

# Fulde-Ferrell-Larkin-Ovchinnikov pairing induced by a Weyl nodal line in an Ising superconductor with a high critical field

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(Received 15 April 2021; revised 30 November 2021; accepted 10 December 2021; published 6 January 2022)

Superconductivity and electron topology are two quantum phenomena that have attracted much interest, but no causal relationship between them has been reported because superconductivity is a many-body effect due to electron-electron interaction, while electron topology is a single-particle manifestation of electron states. Here, we demonstrate that electron topology can induce Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) pairing in Ising Bardeen-Cooper-Schrieffer (IBCS) superconductors. Specifically, we predict that the nonmagnetic metals of the  $MA_2Z_4$  family, including  $\alpha_1$ -TaSi<sub>2</sub>P<sub>4</sub>,  $\alpha_1$ -TaSi<sub>2</sub>N<sub>4</sub>,  $\alpha_2$ -TaGe<sub>2</sub>P<sub>4</sub>,  $\alpha_1$ -NbSi<sub>2</sub>P<sub>4</sub>, and  $\alpha_2$ -NbGe<sub>2</sub>P<sub>4</sub> monolayers, are all IBCS superconductors with a transition temperature ranging from a few to tens of degrees Kelvin. The intrinsic IBCS pairing alone will enhance the in-plane critical field  $B_c$  to  $\sim 20$ – $60$  times the Pauli limit  $B_p$ , and the extrinsic FFLO pairing evoked by topological Weyl nodal lines under a magnetic field can further double the  $B_c/B_p$  ratio. Our findings not only enrich the fundamental relationship between superconductivity and electron topology, but they also yield an effective approach to enhance the robustness of superconductivity.

DOI: [10.1103/PhysRevB.105.024505](https://doi.org/10.1103/PhysRevB.105.024505)

## I. INTRODUCTION

Superconductivity [1] and electron topology [2,3] are two landmark breakthroughs in the fields of condensed-matter physics and materials science. When they are brought together by the proximity effect [4,5], or when they coexist in one material, e.g., superconductors with topological states [6–8] and *vice versa* [9], a more exotic quantum state of topological superconductivity arises, offering a promising route to fault-tolerant Majorana-based quantum computing [10,11]. However, conventional wisdom tells us that there is no causal relationship between superconductivity and electron topology. This is understandable because the former is a many-body effect manifesting an attractive electron-electron interaction of Cooper pairs [12], while the latter is a single-particle effect induced by parity inversion in the electron band structure [13]. Surprisingly, here we reveal an alternative form of Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) superconducting pairing [14,15], induced by topological Weyl nodal lines in the family of two-dimensional (2D) Ising superconductors of  $MA_2Z_4$  monolayers [16,17]. It not only sheds new light on our fundamental understanding of superconductivity in relation with topology, but it also provides a promising approach to enhance the robustness of superconductors.

In addition to the critical transition temperature ( $T_c$ ), another important figure of merit for superconductivity is the critical magnetic field ( $B_c$ ), beyond which the superconductivity vanishes. Generally, a magnetic field destroys superconductivity through orbital and/or Pauli paramagnetic

mechanisms. Because the orbital effect is weak or absent in those materials with a large electron mass [18] or low dimensionality [19], suppressing the Pauli effect has been the focus in order to increase  $B_c$ . In particular, the FFLO pairing [14,15,20–25] has long been shown to be a feasible mechanism to enhance  $B_c$  beyond the Pauli paramagnetic limit ( $B_p$ ). The formation of FFLO pairs, with nonzero momentum, stems from the spin-nondegenerate Fermi surfaces (FSs) induced by an external magnetic field [20,21]. They are favored by low-dimensionality, anisotropic FS, and nesting [26], tending to infinity at the 1D limit at low temperature. The FFLO pairing has been mainly found in quasi-2D clean-limit superconductors, such as organic superconductors [21], cuprate superconductors [22], iron-based superconductors [23], heavy-fermion superconductors [20], and van der Waals (vdW) layered NbS<sub>2</sub> [24,25].

On the other hand, recent studies have shown significantly enhanced  $B_c$  in 2D Ising Bardeen-Cooper-Schrieffer (IBCS) superconductors [27–37], which suppresses the Pauli pair-breaking effect by an effective out-of-plane Zeeman field ( $B_{\text{eff}}$ ), induced by spin-orbit-coupling (SOC) together with inversion asymmetry (type-I) [27–30], or with multiple degenerate orbitals (type-II) [31–33,37]. Such mechanisms have been identified in transition-metal dichalcogenide (TMD) monolayers [27–30,32,34–36], few-layer stanene [33], and Pb films [37] to increase  $B_c$  several times over  $B_p$ . Here, we demonstrate the coexistence of both IBCS and FFLO pairing in one material, namely the 2D  $MA_2Z_4$  monolayer. It is made possible by the coexistence of 2D spin-valley-locking bands and 1D Weyl nodal lines in the vicinity of the Fermi level. The 2D FSs around  $K$  ( $K'$ ) and  $\Gamma$  support IBCS pairing by the spin-valley locking [Fig. 1(a)], while the “effective 1D FSs” arising from the Weyl nodal lines along  $M$ - $\Gamma$ - $M'$  enable

