Unconventional superconductivity in bilayer transition metal dichalcogenides

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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\[7,8\], superconducting heterostructures\[9,10\] and 2D or quasi-2D superconducting materials\[11-13\]. Recently, it was demonstrated that “Ising” superconductivity can exist in monolayer transition metal dichalcogenides (TMDs), such as MoSe\(_2\)\[11,13\] and NbSe\(_2\)\[12\], 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)\[1,2\], 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\[14-16\]. 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\(_2\))\[18\]. The crystal structure of bilayer TMDs is described by the symmetry group \(D_{3h}\) 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). Thus, bilayer TMDs and conventional SCs.

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 \(A_{1u}\), \(A_{1g}\) and \(E_{c}\) pairings, and the metallic phase. (d) Experimental setup of bilayer TMD SC/conventional SC junction.
TMDs\textsuperscript{15,16,30} was first derived for the conduction band of MoS\textsubscript{2} and can also be applied to other TMDs. This model is constructed on a triangle lattice of Mo atoms with 4\textit{d}_{\text{z}^2} orbitals for each monolayer. The conduction band minima appear at two momenta ±\textit{K}, and one can regard ±\textit{K} as valley index and expand the tight-binding model around ±\textit{K} for each layer, as described in Ref.\textsuperscript{15,16}. We extend this model to bilayer TMDs by including layer index. Let us label the annihilation fermion operator as \( \epsilon_{s\tau} \), where \( s = \pi, \downarrow \) is for spin and \( \eta = \pm \) is for two layers. On the basis of \((\epsilon_{1\pi+}, \epsilon_{1\pi-}, \epsilon_{1\downarrow+}, \epsilon_{1\downarrow-})\), the effective Hamiltonian is

\[
\hat{H}_0(p = \epsilon \mathbf{K} + \mathbf{k}) = \xi_k + \epsilon \beta_{\sigma\lambda} s_{\sigma\lambda} + \tau_{\sigma\lambda} \tag{1}
\]

where \( s \) and \( \tau \) are two sets of Pauli matrices for spin and layer degrees, \( \epsilon = \pm \) is for valley index and \( \xi_k = \frac{\hbar^2 k^2}{2m} - \mu \) with chemical potential \( \mu \). Here the \( \beta_{\sigma\lambda} \) term is the Ising SOC while the \( t \) term is the hybridization between two layers. The eigen-energy is given by \( e_{s\tau} = \xi_k + \Delta_0 \) with \( \Delta_0 = \sqrt{\beta_{\sigma\lambda}^2 + t^2} \) 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 consider the symmetry classification of superconducting pairings, similar to that in Cu doped Bi\textsubscript{x}Se\textsubscript{2} Scs\textsuperscript{31} since both materials belong to D\textsubscript{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 \( \tau \) (\( \hat{\Delta} = \sum_{s\tau} \Delta_{s\tau} y_{s\tau} \) where \( y_{s\tau} \) is a \( 4 \times 4 \) 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 \( \xi_k \), \( \xi_{ks\tau} \), \( \xi_{s\tau} \), \( \xi_{s\tau} \), \( \xi_{s\tau} \), \( \xi_{s\tau} \) 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 \( \Delta_{A_{1g}} \) and \( \Delta_{A_{2g}} \) describe intra-layer singlet pairings, \( \Delta_{A_{1u}} \) and \( \Delta_{A_{2u}} \) give inter-layer singlet pairings while \( \Delta_{E_{1u}} \) and \( \Delta_{E_{2u}} \) are inter-layer triplet pairings. The pairing interaction can also be decomposed into different pairing channels as \( V_{A_{1g},1} = V_{A_{1u}} = \frac{\hbar}{2m} \) and \( V_{A_{1g},2} = V_{A_{2u}} = V_{E_{1u}} = V_{E_{2u}} = \frac{\hbar}{2m} \) (See appendix for details).

Possible superconducting pairings are studied based on the linearized gap equations\textsuperscript{32,33} (See appendix). Around the valley \( K \) (or \(-K\)), the Fermi surfaces for two spin states in each layer are well separated by Ising SOC \( \beta_{\sigma\lambda} \) 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 \( kT_c = \frac{2\mu}{\pi T}\exp(-\frac{\epsilon_{k}^{\text{eff}}}{k_{B}T}) \), 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 \( V_{A_{1g},1}^{\text{eff}} = 2U_{0} + 2V_{0} \frac{\hbar^{2}}{2m} \) for the \( A_{1g} \) pairing, \( V_{A_{1u},1}^{\text{eff}} = 2U_{0} \frac{\hbar^{2}}{2m} \) for the \( A_{1u} \) pairing and \( V_{E_{1u},1}^{\text{eff}} = 2V_{0} \frac{\hbar^{2}}{2m} \) 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 \( V_{A_{2u},1}^{\text{eff}} = 0 \). The phase diagram can be extracted by comparing different \( T_{c,i} \) (Fig. 1c). The \( A_{1g} \) pairing is favored by strong attractive in-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 \( U_{0} = \frac{\hbar^{2}}{5m} W \). 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, \( \Delta_{E_{1u}} \) and \( \Delta_{E_{2u}} \) are degenerate. By taking into account the fourth order term in the Landau free energy (See Appendix), either nematic superconductivity (\( \Delta_{E_{1u}}, \Delta_{E_{2u}} = \Delta_{E_{1u}}(\cos \theta, \sin \theta) \) (\( \theta \) is a constant)\textsuperscript{34} or chiral superconductivity with \( \Delta_{E_{1u}}, \Delta_{E_{2u}} = \Delta_{E_{1u}}(1, i) \) can be stabilized\textsuperscript{35}.

