

Ideal Weak Topological Insulator and Protected Helical Saddle Points

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Abstract

We report the design and observation of an ideal weak topological insulator in a quasi-one-dimensional bismuth halide, $\text{Bi}_4\text{I}_{1.2}\text{Br}_{2.8}$. Via angle-resolved photoemission spectroscopy, we identify that $\text{Bi}_4\text{I}_{1.2}\text{Br}_{2.8}$ hosts topological surface states on the (100)' side surface in the form of two anisotropic π -offset Dirac cones while topologically dark on the (001) top surface. The results fully determine a unique side-surface Hamiltonian and thereby identify two pairs of non-degenerate helical saddle points. The fact that both the surface Dirac and helical saddle points are in the global bulk band gap of 195 meV suggests potential of this system to be a fertile ground for topological many-body physics.

In three-dimensional (3D) bulk crystals, a strong topological insulator (STI) hosts an odd number of gapless Dirac surface states on every surface, while a weak topological insulator (WTI) has an even number of such surface states on selected surfaces [1–3]. The label “weak” versus “strong” refers to a perception that the topological surface states (TSS) of a WTI could be easily gapped via annihilation of the even number of Dirac cones (DCs) with Z_2 character due to translation symmetry breaking, making a WTI indistinguishable from a trivial insulator whereas a STI is robust [3]. Recent theoretical developments have shown, however, that not only is the gapless nature of WTIs robust to disorder [4–6], but also the initially perceived vulnerability bestows upon WTIs a desired degree of tunability: layer-dimerization gapping TSS in turn gives birth to gapless hinge modes—a mechanism for a topological phase transition into a higher-order topological insulator [7]. Hence, ideal WTIs possess much stronger potential for tunable transitions between 3D topologically distinct phases of matter and, as elucidated below, for realizing two-dimensional topological many-body physics.

Compared to the vast number of identified STIs, WTIs are challenging to realize experimentally, as implied by the two proposed pathways. The first involves periodic stacks of weakly coupled two-dimensional quantum spin Hall insulators where the TSS are located on non-cleavable side surfaces [8]. The second is to engineer a superlattice with multiple band inversions that requires fine tuning of intra- and inter-layer couplings [9,10]. Besides these challenges, TSS in WTIs reported so far, such as ZrTe_5 [11] and RhBi_2 [12], are buried deep in bulk states. Recently, a new series of quasi-one-dimensional (quasi-1D) material bismuth halides Bi_4X_4 ($\text{X} = \text{I}, \text{Br}$) with two different naturally cleavable surfaces accessible for ARPES has emerged [7,13–17]. Amongst these, $\beta\text{-Bi}_4\text{I}_4$ has been identified as a WTI that is only stable above 300 K below which a structural dimerization gaps out the TSS, leading to a higher-order topological insulator [13]. The high temperature range in which the WTI phase is stable makes it challenging to characterize the unique TSS, resulting in controversial angle-resolved photoemission spectroscopy (ARPES) observations and interpretations [13–15].

In order to realize a material platform with stable, tunable, and accessible surface-selective TSS, we designed a WTI in the form of $\text{Bi}_4\text{I}_{1.2}\text{Br}_{2.8}$ (BIB). Guided by the experimental confirmation that the β -phase is stable in Bi_4I_4 but absent in Bi_4Br_4 [18–20] and by the theoretical prediction that, if

synthesized, β -Bi₄Br₄ is a WTI superior to β -Bi₄I₄ because of a larger bulk gap [17], we searched for an optimized substitution ratio between I and Br for a single-stack crystal structure that is stable at all temperatures. We realized this optimization at the ratio of I:Br = 1.2:2.8 and its vicinity (characterization details are in the Supplemental Material [21]). We note that our X-ray refinement and EDX measurements show a single crystalline structure, demonstrating that BIB is a new phase instead of a phase separated mixture of the two end members. The building block of the quasi-1D crystal structure of BIB is the Bi-X chain along the b axis as shown in Fig. 1(a). The chain stacks along both the a and c axes, leading to two distinct natural cleavage planes—the (001) top surface exposing the ab plane and the (20 $\bar{1}$) side surface exposing the plane parallel to b and $a + 2c$. The (20 $\bar{1}$) direction in BIB corresponds to (100) in β -Bi₄I₄, where the difference originates from the stacking structure. For simplicity we will refer to it as (100)’ surface hereafter. Temperature-dependent resistivity curves confirm the lack of a structural distortion down to 2 K in Fig. 1(b). Large single crystals of BIB with extended thickness along both the a and c axes have been grown as shown in Fig. 1(c), allowing direct accesses to the (001) and the (100)’ surfaces in ARPES measurements.