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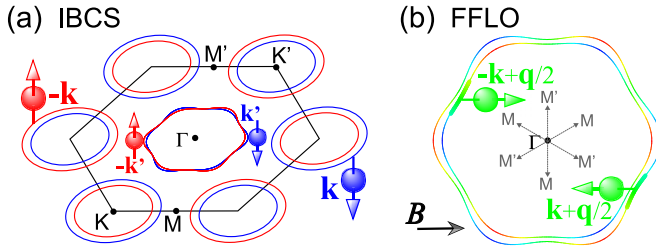


FIG. 1. Schematic illustration of superconducting pairing on the FSs of  $MA_2Z_4$  monolayer. (a) IBCS pairing on all FS contours; (b) FFLO pairing on the FS contour around the  $\Gamma$  point. Red and blue lines denote the out-of-plane up- and down-spin states, respectively, while the green indicates the states with nearly zero out-of-plane and large in-plane spin-polarization due to the in-plane magnetic field  $B$ .

FFLO pairing under a magnetic field [Fig. 1(b)]. The latter is attributed to the field lifting the degeneracy of Weyl nodal lines to generate spin-polarized electrons (anti)parallel to the field direction [green FS in Fig. 1(b)], which are no longer favored for IBCS pairing when the field is close to or exceeds  $B_p$ . Instead, inhomogeneous FFLO pairing with a finite  $\mathbf{q}$  starts to emerge between these spin-polarized electrons on three 1D FSs along the  $M$ - $\Gamma$ - $M'$  paths [Fig. 1(b)], where the local  $B_{\text{eff}}$  is weakest. Importantly, the FFLO pairing formed on 1D FS has the maximum stability [20,21].

It is worth mentioning that centimeter-scale monolayer films of  $\text{MoSi}_2\text{N}_4$  and  $\text{WSi}_2\text{N}_4$  have already been synthesized recently by chemical vapor deposition [16], which opens up a large family of 2D vdW layered materials with the general formula of  $MA_2Z_4$  [16,17] that has no 3D bulk counterparts [38]. Extensive computational research shows that the  $MA_2Z_4$  monolayers generally exhibit outstanding mechanical, thermal, electronic, optical, piezoelectric, thermoelectric, optoelectronic, and photocatalytic properties [16,17,39–44]. Of particular interest to us, certain  $MA_2Z_4$  compounds have been theoretically predicted to be intrinsic superconductors without charge-density-wave (CDW) instability [17]. Since the  $MA_2Z_4$  monolayers lacking inversion symmetry possess similar Zeeman-type spin-valley locking to that of the  $\text{MoS}_2$ -family monolayers [17,42–44], one might expect that if superconducting, they may have a high  $B_c$ . Lo and behold, we found that some superconducting 2D  $MA_2Z_4$  monolayers have the highest  $B_c/B_p$  ratio to date, to the best of our knowledge.

Our discovery is partly enabled by our recent development of a first-principles computational approach for superconductivity [45] by self-consistently solving the superconducting gap equation constructed from density-functional-theory based Wannier functions (WFs) and electron-phonon coupling (EPC) calculations, especially in the presence of an external magnetic field [see details from Note S1 of the supplemental material (SM) [46]]. It allows us to predict not only  $T_c$  but also  $B_c$  of a superconductor, as well as topological superconductors [45,72,73]. To benchmark this newly developed method, we first solved self-consistently the critical magnetic field of electron-doped  $\text{WS}_2$ , whose superconductivity properties are already experimentally available for comparison [36]. Our calculated results reproduce very well the experimental results, especially the measured enhancement of the critical

field  $B_c/B_p$  ratio (see Note S2 of the SM [46]). This gives us confidence in making new predictions for  $MA_2Z_4$ , since they share the same crystal symmetry and similar band structure with  $\text{WS}_2$ . Also, both systems possess the same spin-valley locking features near the  $K$  ( $K'$ ) point giving rise to IBCS pairing. In fact,  $\text{WS}_2$  also has Weyl nodal lines along  $\Gamma$ - $M$  paths, but they lie way below the Fermi level (Fig. S1a [46]), so they do not contribute to superconductivity. This makes  $\text{WS}_2$  a type-I Ising superconductor with solely IBCS pairing. In contrast, the  $\Gamma$ - $M$  nodal lines in the considered  $MA_2Z_4$  lie right at the Fermi level, which led to our discovery of the Weyl-induced FFLO pairing and its effect on enhancing the critical magnetic field.

