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## Unconventional superconductivity in bilayer transition metal dichalcogenides

Chao-Xing Liu<sup>1</sup>

<sup>11</sup>Department of Physics, The Pennsylvania State University, University Park, Pennsylvania 16802-6300, USA; (Dated: January 24, 2017)

Bilayer transition metal dichalcogenides (TMDs) belong to a class of materials with two unique features, the coupled spin-valley-layer degrees of freedom and the crystal structure that is globally centrosymmetric but locally non-centrosymmetric. In this work, we will show that the combination of these two features can lead to a rich phase diagram for unconventional superconductivity, including intra-layer and inter-layer singlet pairings and inter-layer triplet pairings, in bilayer superconducting TMDs. In particular, we predict that the inhomogeneous Fulde-Ferrell-Larkin-Ovchinnikov state can exist in bilayer TMDs under an in-plane magnetic field. We also discuss the experimental relevance of our results and possible experimental signatures.

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Introduction.- Unconventional superconductivity [1-3], which is beyond the simple s-wave spin-singlet superconductivity in the Bardeen-Cooper-Schrieffer theory, can emerge in two dimensional (2D) systems, such as surfaces [4-6] or interfaces [7], superconducting heterostructures [8] and 2D or quasi-2D superconducting materials [9-14]. Recently, it was demonstrated that "Ising" superconductivity can exist in monolayer transition metal dichalcogenides (TMDs), such as MoS<sub>2</sub> [11, 13] and NbSe<sub>2</sub> [12], based on experimental observation that in-plane upper critical field  $H_{c2,\parallel}$  is far beyond the paramagnetic limit. The space symmetry group of the monolayer TMD is the  $D_{3h}$  group without inversion symmetry. Thus, the monolayer superconducting TMDs belong to the so-called non-centrosymmetric superconductors (SCs) [3], for which spin-up and spin-down Fermi surfaces are split by strong spin-orbit coupling (SOC), leading to a mixing of spin singlet and triplet pairings [15, 16]. The existence of triplet components can enhance  $H_{c2,\parallel}$  in non-centrosymmetric SCs [17]. In monolayer TMDs, Ising SOC fixes spin axis along the out-of-plane direction and greatly reduces the Zeeman effect of in-plane magnetic fields, thus explaining the experimental observations of high  $H_{c2,\parallel}$ . A high  $H_{c2,\parallel}$  was also observed in bilayer TMDs (e.g. NbSe<sub>2</sub>) [12]. The crystal structure of bilayer TMDs is described by the symmetry group  $D_{3d}$  with inversion symmetry and the corresponding Fermi surfaces are spin degenerate. This experimental result motivates us to study the difference between bilayer superconducting TMDs and conventional SCs.

We first illustrate the difference from symmetry aspect. Although inversion symmetry exists in bilayer TMDs, the inversion center should be chosen at the center between two layers, labeled by "P" in Fig. 1a. As a result, bilayer TMDs belong to a class of materials which are globally centrosymmetric, but locally non-centrosymmetric (for each layer). The absence of local inversion symmetry can lead to the "hidden" spin polarization [18, 19], the spinlayer locking [20, 21] and other exotic physical phenomena [22]. The superconductivity for these materials has been studied in the CeCoIn<sub>5</sub>/YbCoIn<sub>5</sub> hybrid system [10, 23], SrPtAs [23-26] and other bilayer Rashba systems [27]. Inhomogeneous Fulde-Ferrell-Larkin-Ovchinikov (FFLO) states were

proposed in CeCoIn<sub>5</sub>/YbCoIn<sub>5</sub> hybrid system while chiral topological d + id superconductivity was suggested in SrPtAs. Bilayer TMDs possess global  $D_{3d}$  symmetry and local  $D_{3h}$  symmetry, labeled as  $D_{3d}(D_{3h})$ , and thus it is equivalent to that of SrPtAs [25], but different from CeCoIn<sub>5</sub>/YbCoIn<sub>5</sub> hybrid system with  $D_{3d}(C_{3v})$  symmetry. Due to the  $D_{3h}$  symmetry in each layer, Ising SOC is expected in bilayer TMDs and SrPtAs, while Rashba SOC occurs in CeCoIn<sub>5</sub>/YbCoIn<sub>5</sub> hybrid system.