Next, we examine the accessibility to the two inequivalent surfaces via ARPES (experimental details are in Supplemental Material [21]). As described above, the absence/presence of TSS for each surface provides definitive evidence for the topological character of a WTI candidate. Figures 2(a) and 2(b) show overviews of momentum-energy dispersions from the (001) and (100)’ surfaces, respectively. We observe distinct electronic states, signifying unambiguous single-facet observations for each measurement under a 40 μ m beam spot. The Fermi surface (FS) map from the (001) surface displays a spot-like feature [Fig. 2(a)], contrary to the parallel line FS across the (100)’ surface [Fig. 2(b)]. The clear difference in FS reveals that the interlayer interaction along the a axis is stronger than that along the c axis, consistent with the observations for Bi₄I₄ [13,15].

Having demonstrated the ability to distinguish the two surfaces, we classify the topological character of each surface (Fig. 2). First from the (001) surface, we investigate band dispersions at the two time-reversal-invariant momenta $\bar{\Gamma}$ and $\bar{M}$. While Fig. 2(c) shows a large gap at $\bar{\Gamma}$, at $\bar{M}$ two spectral features are observed with smaller energy scale in Fig. 2(e)—a valence band top at -0.24 eV

and a conduction band bottom just below the Fermi level, E_F . The conduction band bottom is visible likely due to electron doping from halide vacancies. Both the valence band top and the conduction band bottom are guided by the dotted grey lines in the energy distribution curves (EDCs) in Fig. 2(f). To obtain the band gap at $\bar{M}$, we fit the EDC at $k_y = 0$, the red curve in Fig. 2(f), and the gap is determined to be 195 meV with 126 eV photons, varying up to 210 meV with 118 eV photons [21]. Thus, we identify the global bulk gap of 195 ± 10 meV for BIB. Note that the bulk band gap of β -Bi₄I₄ is smaller than 100 meV [13].

By contrast, the (100)' side surface exhibits surface-originated linear crossings at two different time-reversal-invariant momenta. At both $\bar{\Gamma}$ and $\bar{Z}$, as shown in Figs. 2(g) and 2(j), the band dispersions show linear crossings below E_F and the crossing at $\bar{\Gamma}$ appears at -110 meV [Fig. 2(h)] while that at $\bar{Z}$ appears at -140 meV [Fig. 2(k)]. The energy positions of these crossings do not vary with the photon energy, indicating their surface nature [21]. To demonstrate their gapless nature, we plot the EDCs in Figs. 2(i) and (l) with guides on the band dispersions in grey (cyan) to indicate bulk (surface) state nature. We fit the EDCs at $k_y = 0$ and $k_y = \pm 0.01 \text{ \AA}^{-1}$ [21], and found that two peaks are necessary for EDCs at $k_y = \pm 0.01 \text{ \AA}^{-1}$ while only one is needed for $\bar{\Gamma}$ and $\bar{Z}$. Thus, we confirm that the crossings from the (100)' side surface are a pair of gapless TSSs [21].

Taking the observations of the (001) surface and the (100)' surface together, we have demonstrated the necessary conditions to identify BIB as a WTI as illustrated in Figs. 3(a) and 3(b). To verify this, we performed first-principles calculations for BIB. As shown in Fig. 3(c), the bulk electronic states of BIB near E_F are composed of Bi 6*p* states. Two band inversions between the interior and exterior Bi p_x [Fig. 1(a)] bands occur at the L and M points in the 3D Brillouin zone (BZ), which would annihilate when projected onto the (001) surface but be preserved as two linear Dirac crossings when projected onto the (100)' surface. Thus, BIB is a WTI with Z_2 invariants (0;001) based on the Fu-Kane criterion [22].