We have systematically investigated the field-dependent superconductivity of  $\alpha_1$ - $\text{TaSi}_2\text{P}_4$ ,  $\alpha_1$ - $\text{TaSi}_2\text{N}_4$ ,  $\alpha_2$ - $\text{TaGe}_2\text{P}_4$ ,  $\alpha_1$ - $\text{NbSi}_2\text{P}_4$ , and  $\alpha_2$ - $\text{NbGe}_2\text{P}_4$  monolayers. We found that without a magnetic field,  $T_c$  is  $\sim 22.5$  K for  $\alpha_1$ - $\text{TaSi}_2\text{N}_4$  and below 10 K for others. The critical field near 0 K is estimated to be  $\sim 70$  and  $\sim 100$  times  $B_p$ , respectively, for  $\alpha_1$ - $\text{TaSi}_2\text{P}_4$  and  $\alpha_2$ - $\text{TaGe}_2\text{P}_4$ , and it reaches  $\sim 20$ – $30$  times for others, due to the cooperation of the IBCS and FFLO mechanisms. By fitting the self-consistently calculated  $B_c/B_p$  ratios at different temperatures using an extended microscopic model of an Ising superconductor, the FFLO pairing on the FS around the  $\Gamma$  point was demonstrated to show significant enhancements on the  $B_c/B_p$  of  $\alpha_1$ - $\text{TaSi}_2\text{P}_4$ ,  $\alpha_1$ - $\text{TaSi}_2\text{N}_4$ ,  $\alpha_2$ - $\text{TaGe}_2\text{P}_4$ , and  $\alpha_2$ - $\text{NbGe}_2\text{P}_4$  monolayers, while the enhancement is negligible for the  $\alpha_1$ - $\text{NbSi}_2\text{P}_4$  monolayers. This is attributed to the fact that the “effective 1D FS” associated with the Weyl nodal lines, which favors the FFLO pairing, is closely related to the strength of Ising SOC, and SOC that is too weak, as in  $\alpha_1$ - $\text{NbSi}_2\text{P}_4$ , cannot maintain the 1D FS under a magnetic field.

## II. RESULTS AND DISCUSSION

### A. Superconductivity under a magnetic field

The atomic structure of the  $MA_2Z_4$  monolayer can be viewed as the  $\text{MoS}_2$ -type  $MZ_2$  monolayer with the surface dangling bonds passivated by InSe-type  $A_2Z_2$ , constituting a septuple layer of  $Z$ - $A$ - $Z$ - $M$ - $Z$ - $A$ - $Z$  [16,17]. This unique sandwich structure creates a large  $MA_2Z_4$  family with diverse properties arising from varying compositions and relative positions between atomic planes. Here we focus on the non-magnetic metal compounds, which are stable in the  $\alpha_1$  and  $\alpha_2$  phases, including  $\alpha_1$ - $\text{TaSi}_2\text{P}_4$ ,  $\alpha_1$ - $\text{TaSi}_2\text{N}_4$ ,  $\alpha_2$ - $\text{TaGe}_2\text{P}_4$ ,  $\alpha_1$ - $\text{NbSi}_2\text{P}_4$ , and  $\alpha_2$ - $\text{NbGe}_2\text{P}_4$  [17]. From Figs. 2(a) and 2(b), one sees that these noncentrosymmetric phases lack inversion symmetry but contain out-of-plane mirror symmetry  $m_z$ . Consequently, SOC induces a  $B_{\text{eff}}$  to orient electron spins in the out-of-plane direction, manifesting a Zeeman-type spin-valley locking [17,42–44]. This feature can be clearly seen from Figs. 2(c)–2(d) and Fig. S2, as obtained from the first-principles calculations (Note S1 of the SM [46]). Moreover, the up- and down-spin branches cross each other along the  $M'$ - $\Gamma$ - $M$   $\mathbf{k}$ -point paths, forming three  $m_z$ -protected Weyl nodal lines (see the theoretical analysis in Note S3 of the SM [46]). The generic metallic nature with large Ising spin-splitting at the Fermi level provides the precondition for the intrinsic

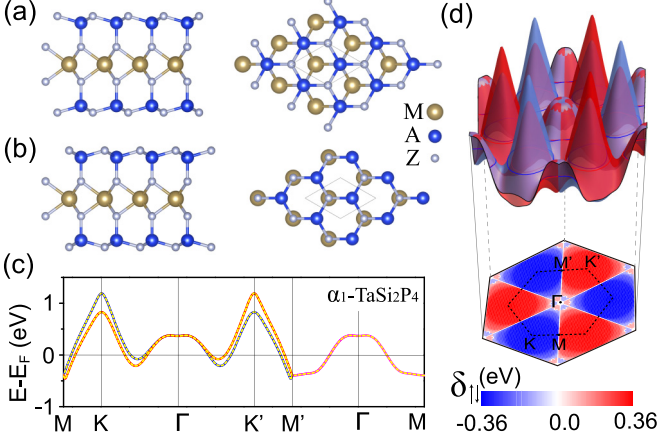


FIG. 2. The atomic and electronic structures. Side (left) and top (right) views of (a)  $\alpha_1$ - $MA_2Z_4$  and (b)  $\alpha_2$ - $MA_2Z_4$  monolayer. (c) Band structure of  $\alpha_1$ - $TaSi_2P_4$  monolayer, showing typical features of spin-valley locking and Weyl nodal lines. Red and blue lines denote, respectively, the split out-of-plane up- and down-spin states, while the pink line indicates the spin-degenerate Weyl nodal line. Yellow dashed lines are the WF fits of the band structure. (d) 3D band structure (top panel) plot and 2D distribution of spin-splitting  $\delta_{\uparrow\downarrow} = E_{\uparrow} - E_{\downarrow}$  (bottom), i.e., the effective Zeeman field  $B_{\text{eff}} = \delta_{\uparrow\downarrow}/\mu_B$  for the two metallic bands, which vanishes along the  $M'$ - $\Gamma$ - $M$  paths (white lines).