In this work, we study possible superconducting pairings based on a prototype model of bilayer TMDs. The superconducting phase diagram as a function intra-layer  $(U_0)$  and interlayer  $(V_0)$  interactions is summarized in Fig. 1c, in which three different pairings, intra-layer  $A_{1g}$  pairing, intra-layer  $A_{1u}$ pairing and inter-layer  $E_u$  pairing, can exist, depending on the strength and sign of  $U_0$  and  $V_0$ . We further study the stability of these superconducting pairings under external magnetic fields. In particular, we predict the FFLO state with a finite momentum pairing [28, 29] induced by the orbital effect of in-plane magnetic fields.

Phase diagram of bilayer TMDs - A prototype model for TMDs [15, 16, 30] was first derived for the conduction band of MoS<sub>2</sub> and can also be applied to other TMDs. This model is constructed on a triangle lattice of Mo atoms with  $4d_{r^2}$  orbitals for each monolayer. The conduction band minima appear at two momenta  $\pm K$ , and one can regard  $\pm K$  as valley index and expand the tight-binding model around  $\pm K$  for each layer, as described in Ref. [15, 16]. We extend this model to bilayer TMDs by including layer index. Let us label the annihilation fermion operator as  $c_{\sigma,\eta}$ , where  $\sigma = \uparrow, \downarrow$  is for spin and  $\eta =$  $\pm$  is for two layers. On the basis of  $(c_{\uparrow,+}, c_{\downarrow,+}, c_{\uparrow,-}, c_{\downarrow,-})$ , the effective Hamiltonian is

$$\hat{H}_0(\mathbf{p} = \epsilon \mathbf{K} + \mathbf{k}) = \xi_k + \epsilon \beta_{so} s_z \tau_z + t \tau_x \tag{1}$$

where s and  $\tau$  are two sets of Pauli matrices for spin and layer degrees,  $\epsilon = \pm$  is for valley index and  $\xi_k = \frac{\hbar^2}{2m}k^2 - \mu$  with chemical potential  $\mu$ . Here the  $\beta_{so}$  term is the Ising SOC while the t term is the hybridization between two layers. The eigenenergy is given by  $\varepsilon_{s,\lambda} = \xi_k + \lambda D_0$  with  $D_0 = \sqrt{\beta_{s0}^2 + t^2}$  and  $s, \lambda = \pm$ . s does not appear and thus the eigen-states with opposite s are degenerate, as shown in Fig. 1b. We next con-



FIG. 1. (a) Crystal structure of bilayer TMDs  $MX_2$  with the inversion center labelled by P. (b) Schematics for energy dispersion of bilayer TMDs where red and blue are for spin up and spin down, and solid and dashed lines are for the top and bottom layers. Here each band is doubly degenerate and we shift the dashed lines a little for the view. (c) The phase diagram as a function of  $U_0$  and  $V_0$ . The red, blue and green lines are the phase boundary, separating three superconducting phases, the  $A_{1g}$ ,  $A_{1u}$  and  $E_u$  pairings, and the metallic phase. (d) Experimental setup of bilayer TMD SC/conventional SC junction.