The existence of two distinct DCs on the (100)' surface, together with the quasi-1D geometry, leads to a unique topological structure with two protected pairs of helical saddle points. To illustrate this, we construct an effective Hamiltonian [7] constrained by the quasi-1D geometry, surface

symmetry, and WTI topology for the (100)' surface states (details in Supplemental Material [21]). The surface states of this minimal model are shown in Fig. 4(a), where there are two pairs of helical saddle points. The presence of these surface saddle points is a direct consequence of the weak dispersion in the stacking direction and the ultimate merger of the two surface DCs. Note that this model is constructed from the experimentally determined crystal structure and informed by the experimentally observed gapless DCs at $\bar{\Gamma}$ and $\bar{Z}$. The presence of the pairs of helical saddle points and the energy gap between $\bar{\Gamma}$ and $\bar{Z}$ is guaranteed because the two DCs must be connected between $\bar{\Gamma}$ and $\bar{Z}$, and time-reversal symmetry cannot protect any extra crossing between $\bar{\Gamma}$ and $\bar{Z}$. We note that it is challenging to directly resolve the saddle points separately from the Dirac points, given that the energy and momentum differences between a saddle point and its nearest Dirac point are 4.4 meV and $0.07 \text{ \AA}^{-1}$, respectively, while the energy resolution of our ARPES experiment is $\sim 20 \text{ meV}$ [21].

Having fully constrained the minimal model from a direct comparison of the theoretically calculated and experimentally obtained constant energy surfaces (CESs), shown in Figs. 4(d) and 4(e), respectively, we can now precisely determine the location of the helical saddle points with the Dirac points [Figs. 4(a) and 4(c)]. We also overlay the bulk conduction band (BCB) and bulk valence band (BVB) locations determined previously in Fig. 2(f). As shown in Fig. 4(c), the entire surface state structure is located within the global bulk band gap, which justifies that BIB exhibits the ideal WTI surface characteristics. Interestingly, this TSS gives rise to a series of four potential Lifshitz transitions that can be realized by tuning the chemical potential, as shown by the evolution of the CESs as a function of energy (Fig. 4(d)) [17].

A few remarks are in order. The existence of two pairs of helical saddle points is a part of the topological structure of the unique WTI surface state: the double DCs are π -offset in momentum, which is in sharp contrast to STI with a single surface DC. In BIB, the weak inter-layer coupling ensures the Dirac velocity v_z to be small and hence the saddle points to be in-gap. Recently, it has been suggested that one can artificially engineer Dirac flat bands by overlaying two STIs with a small twist [23,24], analogous to twisted bilayer graphene [25]. We emphasize that, in an ideal WTI such as BIB, it is naturally guaranteed to host two distinct DCs in the bulk gap with a narrow bandwidth between the two

Van Hove singularities (VHSs). In the case of BIB, this bandwidth is ~ 40 meV, which may be realized by trimerizing the layers or adding spacer layers, both of which are realizable amongst the class of layered bismuth halides. Last but not least, the non-degenerate surface CESs have non-trivial spin texture and can be closed or open.

The exotic topological manifold observed on the side surface of an ideal WTI has potentials to be a fertile ground for topological many-body physics, encouraging further investigations. One example is coupled helical Luttinger-liquid behavior when E_F is tuned beyond either of the two VHSs but still in the bulk gap where the corresponding CES is open. Indeed, the weak dispersion along the LM direction [Figs. 3(c) and 4] suggests that each (001) monolayer of BIB is a large-gap quantum spin Hall insulator. Appealingly, when such a surface state is proximitized [26] by a linear junction of a ferromagnetic insulator and an s -wave superconductor, a Majorana chain with precisely one Majorana zero mode per layer and uniform nearest-neighbor couplings are expected as shown in Fig. 5(a) [27]. Another example is unconventional indirect exciton condensation when E_F is tuned to the middle point between the $\bar{\Gamma}$ and $\bar{Z}$ Dirac point energies [28]. In this case, the electron and hole pockets are of the same size and thus enjoy perfect nesting as presented in Fig. 4(b). Notably, the spin textures of the two pockets are only half aligned for both the intra- and inter-surface pairing channels, given their helical characteristics as shown in Fig. 5(b) revealed by the effective model.

To summarize, we have designed and discovered an ideal WTI in BIB that is topologically dark on the (001) top surface yet hosts two π -offset DCs and two pairs of helical saddle points on the (100)' side surface. Excitingly, the complete set of topological objects on the (100)' surface state is exposed within the global bulk gap and hence accessible to quantum transport. As the chemical potential of bulk crystals is observed here to be at the edge of the bulk gap, gate tuning allows access to the various regimes in which novel physics is expected, given that gate-tunable boundary transport has recently been achieved in Bi_4I_4 field effect transistors and Josephson junctions [29]. Superconductivity under hydrostatic pressure have also been reported [30–32]. Altogether, the ideal WTI, BIB, offers a new surface platform for exploring the interplay between band topology and FS instabilities.