IBCS pairing, while the 1D Weyl states are shown to initiate and stabilize the extrinsic FFLO pairing under a magnetic field by lifting the spin degeneracy [20,21,74].

To quantitatively characterize the anticipated superconductivity in  $MA_2Z_4$  monolayers, we first calculated the EPC strength  $\lambda$  and estimated critical temperature  $T_c^{\text{AD}}$  using the Allen-Dynes (AD) modified McMillan's formula (see Note S1 of the SM [46]). Specifically, taking  $\alpha_1$ - $TaSi_2P_4$  as an example, the calculated phonon spectra [Fig. 3(a)] indicate that EPC induces a phonon mode softening, but no CDW instability that is known to be detrimental to IBCS [29,30,34,35] and FFLO pairing [74]. The CDW instability is still absent when temperature is increased to 100 K (see Note S4 and Fig. S3 in the SM [46]). The total EPC  $\lambda$  is calculated to be 0.77 from the cumulative EPC  $\lambda(\omega)$  [Fig. 3(b)], which stems mainly from the couplings between the in-plane phonon vibrations of Ta and the electrons on the  $d_{xy}$ ,  $d_{x^2-y^2}$ , and  $d_{z^2}$  orbitals of the Ta atom (Fig. S4 [46]). With the logarithmically averaged frequency  $\langle\omega\rangle_{\log}$  being evaluated to be 105.56 K from the Eliashberg spectral function  $\alpha^2F(\omega)$  [Fig. 3(b)], the  $T_c^{\text{AD}}$  was estimated to be  $\sim 4.60$  K. The key features of  $\alpha_1$ - $TaSi_2P_4$  are also found in other  $MA_2Z_4$  monolayers (Fig. S5 [46]).

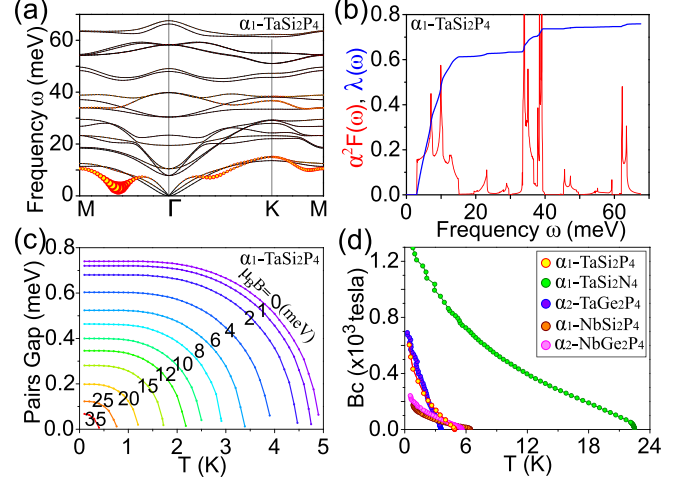


FIG. 3. The superconductivity under an in-plane magnetic field. (a) Phonon spectra with the magnitude of the EPC strength  $\lambda_{qv}$  being drawn proportional to the size of the yellow filled circles. (b) Plots of Eliashberg spectral function  $\alpha^2F(\omega)$  and cumulative frequency-dependent EPC strength  $\lambda(\omega)$ . (c) The temperature-dependent pairing gaps under different in-plane magnetic fields (color bar). (d) The temperature-dependent in-plane critical magnetic field for the five  $MA_2Z_4$  monolayers.

We summarize the superconductivity related parameters, e.g., EPC  $\lambda$ , the density of states (DOS) at the Fermi level  $N_F$ , logarithmically averaged frequency  $\langle\omega\rangle_{\log}$ , and  $T_c^{\text{AD}}$  in Table I. The convergence of these values was carefully checked (Tables S1 and S2 [46]). Qualitatively, the finite  $T_c^{\text{AD}}$  (Table I) indicates that all five  $MA_2Z_4$  monolayers are likely to exhibit intrinsic superconductivity without the need for doping.

Since the above conventional first-principles methods of estimating critical temperature cannot be applied to the systems without time-reversal symmetry, we next investigate further the superconductivity and its dependence on an in-plane magnetic field for the five  $MA_2Z_4$  monolayers using the newly developed method of self-consistently solving the first-principles WF gap equation (see Note S5 of the SM [46]). We note that for 2D Ising superconductors under a perfectly aligned in-plane field, the orbital depairing mechanism can be neglected, and then there is only one in-plane critical field  $B_c$  above which superconductivity vanishes [27,33,37]. Thus, in the present study, only the suppressing effect of Pauli paramagnetic mechanisms on superconductivity is considered.