sider the symmetry classification of superconducting pairings, similar to that in Cu doped Bi<sub>2</sub>Se<sub>3</sub> SCs [31] since both materials belong to  $D_{3d}$  group. We only consider s-wave pairing, and thus the gap function  $\hat{\Delta}$  is independent of momentum and can be expanded in terms of s and  $\tau$  ( $\hat{\Delta} = \sum_{i,\mu} \Delta_{i,\mu} \gamma_{i,\mu}$ where  $\gamma_{i,\mu}$  is a 4 × 4 matrix composed of s and  $\tau$  and  $i,\mu$ are the indices labelling different representations). Due to anti-commutation relation between fermion operators, the gap function needs to be anti-symmetric, and thus only six matrices  $s_y$ ,  $s_y\tau_x$ ,  $s_y\tau_z$ ,  $\tau_y$ ,  $s_x\tau_y$ ,  $s_z\tau_y$  can couple to s-wave pairing. The classification of these representation matrices, as well as their explicit physical meanings, are listed in the Table I, from which  $\Delta_{A_{1g},1}$  and  $\Delta_{A_{1u}}$  describe intra-layer singlet pairings,  $\Delta_{A_{1g},2}$  and  $\Delta_{A_{2u}}$  give inter-layer singlet pairings while  $\Delta_{E_u,1}$ and  $\Delta_{E_u,2}$  are inter-layer triplet pairings. The pairing interaction can also be decomposed into different pairing channels as  $V_{A_{1g},1} = V_{A_{1u}} = \frac{U_0}{2}$  and  $V_{A_{1g},2} = V_{A_{2u}} = V_{E_u,1} = V_{E_u,2} = \frac{V_0}{2}$ (See appendix for details).

Possible superconducting pairings are studied based on the linearized gap equations [1–3] (See appendix). Around the valley K (or -K), the Fermi surfaces for two spin states in each layer are well separated by Ising SOC  $\beta_{so}$  term. Therefore, we below assume the Fermi energy only crosses the lower energy band at each valley (Fig. 1b), for simplicity. The pairings with different representations do not couple to each other and thus, we can compute the critical temperature  $T_c$  in each representation, separately. The critical temperature normally takes the form  $kT_{c0,i} = \frac{2\gamma\omega_D}{\pi} exp\left(-\frac{1}{N_0 V_{ieff}}\right)$ , with the representation index *i*, density of states  $N_0$ , the Debye frequency  $\omega_D$  and  $\gamma \approx 1.77$ . The effective interaction is given by

TABLE I. The matrix form and the explicit phyiscal meaning of Cooper pairs in the representations  $A_{1g}$ ,  $A_{1u}$ ,  $A_{2u}$  and  $E_u$  of the  $D_{3d}$  group. Here  $c_{\sigma\eta}$  is electron operator with  $\eta = \pm$  for layer index  $\sigma$  for spin. *s* and  $\tau$  are Pauli matrices for spin and layer.

Representation	Matrix form	Explicit form
$A_1 \cdot \Delta_{A_{1g},1}$	$s_y$	$c_{\uparrow +}c_{\downarrow +}+c_{\uparrow -}c_{\downarrow -}$
$\Delta_{A_{1g},2}$	$s_y \tau_x$	$c_{\uparrow +}c_{\downarrow -}+c_{\uparrow -}c_{\downarrow +}$
$A_{1u}$ : $\Delta_{A_{1u}}$	$s_y \tau_z$	$c_{\uparrow +}c_{\downarrow +}-c_{\uparrow -}c_{\downarrow -}$
$A_{2u}$ : $\Delta_{A_{2u}}$	$s_x \tau_y$	$c_{\uparrow +}c_{\downarrow -}-c_{\uparrow -}c_{\downarrow +}$
$E \cdot \Delta_{E_u,1}$	$ au_y$	$C_{\uparrow +} C_{\uparrow -}$
$\Delta_{E_u,2}$	$s_z  au_y$	$C_{\downarrow +} C_{\downarrow -}$