Acknowledgements

This work is mainly supported by National Science Foundation (NSF) through the DMREF program. This research used resources of the Advanced Light Source and the Stanford Synchrotron Radiation Lightsource, both U.S. DOE Office of Science User Facilities under contract Nos. DE-AC02-05CH11231 and DE-AC02-76SF00515, respectively. The work at UC Berkeley is supported by NSF Grant No. DMR-1921798. The work at Rice is supported by NSF under Grant No. DMR-1921847, the Robert A. Welch Foundation under Grant No. C-2024, and the Gordon and Betty Moore Foundation's EPiQS Initiative through grant No. GBMF9470. We acknowledge the Texas Advanced Computing Center (TACC) for providing resources that have contributed to the research results reported in this work. The work at UT Dallas is supported by NSF under Grant Nos. DMR-1921581 and DMR-1945351, AFOSR under Grant No. FA9550-19-1-0037, and Army Research Office (ARO) under Grant No. W911NF-18-1-0416.

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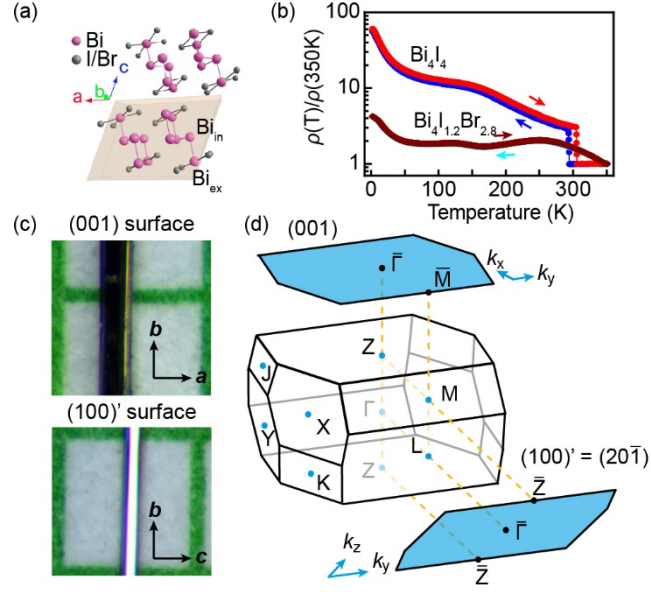


FIG. 1 (a) Crystal structure of BIB. A parallelepiped shows the unit cell. (b) Temperature-dependent resistivity for Bi_4I_4 and BIB. (c) Images for BIB crystals showing the accessible top (001) and side (100)' surfaces. The size of the grid is $1 \times 1 \text{ mm}^2$. (d) Bulk BZ and the (001) and (100)' surface BZs.