Without a magnetic field, the self-consistently calculated critical temperature  $T_c^{\text{SCF}}$  agrees well with  $T_c^{\text{AD}}$  for those  $MA_2Z_4$  monolayers with intermediate EPC strength  $\lambda < 1.0$ , which gives a good starting point to perform further calcula-

TABLE I. The parameters related to the superconductivity of  $MA_2Z_4$  monolayers.

	$\alpha_1$ - $TaSi_2P_4$	$\alpha_1$ - $TaSi_2N_4$	$\alpha_2$ - $TaGe_2P_4$	$\alpha_1$ - $NbSi_2P_4$	$\alpha_2$ - $NbGe_2P_4$
$\lambda$	0.77	1.29	0.66	0.79	0.80
$N_F$ ( $eV^{-1}$ )	2.04	1.92	1.96	2.22	2.18
$\langle\omega\rangle_{\log}$ (K)	105.56	231.17	105.48	136.93	115.25
$T_c^{\text{AD}}$ (K)	4.60	22.46	3.12	6.31	5.35

TABLE II. The parameters used to calculate and analyze the superconductivity of  $MA_2Z_4$  monolayers with IBCS and FFLO pairing under a magnetic field.

	$\alpha_1$ -TaSi <sub>2</sub> P <sub>4</sub>	$\alpha_1$ -TaSi <sub>2</sub> N <sub>4</sub>	$\alpha_2$ -TaGe <sub>2</sub> P <sub>4</sub>	$\alpha_1$ -NbSi <sub>2</sub> P <sub>4</sub>	$\alpha_2$ -NbGe <sub>2</sub> P <sub>4</sub>
$g$	0.28	0.41	0.23	0.27	0.27
$T_c^{\text{SCF}}$ (K)	4.90	22.50	3.55	6.45	5.60
$\delta_{F\uparrow\downarrow}^{\text{K}(K')}$ (meV)	$\sim 179.7$	$\sim 197.1$	$\sim 171.5$	$\sim 59.1$	$\sim 59.6$
$\delta_{F\uparrow\downarrow}^{\Gamma}$ (meV)	$\sim 31.0$	$\sim 46.1$	$\sim 64.8$	$\sim 5.1$	$\sim 15.4$
$\tilde{\beta}_{\text{SOC}}^{\text{K}(K')}$ (meV)	40	80	40	30	20

tions with a magnetic field by employing the evaluated pairing strength  $g$  (Table II). But for  $\alpha_1$ -TaSi<sub>2</sub>N<sub>4</sub> with  $\lambda = 1.29$ , a case of strong EPC,  $T_c^{\text{SCF}} \sim 47.00$  K is obtained by using the evaluated  $g = 0.57$ , which overestimates by about two times compared with  $T_c^{\text{AD}} \sim 22.46$  K. This indicates that the first-principles WF gap equation can be safely applied only to superconductors with intermediate EPC strength. We thus purposely reduce the  $g$  to 0.41 for  $\alpha_1$ -TaSi<sub>2</sub>N<sub>4</sub> to reproduce  $T_c^{\text{AD}}$  for investigating its field-dependent superconductivity.

Figure 3(c) shows the calculated pairing gap  $\Delta$  of  $\alpha_1$ -TaSi<sub>2</sub>P<sub>4</sub> as a function of temperature  $T$  and in-plane field  $B$ . When  $B = 0$ , the pairing gap at 0 K is  $\Delta_0 \sim 0.74$  meV, which is fully suppressed at  $T_c^{\text{SCF}} \sim 4.90$  K, in good agreement with  $T_c^{\text{AD}} \sim 4.60$  K. Due to the Pauli paramagnetic pair-breaking effect,  $T_c^{\text{SCF}}$  decreases with increasing field. The gap is found to close at 0 K when  $\mu_{\mathbf{B}}B$  ( $\mu_{\mathbf{B}}$  is the Bohr magneton) is larger than  $\sim 35$  meV, translating to an in-plane critical magnetic field  $B_c \sim 600$  tesla for  $\alpha_1$ -TaSi<sub>2</sub>P<sub>4</sub> monolayer. Accordingly,  $\Delta(T, B)$  is evaluated for  $\alpha_1$ -TaSi<sub>2</sub>N<sub>4</sub>,  $\alpha_2$ -TaGe<sub>2</sub>P<sub>4</sub>,  $\alpha_1$ -NbSi<sub>2</sub>P<sub>4</sub>, and  $\alpha_2$ -NbGe<sub>2</sub>P<sub>4</sub> using the parameters listed in Tables I and II (see Fig. S7 [46]), which enables us to extract their  $B_c$  versus  $T$  [Fig. 3(d)]. In all five cases, the  $B_c$  show an upturn when  $T \rightarrow 0$  K, consistent with IBCS and/or the FFLO mechanism. The  $B_c$  at 0 K for  $\alpha_1$ -TaSi<sub>2</sub>N<sub>4</sub>,  $\alpha_2$ -TaGe<sub>2</sub>P<sub>4</sub>,  $\alpha_1$ -NbSi<sub>2</sub>P<sub>4</sub>, and  $\alpha_2$ -NbGe<sub>2</sub>P<sub>4</sub> are  $\sim 1300$ ,  $\sim 700$ ,  $\sim 170$ , and  $\sim 240$  tesla, respectively. In general, a higher  $T_c^{\text{SCF}}$  together with larger Ising spin-splitting  $\delta_{F\uparrow\downarrow}^{\Gamma}$  and  $\delta_{F\uparrow\downarrow}^{\text{K}(K')}$  on the FSs around  $\Gamma$  and  $K$  ( $K'$ ) (Table II) tend to induce larger  $B_c$ .

