Sb$_2$O. Further spin-resolved measurements confirm its spin-helical surface states around $E_F$, which are topologically protected and give an opportunity for realization of Majorana zero modes and Majorana flat bands in one material. Hosting dual topological superconducting states, the intrinsic superconductor BaTi$_2$Sb$_2$O is expected to be a promising platform for further investigation of topological superconductivity.

Topological superconductors have attracted tremendous interest for harboring Majorana bound states on their boundaries [1–3]. The non-Abelian Majorana zero modes in the vortex of topological superconductors are potential for topological quantum computations without decoherence [4–6]. To date, several systems have been predicted to host topological superconductivity (TSC). Examples include intrinsic odd-parity superconductors [7,10] and those heterostructures constructed by proximity coupling of topological insulators (TIs) to conventional $s$-wave superconductors [11–13], which inspired enormous experimental efforts to the exploration of Majorana fermions [14–34]. However, experimentally, the presence of Majorana zero modes in some of these systems is still in hot debate. For example, pairing symmetries of several proposed $p$-wave superconductors, such as Sr$_2$RuO$_4$ [14,15] and Cu$_4$Bi$_2$Se$_3$ [16,20], are yet inconclusive. On the other side, although signs of Majorana bound states have been reported in conventional $s$-wave superconductors with the proximity to TIs, difficulties in fabricating such these heterostructures and the disturbance of interface physics would inevitably prohibit their further studies [23–34].

The recent discovery of TI states in the iron-based superconductor Fe(Se,Te), combining nontrivial topological states and superconductivity in a single material, pointed out a new dimension in realizing Majorana bound states [35–41]. One later research on Li(Fe,Co)As revealed that iron-based superconductors might generically host multiple types of non-trivial topological states, e.g., both the TI and topological Dirac semimetal (TDS), together with unconventional superconductivity [42]. Thus, the possible intrinsic TSC therein, which takes advantage of the proximity effect in the momentum space, would overcome disadvantages in other implementations of TSC. However, the Dirac point (DP) from the TDS states of superconducting Li(Fe,Co)As with 3% Co is located above the Fermi level ($E_F$), and thus the proposed one-dimensional Majorana fermion would not be expected to dominate the low-energy electronic structure [42]. Meanwhile, seemingly the superconductivity would be inexorably suppressed by introducing further charge carriers in Li(Fe,Co)As [43,44]. In this regard, it is critical to search for more practical superconductors with both Dirac cones from TI and TDS states below $E_F$ to realize multiple topological superconducting states in one material.

In this Letter, we have identified both TI and TDS states reminiscent of those in iron-based superconductors but with Dirac cones below $E_F$ in superconducting BaTi$_2$Sb$_2$O samples using angle-resolved photoemission spectroscopy (ARPES) and first-principles calculations. Furthermore, spin-resolved
FIG. 1. (a) Crystal structure of layered BaTi$_2$Sb$_2$O. (b) Three dimensional bulk Brillouin zones (black) with (001) surface Brillouin zone (yellow) projected. High symmetry points are marked. (c) Core-level photoemission spectrum shows characteristic Ba, Ti and Sb peaks. Inset: Image of high quality BaTi$_2$Sb$_2$O single crystal. (d)(i) and (ii) Comparison between the experimental and bulk calculated constant-energy surface. (i) is taken at 84 eV photons integrated over the energy window of $|E_F-25$ eV, $E_F+25$ eV]. (e) Temperature dependence of resistivity of BaTi$_2$Sb$_2$O. (f) Calculated band structure in the presence of SOC. The enlarged image of the light red rectangle announces the TDS and TI region in the white box. Two surface states marked as SS1 and SS2 are part of the Dirac cone from TDS and TI, respectively.

ARPES measurements confirmed both of the predicted spin-helical surface bands originated from TDS and TI states, which are prospective to harbour Majorana zero modes and Majorana flat bands in one single material. This titanium-based oxypnictide superconductor which has the similar multiple topological states as iron-based superconductors would provide another parallel but more practical playground for comparative study on TSC.