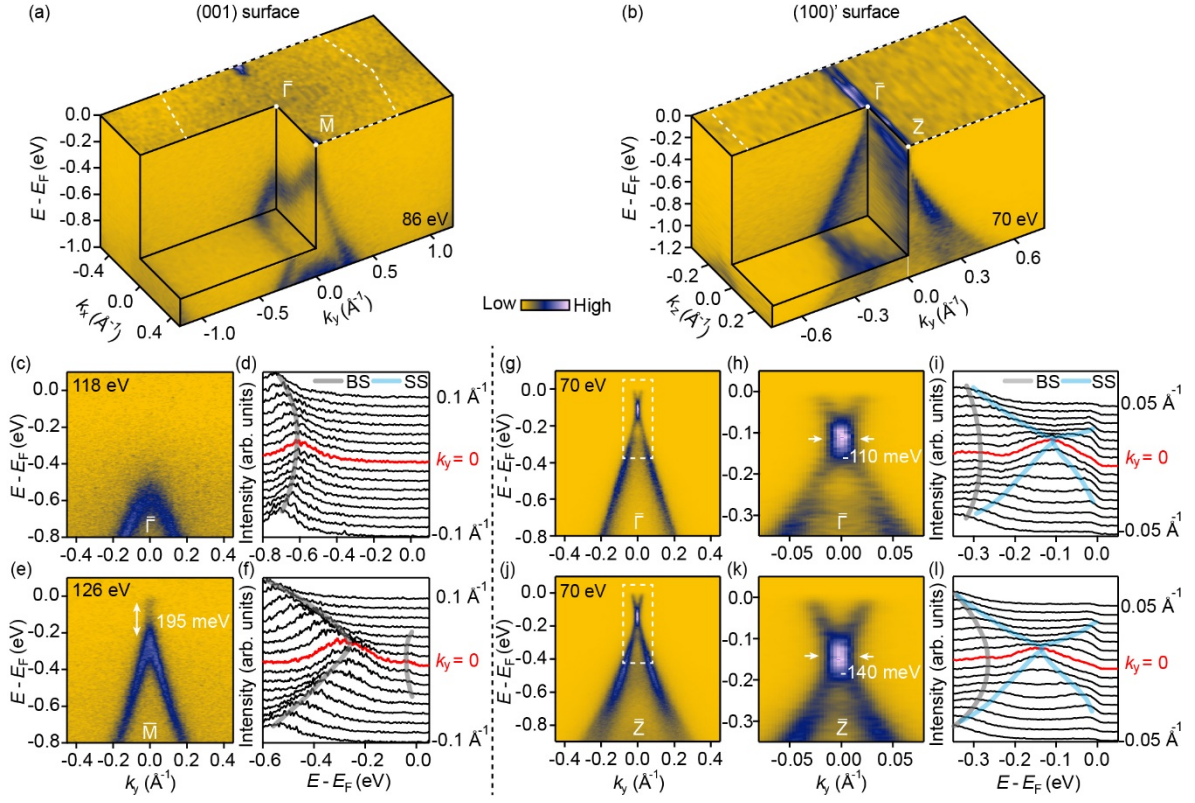


FIG. 2 (a) Overview of ARPES spectra with band dispersions across high symmetry points and FS maps on the (001) and (100)' surfaces. 86 eV and 70 eV photons were used for the (001) and (100)' surfaces, respectively. (c) and (d) Band dispersion across $\bar{\Gamma}$ on the (001)-projected surface BZ and the corresponding EDCs. (e) and (f) Analogous plots to (c) and (d) at $\bar{M}$ on the (001) surface. (g) Band dispersions across $\bar{\Gamma}$ on the (100)'-projected surface BZ. (h) Zoomed-in view of the white box in (g), showing the Dirac crossing at -110 meV. (i) corresponding EDCs from (h). (j)–(l) Equivalent plots as (g)–(i) for the $\bar{Z}$ point on the (100)'-projected surface BZ with a Dirac crossing at -140 meV.

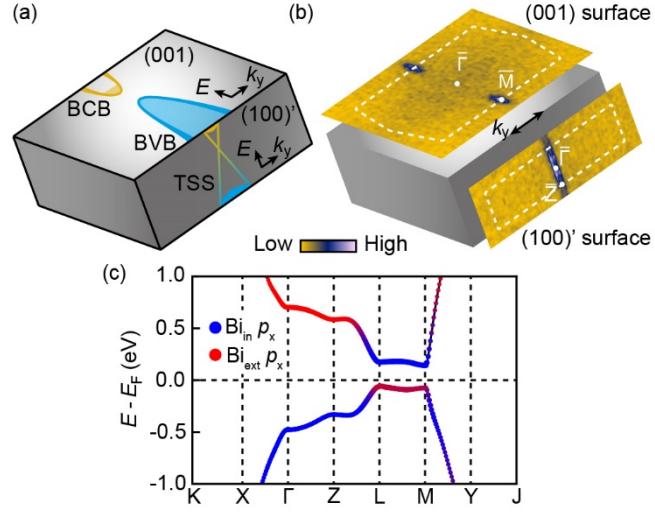


FIG. 3 (a) A schematic overview for the WTI nature of BIB based on experimental observations. (b) FSs measured on the (001) and (100)' surfaces. (c) First-principles bulk calculations for BIB.