 $V_{A_{1g},eff} = 2U_0 + 2V_0 \frac{t^2}{D_0^2}$  for the  $A_{1g}$  pairing,  $V_{A_{1u},eff} = 2U_0 \frac{\beta_{so}^2}{D_0^2}$ for the  $A_{1u}$  pairing and  $V_{E_u,eff} = 2V_0 \frac{\beta_{so}^2}{D_c^2}$  for the  $E_u$  pairing, from which the corresponding critical temperature in each channel can be determined. The  $A_{2u}$  pairing does not exist because  $V_{A_{2u},eff} = 0$ . The phase diagram can be extracted by comparing different  $T_{c0,i}$  (Fig. 1c). The  $A_{1g}$  pairing is favored by strong attractive intra-layer interaction ( $U_0 > 0$ ), while the  $E_u$  pairing is favored by strong attractive inter-layer interaction ( $V_0 > 0$ ). These two phases are separated by the critical line  $U_0 = \frac{\beta_{xo}^2 - t^2}{D_z^2} V_0$ . The  $A_{1u}$  pairing appears when the repulsive inter-layer interaction is stronger than the attractive intralayer interaction  $(-V_0 > U_0 > 0)$  because repulsive inter-layer interaction will favor opposite phases of pairing functions between two layers. The  $A_{1u}$  phase is separated from the  $A_{1g}$ phase by a critical line  $U_0 = -V_0$ . When both  $U_0$  and  $V_0$  are repulsive interaction  $(U_0, V_0 < 0)$ , no superconductivity can exist. For the 2D  $E_u$  pairing,  $\Delta_{E_u,1}$  and  $\Delta_{E_u,2}$  are degenerate. By taking into account the fourth order term in the Landau free energy (See Appendix), either nematic superconductivity  $(\Delta_{E_u,1}, \Delta_{E_u,2}) = \Delta_{E_u}(\cos\theta, \sin\theta)$  ( $\theta$  is a constant) [32] or chiral superconductivity with  $(\Delta_{E_u,1}, \Delta_{E_u,2}) = \Delta_{E_u}(1, i)$  can be stabilized[33].

*Magnetic field effect* – Next we study the effect of magnetic fields on bilayer superconducting TMDs. Generally, magnetic fields have two effects, the Zeeman effect and the orbital effect. The Zeeman coupling is given by

$$\hat{H}_{Zee} = g\mathbf{B} \cdot \mathbf{s} \tag{2}$$

where **B** labels the magnetic field and the Bohr magneton is absorbed into *g* factor. The orbital effect is normally included by replacing the momentum **k** in  $\xi_k$  with the canonical momentum  $\pi = \mathbf{k} + \frac{e}{\hbar}\mathbf{A}$  with vector potential **A** (Peierls substitution). The orbital effect of in-plane magnetic fields is normally not important for a quasi-2D system. However, it is not the case in bilayer TMDs due to its unusual band structure. Let's choose  $\mathbf{A} = (0, -B_x z, 0)$  for the in-plane magnetic field  $B_x$ , in which the origin z = 0 is located at the center between two layers. As a result,  $\xi_k$  is changed to  $\xi_{\pi} = \frac{\hbar^2}{2m} (k_x^2 + (k_y - \frac{eB_x z_0}{2\hbar} \tau_z)^2) - \mu$ after the substitution, where  $z_0$  is the distance between two layers. The Ginzburg-Landau free energy is constructed as

$$L = \frac{1}{2} \sum_{\mathbf{q},i\mu} \Delta_{i,\mu}^*(\mathbf{q}) \left( \frac{1}{V_i} \delta_{ij} \delta_{\mu\nu} - \chi_{ij,\mu\nu}^{(2)}(\mathbf{q},\mathbf{B}) \right) \Delta_{j,\nu}(\mathbf{q}) + L_4,$$
(3)

where  $L_4$  describes the fourth order term. The superconductivity susceptibility  $\chi_{ij,\mu\nu}^{(2)}$  can be expanded up to the second order of **q** and **B** ( $q_iq_j$ ,  $B_iB_j$  and  $q_iB_j$  with i, j = x, y, z). The magnetic field correction to  $T_{c0,i}$  for different pairings can be extracted by minimizing the above free energy (See appendix).

Due to the orbital effect, the Hamiltonian (1) is changed to

$$\hat{H}'_0 = \xi_k - \hbar v_Q k_y \tau_z + \epsilon \beta_{so} s_z \tau_z + t \tau_x, \tag{4}$$