Details on the sample growth method, first-principles calculation and ARPES measurement and can be found in Note 1 to 3 in Supplemental Material (SM) [45]. The crystal structure of BaTi$_2$Sb$_2$O is illustrated in Fig. 1(a). It is composed of [Ti$_2$Sb$_2$O]$_6^{2+}$ octahedron layers, which are stacked with Ba atoms along the c axis. Thus, the natural cleavage plane should be parallel to the a-b plane and between two neighbouring [Ti$_2$Sb$_2$O]$_6^{2+}$ layers. The bulk and (001)-projected surface Brillouin zones (BZs) of BaTi$_2$Sb$_2$O are shown in Fig. 1(b). Fig. 1(c) displays the angle-integrated photoemission spectrum of BaTi$_2$Sb$_2$O over a large range of binding energy, in which we can clearly identify the Ba (4p and 4d), Ti (3p) and Sb (4d) core levels, confirming the element composition of our samples. After cleaved in the air, the sample shows typical flat and shining surface as illustrated in the inset of Fig. 1(c). Besides, we present the Fermi surface (FS) map obtained by ARPES in Fig. 1(d)(i), which clearly suggests the square BZ with four-fold symmetry and is in remarkable agreement with the calculation [Fig. 1(d)(ii)], further proving its tetragonal crystal structure and (001) cleavage plane. In addition, the sample exhibits a metallic temperature dependence and shows zero resistance below $T_c=1.05$ K [the inset of Fig. 1(e)], in accordance with previous reports [46].

Fig. 1(f) declares the calculated band structure of BaTi$_2$Sb$_2$O with the spin-orbit coupling (SOC). We concentrate on crossings of several bands around $E_F$, which are highlighted by the light red rectangle with the enlarged image in the right panel. These three bands belong to irreducible representations $\Gamma_6$, $\Gamma_7$, and $\Gamma_7^-$, respectively. The crossing of $\Gamma_6^-$ and $\Gamma_7^-$ at an arbitrary $k$ along $\Gamma-Z$ features a symmetry-protected DP. Slightly lower in energy, $\Gamma_7^-$ and $\Gamma_7^-$ cross, leading to a small gap. The distinct behavior of these two band crossings stems from different mechanisms. BaTi$_2$Sb$_2$O belongs to the space group no. 123, which respects the inversion symmetry $\hat{I}$. The joint operation of time-reversal ($\hat{T}$) and $\hat{I}$ promises the Kramer’s double degeneracy everywhere in the BZ. In this case, $\epsilon\hat{z}$, which leaves $k_z$ invariant along $\Gamma-Z$, protects a stable DP between $\Gamma_6^-$ and $\Gamma_7^-$ bands. However, as $\Gamma_7^-$ and $\Gamma_7^-$ share the same basis functions with the only difference on their response to $\hat{I}$, the two bands will unavoidably open a gap when they cross. Such particular alignment of bands is essential for BaTi$_2$Sb$_2$O to resemble the electronic structure of iron-based superconductors which features a great potential to coexist two distinct types of topological superconductivity in one system, i.e., the one-dimensional Majorana fermion from topological DPs and Majorana zero modes from the topological insulator or DPs [42, 43]. To better understand the different symmetry-protection and gap opening mechanisms, we compose a $k \cdot p$ model around the DP and the gap below with $\epsilon\hat{z}$ and $\hat{T}\hat{I}$, see Note 4 in SM for more details [45].

The above two topological nontrivial states can be further displayed in the surface states calculation along $\hat{T}-\hat{M}$. Here, two bulk bands near $\hat{F}$ are marked as BB1 and BB2, respectively [Fig. 1(g)]. Moreover, by zooming in the region of the
We note that these surface states are both below $E_F$, which can be assigned as SS1 and SS2, respectively. Although these two DPs are as close as $-23$ meV and their overlapped states have gradually merged into bulk states, we could still resolve the rest parts of these surface states, which can be assigned as SS1 and SS2, respectively. We note that these surface states are both below $E_F$ and thus can be probed by ARPES without further carrier doping.