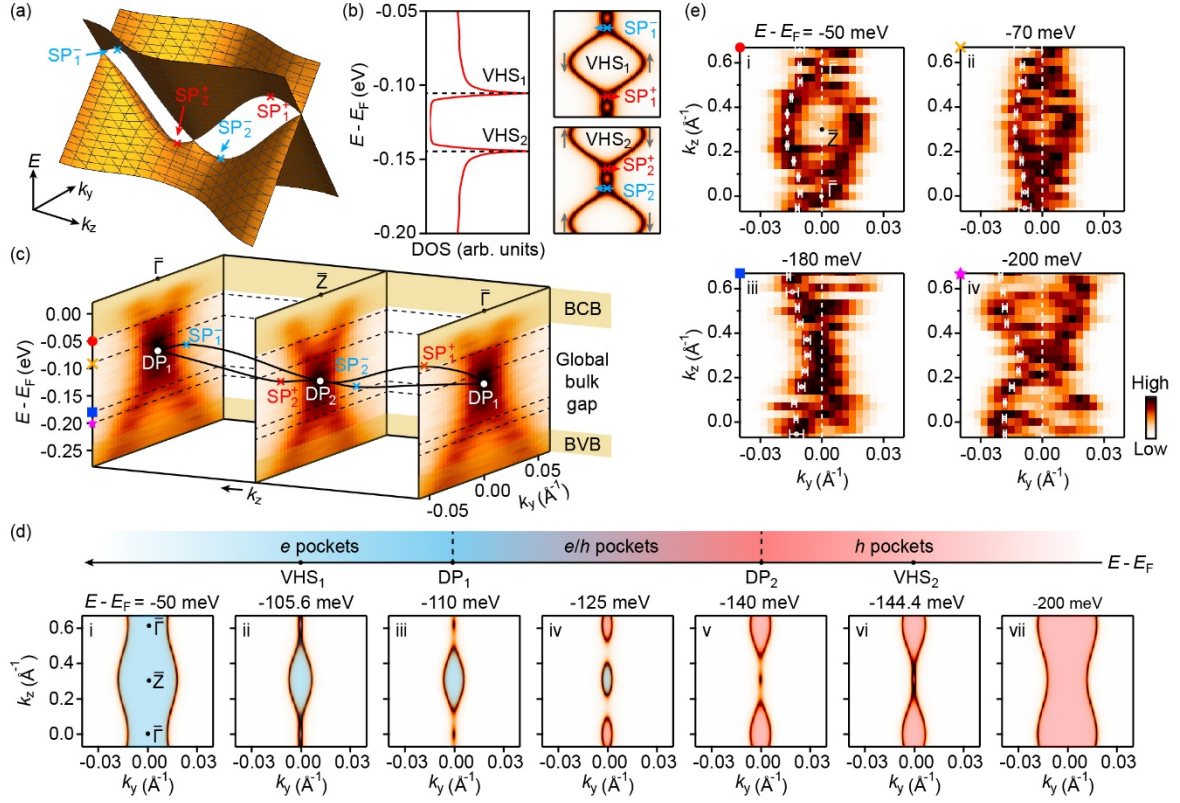


FIG. 4 (a) 3D electronic structure given by the minimal model. Two pairs of helical saddle points are labeled as $SP_{1,2}^{\pm}$. Subscripts 1 (2) indicate the energy location higher (lower) than that of the Dirac points, and superscripts $\pm$ indicate the spin polarizations. (b) Left: Calculated surface state density of states using the minimal model. Two saddle-point Van Hove singularities ($VHS_{1,2}$) are shown. Right: Spin textures on CESs at $VHS_{1,2}$. (c) 3D plot for a comprehensive description of the electronic structure on the $(100)'$ surface. $DP_{1,2}$ show the Dirac points at $\bar{\Gamma}$ and $\bar{Z}$, respectively. (d) CESs obtained from the minimal model at key energies indicated by the bar along the energy axis. Shades on CESs indicate electron- (blue) and hole- (red) pockets. (e) Experimentally obtained CESs above and below $VHS_{1,2}$ and $DP_{1,2}$. Colored marks on the left top corners indicate the energy positions in (c). White dots with error bars (standard deviation for the results) are fitted positions of CESs.

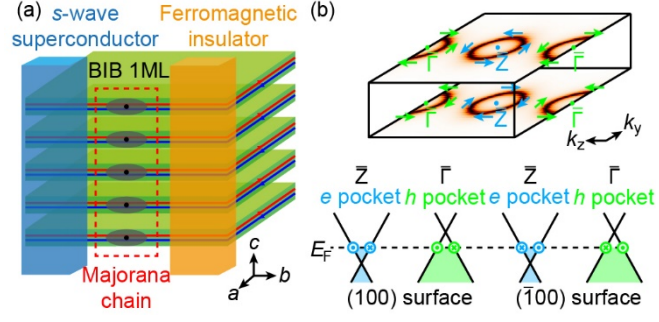


FIG. 5 (a) Emergence of Majorana chain when the (100)' TSS is proximitized to a linear junction of a ferromagnetic insulator and an *s*-wave superconductor. (b) Unconventional exciton condensation by perfect nesting condition between electron and hole pockets in the TSS where their spin textures are only half-aligned for both the intra- and inter-surface pairing channels.