To give a semiquantitative measure on the robustness of superconductivity against the field in the five  $MA_2Z_4$  monolayers, we evaluate the normalized critical field by the Pauli limit  $B_p = \Delta_0/(\sqrt{2}\mu_{\mathbf{B}})$ , i.e.,  $B_c/B_p$ , as a function of the normalized temperature,  $T/T_c^{\text{SCF}}$ , as shown by the filled circles in Figs. 4(a)–4(e). At 0 K, the  $B_c/B_p$  ratio can reach as high as  $\sim 100$  in  $\alpha_2$ -TaGe<sub>2</sub>P<sub>4</sub> monolayer [Fig. 4(c)], and that of  $\alpha_2$ -TaSi<sub>2</sub>P<sub>4</sub> exceeds  $\sim 70$  [Fig. 4(a)].  $\alpha_1$ -TaSi<sub>2</sub>N<sub>4</sub>,  $\alpha_2$ -NbGe<sub>2</sub>P<sub>4</sub>, and  $\alpha_1$ -NbSi<sub>2</sub>P<sub>4</sub> possess the  $B_c/B_p$  ratio within the range of 20–30 [Figs. 4(b), 4(d), and 4(e)]. The predicted  $B_c/B_p$  ratios are all higher than most known Ising superconductors.

### B. Analysis of the high critical field

To better understand the physical origin of such a high critical field, we extend the conventional microscopic model of the Ising superconductor with one effective Ising SOC strength [32,33,37,75] to the case with different SOC strengths (see Note S6 of the SM [46]), which resulted in the following

equations:

$$F(\mu_{\mathbf{B}}B_c, \tilde{\beta}_{\text{SOC}}^{\Gamma}) + 2 \times F(\mu_{\mathbf{B}}B_c, \tilde{\beta}_{\text{SOC}}^{\text{K}(K')}) = 0, \quad (1)$$

$$\begin{aligned} & F(\mu_{\mathbf{B}}B_c, \tilde{\beta}_{\text{SOC}}) \\ & \equiv \ln(T/T_c) + \frac{(\mu_{\mathbf{B}}B_c)^2}{(\tilde{\beta}_{\text{SOC}})^2 + (\mu_{\mathbf{B}}B_c)^2} \\ & \times \text{Re} \left[ \psi \left( \frac{1}{2} + \frac{i\sqrt{(\tilde{\beta}_{\text{SOC}})^2 + (\mu_{\mathbf{B}}B_c)^2}}{2\pi k_{\mathbf{B}}T} \right) - \psi \left( \frac{1}{2} \right) \right]. \end{aligned} \quad (2)$$

Here  $\psi(X)$  is the digamma function. It enables us to estimate the in-plane critical field of IBCS pairing for  $MA_2Z_4$  monolayers, which consist of one FS with the effective Ising SOC strength of  $\tilde{\beta}_{\text{SOC}}^{\Gamma}$  and two FSs with  $\tilde{\beta}_{\text{SOC}}^{\text{K}(K')}$ . The  $\tilde{\beta}_{\text{SOC}}^{\Gamma}$  and  $\tilde{\beta}_{\text{SOC}}^{\text{K}(K')}$  are closely related to the average magnitude of the Ising spin-splitting  $\delta_{F\uparrow\downarrow}^{\Gamma}$  and  $\delta_{F\uparrow\downarrow}^{\text{K}(K')}$  on the FSs around  $\Gamma$  and  $K$  ( $K'$ ), respectively (Table II). Without losing generality, we set  $\tilde{\beta}_{\text{SOC}}^{\Gamma} = \tilde{\beta}_{\text{SOC}}^{\text{K}(K')} \times \delta_{F\uparrow\downarrow}^{\Gamma}/\delta_{F\uparrow\downarrow}^{\text{K}(K')}$  and tune the value of  $\tilde{\beta}_{\text{SOC}}^{\text{K}(K')}$  to fit the self-consistent solutions at low field, where the IBCS pairing is dominant over the FFLO pairing.