where  $v_Q = \frac{eB_x z_0}{2m}$  and the chemical potential  $\mu$  in  $\xi_k$  is redefined to include the  $B_x^2$  term. We first focus on the limit  $t \rightarrow 0$ , in which the energy dispersion of the Hamiltonian (4) is shown in Fig. 2a. The energy bands on the top and bottom layers are shifted in the opposite directions in the momentum space by  $Q = \frac{eB_x z_0}{2\hbar}$ . This momentum shift cannot be "gauged away" and thus the intra-layer spin-singlet pairing must carry a non-zero total momentum. This immediately suggests the possibility of the FFLO state [28, 29, 34] for the intra-layer singlet  $A_{1g}$  and  $A_{1u}$  pairings. Since in-plane magnetic fields break the  $D_{3d}$  symmetry, the orbital effect can mix the singlet  $A_{1g}$  and  $A_{1u}$  pairings. In the limit  $t \rightarrow 0$  with  $T_{c0,A_{1g}} = T_{c0,A_{1u}} =$  $T_{c0}$ , we derive the free energy for the coupled  $A_{1g}$  and  $A_{1u}$ pairings as

$$L_{2} = \frac{1}{2} \sum_{\mathbf{q}} \left[ \left( 4N_{0}ln\left(\frac{T}{T_{c0}}\right) - \mathcal{P}(h_{x},\mathbf{q}) \right) \sum_{i=A_{1g},A_{1u}} |\Delta_{i}|^{2} -\Delta_{A_{1g}}^{*} \mathcal{Q}\Delta_{A_{1u}} - \Delta_{A_{1u}}^{*} \mathcal{Q}\Delta_{A_{1g}} \right],$$
(5)

in which the detailed form of  $\mathcal{P}$  and Q are defined in Appendix. The term  $Q = \tilde{K}B_xq_y$  with a constant  $\tilde{K}$  mixes  $A_{1g}$  and  $A_{1u}$  pairings. With a transformation  $\Delta_{\pm} = \frac{1}{\sqrt{2}} \left( \Delta_{A_{1g}} \pm \Delta_{A_{1u}} \right)$ , the free energy is changed to

$$L_2 = \frac{1}{2} \sum_{\alpha=\pm,\mathbf{q}} \left( 4N_0 ln\left(\frac{T}{T_{c0}}\right) - \mathcal{P}(B_x,\mathbf{q}) - \alpha \mathcal{Q}(B_x,\mathbf{q}) \right) |\Delta_\alpha|^2 (6)$$

The corresponding critical temperature is determined by maximizing  $ln\left(\frac{T_c}{T_{c0}}\right) = \frac{1}{4N_0} \left(\mathcal{P}(B_x, \mathbf{q}) + \alpha Q(B_x, \mathbf{q})\right)$  with respect to  $\mathbf{q}$  and  $\alpha$ . From the explicit form of  $\mathcal{P}$  and Q, the maximum is achieved by  $q_x = 0$  and  $|q_y| = q_c = \frac{eB_x z_0}{\hbar} = 2Q$ , thus realizing the FFLO state. The corresponding correction to  $T_c$  vanishes  $(T_c = T_{c0})$ . As a comparison, the  $T_c$ of zero momentum pairing decreases with magnetic fields as  $ln\left(\frac{T_c(\mathbf{q}=0)}{T_{c0}}\right) = -C\left(\frac{\hbar v_Q k_f}{2\pi k T}\right)^2 \propto -B_x^2$  and the FFLO state is always favored in the limit  $t \to 0$  for in-plane magnetic fields.

The form of the stable pairing function depends on the sign of Q. Let's assume  $B_x > 0$  and  $\tilde{K} > 0$  in  $Q = \tilde{K}B_xq_y$ . If  $q_y = q_c > 0$ , Q > 0 and thus  $\Delta_+$  pairing is favored. If  $q_y = -q_c < 0$ , Q < 0 and  $\Delta_-$  is favored.  $\Delta_+(q_c)$  and  $\Delta_-(-q_c)$  are degenerate for the second order term of free energy. The FFLO state in the real space is

$$\Delta(\mathbf{r}) = \Delta_+(q_c)e^{iq_c y} + \Delta_-(-q_c)e^{-iq_c y}.$$
(7)