Experimentally, we first revealed both the TDS and TI bulk states of BaTi$_2$Sb$_2$O. Due to considerable entanglement between the bulk and surface states around $E_F$ [Fig. 1(g)], we took advantage of the matrix element effect to distinguish between them by using $p$- and $s$-polarized photons. The detailed experiment geometry is shown in Fig. S2 and Note 5 in SM. Fig. 2(a) exhibits the photoemission intensity plot along $\Gamma$-$M$ with $p$-polarized 100 eV photons, corresponding to the $k_z$ plane nearly intersecting both predicted DPs [see details in Fig. S3 and Note 6 in SM]. We can directly identify the predicted BB1 (TDS) and BB2 (TI) bulk bands around the BZ center. Figs. 2(b)(i) and (ii) present second derivative photoemission intensity maps on the $k_m$-$k_z$ plane [the $k_m$ direction is indicated in Fig. 1(b)] taken with $p$-polarized photons at $E_F$ and $E_F - 0.4$ eV, respectively. Both band dispersions with apodicic periodic modulation along $k_z$ could be recognized, confirming the bulk nature of BB1 and BB2. Furthermore, we show four photoemission intensity plots taken at different $k_z$ as schematically illustrated in Fig. 2(e), from Cut 1 [Fig. 2(c)(i)] to Cut 4 [Fig. 2(c)(iv)]. The dispersion of BB2 varies from the parabolic lineshape to quasi-linear, and the apex keeps drifting up (highlighted by the yellow dashed line), showing the typical feature of a TI bulk cone [Fig. 2(e)]. In addition, the corresponding second derivative plots in Fig. 2(d)(i)-(iv) manifest the gradual increase of the Fermi crossing and more linear dispersion when close to the DP, in accordance with the predicted TDS feature of BB1 [Fig. 2(e)].

Next, we performed similar ARPES measurements but with $s$-polarized photons to identify topological surface states of BaTi$_2$Sb$_2$O. Fig. S2(a) shows the intensity plot along $\Gamma$-$M$ with $s$-polarized photons. The predicted surface bands SS1 and SS2 around $\Gamma$ were unambiguously revealed. Figs. S2(b) and (ii) represent second derivative intensity maps on the $k_m$-$k_z$ plane with $s$-polarized photons taken at $E_F$ and $E_F - 0.4$ eV, respectively. Our results demonstrate negligible $k_z$ dispersions for both bands, suggesting that bulk states have been significantly suppressed and surface states from TDS and TI are dominating here. Fig. S2(c) displays the enlarged second derivative plot of band structure in the red box of Fig. S2(a). Together with the appended band dispersions (blue and red dashed lines) extracted from the corresponding momentum distribution curves (MDCs) [Fig. S2(d)], we can well distinguish between those cone-like surface states from TDS and TI, in spite of rather small energy gap between these dual topological states. Furthermore, as exhibited by second derivative intensity plots in Figs. S2(e) and (f), detailed evolution of these surface cones along $k_z$ and $k_m$ could be recognized in evidence. The in-plane dispersion migrates apparently deviating $\Gamma$ at different $k_z$ and $k_m$ positions, suggesting that both surface DPs are exactly located at $\Gamma$.
Moreover, as displayed in Figs. 4(e), spin-resolved MDCs to those of right hand parts, consistent with our calculations. Evidently, for both TI and TDS surface cones, the left hand parts of these cones show the opposite spin direction. Consequently, for both TI and TDS surface cones, the left hand parts of these cones show the opposite spin direction. Evidently, for both TI and TDS surface cones, the left hand parts of these cones show the opposite spin direction. Evidently, for both TI and TDS surface cones, the left hand parts of these cones show the opposite spin direction. Evidently, for both TI and TDS surface cones, the left hand parts of these cones show the opposite spin direction. Evidently, for both TI and TDS surface cones, the left hand parts of these cones show the opposite spin direction. Evidently, for both TI and TDS surface cones, the left hand parts of these cones show the opposite spin direction. 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in the titanium-based oxypnictide superconductor BaTi$_2$Sb$_2$O$_7$ by ARPES measurements in association with first-principles calculations. The helical surface states from TDS which is in the vicinity of $E_F$ could produce the Majorana zero mode and Majorana flat band in this system. Therefore, BaTi$_2$Sb$_2$O$_7$ is a good platform to obtain various types of Majorana fermions and further research of topological superconductivity.

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FIG. 4. (a) Calculated spin texture along $\bar{\Gamma}$-$\bar{M}$ direction. Inset: the enlarged image of the white box region. (b) Sketch of the spin-polarized characteristics of TDS and TI surface states. The red and blue lines express spin down and up, respectively. (c) and (d) Intensity plot measured without and with $\hat{y}$ direction spin-polarized along $\bar{\Gamma}$-$\bar{M}$ direction. The red and blue parts express spin direction along $-\hat{y}$ and $+\hat{y}$, respectively. (e) Spin polarization at Cut 1 to 3 in (d), indicating the spin texture of SS1 to SS3. (f) Spin resolved MDCs taken at Cut 1 to 3. The red (down) and the blue (up) triangles indicate the spin direction along $-\hat{y}$ and $+\hat{y}$, respectively. (g) Sketch of the dual topological superconductivity states induced by TDS states.

* Equal contributions

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