The dependences of  $B_c/B_p$  on  $T/T_c^{\text{SCF}}$  for the five  $MA_2Z_4$  monolayers are fitted using the values of  $\tilde{\beta}_{\text{SOC}}^{\text{K}(K')}$  summarized in Table II. Taking  $\alpha_1$ -TaSi<sub>2</sub>P<sub>4</sub> as an example [Fig. 4(a)], one can clearly see that the dependence can be well reproduced at low magnetic field. Upon further enhancing the field, a significant deviation between the extended microscopic model and the self-consistent solution emerges, which increases with the increasing field noticeably. This is because more and more FFLO pairs start to form, as more electron spins in the vicinity of Weyl nodal lines will be reoriented with those having a zero out-of-plane component to satisfy the FS nesting condition [green color in Fig. 1(b)]. It clearly illustrates that the additional FFLO pairing can indeed further enhance the robustness of superconductivity under magnetic field [light blue shaded region in Fig. 4(a)], by as much as double the critical field enhanced by IBCS pairing [inset of Fig. 4(a)]. Similar analysis is applied to  $\alpha_1$ -TaSi<sub>2</sub>N<sub>4</sub> [Fig. 4(b)],  $\alpha_2$ -TaGe<sub>2</sub>P<sub>4</sub> [Fig. 4(c)], and  $\alpha_2$ -NbGe<sub>2</sub>P<sub>4</sub> [Fig. 4(d)] monolayers. However, the enhancement of FFLO pairing is negligible for  $\alpha_1$ -NbSi<sub>2</sub>P<sub>4</sub> monolayer, since the self-consistent solution can be well fitted by the extended model of IBCS pairing over the full temperature range [Fig. 4(e)].

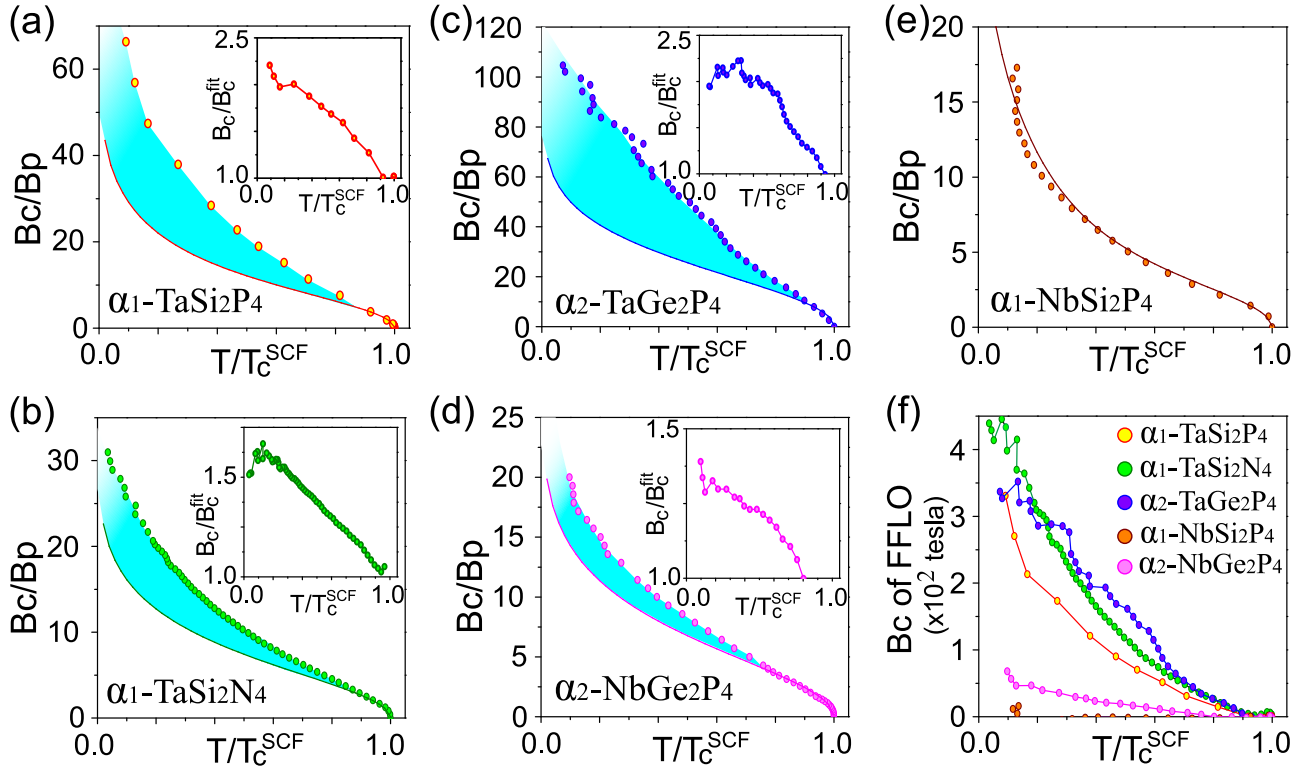


FIG. 4. The relative contribution of IBCS and FFLO pairing on field enhancement. (a)–(e) The dependences of  $B_c/B_p$  on normalized temperature  $T/T_c^{SCF}$  for (a)  $\alpha_1$ -TaSi<sub>2</sub>P<sub>4</sub>, (b)  $\alpha_1$ -TaSi<sub>2</sub>N<sub>4</sub>, (c)  $\alpha_2$ -TaGe<sub>2</sub>P<sub>4</sub>, (d)  $\alpha_2$ -NbGe<sub>2</sub>P<sub>4</sub>, and (e)  $\alpha_1$ -NbSi<sub>2</sub>P<sub>4</sub> monolayer, which are fitted by the extended microscopic model at low field (solid lines). Light blue shaded regions represent the component of critical field enhanced by additional FFLO pairing. The inset shows the value of the self-consistently calculated total critical fields ( $B_c$ ) overlaid with model fitted values ( $B_c^{fit}$ , i.e., contribution of IBCS pairing) at different temperatures. (f) The normalized temperature-dependent field enhancement of FFLO pairing estimated roughly by subtracting the  $B_c^{fit}$  from the total enhancement  $B_c$ .