The exact form of pairing function is determined by the fourth order term of  $\Delta_+(q_c)$  and  $\Delta_-(-q_c)$ , which is phenomenologically given by

$$L_{4} = \mathcal{B}_{s} \left( |\Delta_{+}(q_{c})|^{2} + |\Delta_{-}(-q_{c})|^{2} \right)^{2} + \mathcal{B}_{a} \left( |\Delta_{+}(q_{c})|^{2} - |\Delta_{-}(-q_{c})|^{2} \right)^{2}$$
(8)

If  $\mathcal{B}_a > 0$ , we need  $|\Delta_+(q_c)| = |\Delta_-(-q_c)| = \Delta_0$  to minimize  $L_4$ . This state is known as LO phase [28, 35] or stripe phase [4, 6, 8, 36] or pair density wave [10, 37, 38]. If  $\mathcal{B}_a < 0$ , we have either  $\Delta_+(q_c) = 0$  or  $\Delta_-(-q_c) = 0$ . In either case, the amplitude of  $\Delta(\mathbf{r})$  is fixed while its phase oscillates, thus correponding to FF phase [29, 35] or helical phase [3, 36, 39–41]. In the limit  $t \to 0$ , the coefficients are computed as  $\mathcal{B}_s = \mathcal{B}_a = \frac{7N_0\zeta(3)}{16(\pi kT_{c_0})^2} > 0$ . Therefore, the stripe phase will be favored under an in-plane magnetic field near the critical temperature.

In the limit  $t \to 0$ ,  $\Delta_+$  and  $\Delta_-$  are just the singlet pairing on the top and bottom layers according to Table I, and the free energies for  $\Delta_+$  and  $\Delta_-$  become decoupled (see Eq. (6) for  $L_2$  term and Eq. (96) of the appendix for  $L_4$  term). Thus, the FFLO state in Eq. (7) can be viewed as two independent helical phases in two separate layers. No supercurrent or other observables can exist in helical phases [39, 40] for infinite large systems. To identify this phase, one needs to consider a Josephson junction structure between bilayer TMDs and conventional SCs (Fig. 1d), similar to that discussed in Ref. [3, 40, 42] (See appendix for details). For a finite tunneling *t*, the interference between two layers leads to the gap oscillation of stripe phase in Eq. (7).

We notice that the FFLO phase has been proposed in noncentrosymmetric SCs under a magnetic field [6, 23], and emphasize two essential differences between our case and noncentrosymmetric SCs. (1) In non-centrosymmetric SCs, the FFLO phase is induced by a linear gradient term  $\tilde{K}_{ij}\Delta^* B_i q_j \Delta$ ( $\tilde{K}_{ij}$  is a parameter) that breaks inversion symmetry. In contrast, inversion symmetry is preserved in our system, and the linear gradient term ( $\tilde{K}_{ij}\Delta^*_{A_{1g}}B_iq_j\Delta_{A_{1u}}$ ) couples two pairings with opposite parities. (2) In non-centrosymmetric SCs, the FFLO phase results from the combination of Rashba SOC and Zeeman effect of magnetic fields. In our system, the FFLO phase is from the combination of Ising SOC and the orbital effect of magnetic fields. In particular, this phase can occur for any magnetic field strength in the weak interlayer coupling limit  $t \rightarrow 0$ .

When  $t \neq 0$ , the occurence of the FFLO phase will be shifted to a finite magnetic field. We numerically minimize free energy with respect to the momentum **q** and calculate the magnetic field correction to  $T_c$ . In Fig. 2b,  $T_c/T_{c0}$  is plotted as a function of magnetic field  $B_x$  for three hybridization parameters t. The momenta for the corresponding stable states, labeled by  $q_c$ , are shown in Fig. 2c. For a weak hybridization  $(t = 1 \text{meV} \ll \beta_{so} = 40 \text{meV})$ , FFLO phase appears at a small  $B_x$ , and the corresponding  $q_c$  approaches 2Q with increasing  $B_x$ . There is only a weak correction to  $T_c$  for the FFLO phase (black line in Fig. 2b). When increasing hybridization (t = 5, 10 meV), zero momentum pairing is favored for small



FIG. 2. (a) Schematics of energy dispersion for bilayer TMDs with an in-plane magnetic field. Here red and blue colors are for opposite spins and solid and dashed lines are for top and bottom layers. (b) The magnetic field dependence of the critical temperature  $T_c$ . Here the black line is for t = 1meV, the red is for t = 5meV while the blue is for t = 10meV. Other parameters are chosen as  $\beta_{so} = 40meV$ ,  $\hbar v_F = 30meV \cdot nm$  and  $m = 0.6m_e$  with electron mass  $m_e$ ,  $N_0U_0 = 0.3$ and  $N_0V_0 = 0.1$ . Only the orbital effect is taken into account. (c) The momentum  $q_c$  for the stable pairing state as a function of  $B_x$ . (d) Phase diagram as a function of  $B_x$  and  $T_c$ . Here I is for conventional SC phase, II is for FFLO state and III is for normal metal.  $B_N = \frac{2KT_{c0}}{M(r)}$ .

 $B_x$  and lead to a rapid decrease of  $T_c$  with its correction given by  $\frac{T_c - T_{c0}}{T_{c0}} \propto -B_x^2$  (red and blue lines in Fig. 2b). When  $B_x$ becomes larger, a transition from zero momentum pairing to the FFLO state occurs. The decreasing in  $T_c$  deviates from the  $B_r^2$  dependence and becomes weaker. Experimentally, one can control the hybridization between two layers by inserting an insulating layer in between, and the deviation of the  $T_c$  correction from the  $B_x^2$  dependence implies the occurrence of FFLO states in this system. We further construct the phase diagram by evaluating gap functions as a function of temperatures and magnetic fields for t = 10meV in Fig. 2d. As discussed in appendix, The transition from the normal metal (III region in Fig. 2d) to uniform SC (I region) or FFLO state (II region) is of the second order type (dashed red line in Fig. 2d) while the transition between uniform SC and FFLO state is of the first order type (dashed black line in Fig. 2d).

Besides the orbital effect, the correction of  $T_c$  due to the Zeeman effect, which is the same for zero-momentum pairing and the FFLO phase, is given by  $ln\left(\frac{T_{cA_{1g}}}{T_{c0A_{1g}}}\right) \propto -\frac{t^2}{\beta_{so}^2}B_x^2$  for  $A_{1g}$  pairing and  $ln\left(\frac{T_{cA_{1g}}}{T_{c0A_{1g}}}\right) \propto -\frac{t^4}{\beta_{so}^4}B_x^2$  for  $A_{1u}$  pairing. Additional factors  $t^2/\beta_{so}^2$  and  $t^4/\beta_{so}^4$  greatly reduce the  $B_x^2$  dependence for the  $A_{1g}$  and  $A_{1u}$  pairings in the limit  $t \ll \beta_{so}$ . The behavior of out-of-plane magnetic field  $(B_z)$  in bilayer TMDs is similar to that of conventional SCs (See Appendix).

*Discussion and Conclusion* – In realistic bilayer superconducting TMDs, the Fermi energy will cross both spin states in

each layer. However, once the Ising SOC is larger than other energy scales ( $\beta_{so} \gg t, \hbar k_f v_O, \hbar v_f q$ ), the Fermi surfaces for two spin states in one layer are well separated and the physics discussed here should be valid qualitatively. Based on the existing experiments, the  $A_{1g}$  pairing is mostly likely to exist at a zero magnetic field. In this case, we predict the occurence of the FFLO phase under an in-plane magnetic field. The onset magnetic field is determined by the ratio between inter-layer hybridization t and Ising SOC  $\beta_{so}$  ( $\frac{t}{\beta_{so}} \sim 0.27$  in NbSe<sub>2</sub>) [12]. Our results suggest a weak correction to  $T_c$  for both the orbital and Zeeman effects of in-plane magnetic fields, thus consistent with experimental observations of high in-plane critical fields in bilayer superconducting TMDs [12]. The central physics in this work originates from the unique crystal symmetry property, and similar physics can occur in SrPtAs [25]. Similar physics also occurring for exciton condensate in a bilayer system [43, 44]. Our work paves a new avenue to search for unconventional superconductivity in 2D centrosymmetric SCs.

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