We quantify the critical field enhanced by FFLO pairing in Fig. 4(f). One sees that the FFLO pairing alone can increase the critical field by more than  $\sim 300$ ,  $\sim 400$ ,  $\sim 300$ ,  $\sim 50$ , and  $\sim 0$  tesla for  $\alpha_1$ -TaSi<sub>2</sub>P<sub>4</sub>,  $\alpha_1$ -TaSi<sub>2</sub>N<sub>4</sub>,  $\alpha_2$ -TaGe<sub>2</sub>P<sub>4</sub>,  $\alpha_2$ -NbGe<sub>2</sub>P<sub>4</sub>, and  $\alpha_1$ -NbSi<sub>2</sub>P<sub>4</sub> monolayers at the 0 K limit, respectively. We argue that the FFLO pairing formed on the “effective 1D FS” associated with the Weyl nodal line is closely related to the strength of Ising SOC quantified by  $\delta_{F\uparrow\downarrow}^\Gamma$  (Table II). SOC that is too weak cannot maintain the “1D FS” under a magnetic field, such as in the case of  $\alpha_1$ -NbSi<sub>2</sub>P<sub>4</sub> with negligible FFLO pairing. This demonstrated the critical role that the 1D Weyl nodal lines play in inducing FFLO pairing in a system with strong SOC.

It is worth noting that even without FFLO pairing, the  $B_c/B_p$  ratios are still higher than most of the experimentally confirmed Ising superconductors except WS<sub>2</sub> [36]. This is likely because the  $MA_2Z_4$  monolayer does not possess CDW instability, nor does it need ionic gating to evoke superconductivity. The novel Weyl-nodal-induced FFLO pairing and the unprecedentedly high critical field in the  $MA_2Z_4$  monolayers may be revealed from field dependence measurements, as was done previously for superconductors with either IBCS or FFLO pairing. We emphasize that the  $MA_2Z_4$  monolayers are the first materials in which the field enhancement is enabled by both IBCS and FFLO pairing simultaneously. Also, exploring the possible topological superconducting state associated with the Weyl nodal lines, such as the one associated with the Sarma state [76], in these Ising

superconductors could be another interesting topic of future study.

### III. CONCLUSION

In summary, we have demonstrated the causal relationship between superconductivity and electron topology by revealing a topological Weyl nodal line induced FFLO pairing in an IBCS superconductor  $MA_2Z_4$  monolayer. The superconducting transition temperatures are predicted to range from a few to tens of degrees Kelvin for the considered  $\alpha_1$ -TaSi<sub>2</sub>P<sub>4</sub>,  $\alpha_1$ -TaSi<sub>2</sub>N<sub>4</sub>,  $\alpha_2$ -TaGe<sub>2</sub>P<sub>4</sub>,  $\alpha_1$ -NbSi<sub>2</sub>P<sub>4</sub>, and  $\alpha_2$ -NbGe<sub>2</sub>P<sub>4</sub> monolayers when there is no external magnetic field. In the presence of an in-plane field, the cooperation of the IBCS and FFLO mechanisms will enable the  $B_c/B_p$  ratio to reach  $\sim 100$ ,  $\sim 70$ ,  $\sim 30$ , and  $\sim 20$  for  $\alpha_2$ -TaGe<sub>2</sub>P<sub>4</sub>,  $\alpha_1$ -TaSi<sub>2</sub>P<sub>4</sub>,  $\alpha_1$ -TaSi<sub>2</sub>N<sub>4</sub>, and  $\alpha_2$ -NbGe<sub>2</sub>P<sub>4</sub> monolayers, respectively. We found that the FFLO pairing condensed on the “effective 1D FS” of the Weyl nodal line is closely related to the strength of SOC, which cannot be maintained under a magnetic field when the SOC is weak, such as in the case of  $\alpha_1$ -NbSi<sub>2</sub>P<sub>4</sub> monolayer with negligible FFLO pairing. Our findings not only enrich the fundamental relationship between superconductivity and electron topology, but they also yield an effective approach to enhance the robustness of superconductivity against a magnetic field.

## ACKNOWLEDGMENTS

F.L. acknowledges financial support from DOE-BES (No. DE-FG02-04ER46148). X.Z. acknowledges financial support by the National Natural Science Foundation of China (No.

12004357), the Natural Science Foundation of Shandong Province (No. ZR2020QA053), and the Young Talents Project at Ocean University of China.

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