### 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

\[
\hat{H}_{\text{Zeeman}} = g\mathbf{B} \cdot \mathbf{s} \tag{2}
\]

where \( \mathbf{B} \) 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 \( k = \mathbf{k} + \frac{\hbar}{2m} \mathbf{A} \) with vector potential \( \mathbf{A} \) (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 \( \mathbf{A} = (0, -B_{z\text{c}}, 0) \) for the in-plane magnetic field \( B_{z\text{c}} \), in which the origin \( z = 0 \) is located at the center between two layers. As a result, \( \xi_k \) is changed to \( \xi_{k} = \frac{\hbar^{2}}{2m} (k^{2} + (k_{z} - eB_{z\text{c}}/2m\tau_{z})^{2} - \mu \) after the substitution, where \( z_{0} \) is the distance between two layers.

The Ginzburg-Landau free energy is constructed as

\[
\mathcal{L} = \frac{1}{2} \sum_{\mathbf{q} \mu} \Delta_{\mu}^{*}(\mathbf{q}) \left( \frac{1}{V_{i}} \delta_{ij} \beta_{\mu\nu} - \chi_{ij,\mu\nu}(\mathbf{q}, \mathbf{B}) \right) \Delta_{\nu}(\mathbf{q}) + L_{4}, \tag{3}
\]

where \( L_{4} \) describes the fourth order term. The superconductivity susceptibility \( \chi_{ij,\mu\nu} \) can be expanded up to the second order of \( \mathbf{q} \) and \( \mathbf{B} \) (\( q_{i} q_{j}, B_{i} B_{j} \) and \( q_{i} B_{j} \) with \( i, j = x, y, z \)). The mag-

### Table I: The matrix form and the explicit physical meaning of Cooper pairs in the representations \( A_{1g}, A_{1u}, A_{2u} \) and \( E_{u} \) of the D\textsubscript{3d} group. Here \( c_{\sigma\tau} \) is electron operator with \( \eta = \pm \) for layer index \( \sigma \) for spin. \( s \) and \( \tau \) are Pauli matrices for spin and layer.
netic field correction to $T_{cij}$ for different pairings can be extracted by minimizing the above free energy (See appendix).

Due to the orbital effect, the Hamiltonian \( H \) is changed to

\[
\hat{H}_0 = \xi_k - h v_Q k_y + \epsilon_B s_y \tau_z + \tau_x,
\]

where $v_Q = \frac{e A}{\hbar c}$ and the chemical potential $\mu$ in $\xi_k$ is redefined to include the $B^2$ term. We first focus on the limit $t \to 0$, in which the energy dispersion of the Hamiltonian \( \hat{H}_0 \) 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 $Q = \frac{2 \pi n_s}{a}$. 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\(^{28,29,35}\) for the intra-layer singlet $A_{1g}$ and $A_{1u}$ pairings. Since in-plane magnetic fields break the centrosymmetric SCs. (1) In non-centrosymmetric SCs, the FFLO phase is induced by a linear gradient term $\tilde{K}_{ij} \Delta^* B_{ij} \Delta$ ($\tilde{K}_{ij}$ is a parameter) that breaks inversion symmetry. In contrast, inversion symmetry is preserved in our system, and the linear gradient term ($\tilde{K}_{ij} \Delta^* A_{ij} \Delta$) 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 $t \to 0$.

When $t \neq 0$, the occurrence 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, $T_c/T_{c,0}$ is plotted as a function of magnetic field $B_z$ for three hybridization parameters $t$. The momenta for the corresponding stable states, labeled by $q_s$, 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 $B_z$, and the corresponding $q_s$ approaches $Q$ with increasing $B_z$. There is only a weak correction to $T_c$ for the FFLO phase (black line in Fig. 2b). When increasing hybridization ($t = 5, 10 \text{meV}$), zero momentum pairing is favored for small $B_z$, and lead to a rapid decrease of $T_c$, with its correction given by $T_c \sim -B_z^2$ (red and blue lines in Fig. 2b). When $B_z$ becomes larger, a transition from zero momentum pairing to the FFLO state occurs. The decreasing in $T_c$ deviates from the $B_z^2$ 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 $B_z^2$ dependence implies the occurrence of FFLO states in this system. We further construct the phase diagram
magnetic fields for
In the path integral formula,
where the action is given by
where the
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
transition between uniform SC and FFLO state is of the first
order type (dashed black line in Fig. 2d). As discussed in
existing experiments, the
Ising SOCs is larger than other
energy scales ($\beta_{so} \gg t, \hbar k_f V_Q, \hbar v_f q$), 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 ex-
isting experiments, the $A_{1g}$ pairing is mostly likely to exist at
a zero magnetic field. In this case, we predict the occurrence 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 $\beta_{so}$ ($t/\beta_{so} \sim 0.27$ in NbSe$_2$).42
Our results suggest a weak correction to $T_c$. For both the orbital
and Zeeman effects of in-plane magnetic fields, thus consist-
ent with experimental observations of high in-plane crit-
ical fields in bilayer superconducting TMDs.12 The central
physics in this work originates from the unique crystal sym-
metry property, and similar physics can occur in SrPtAs.26
Similar physics also occurring for exciton condensate in a bi-
layer system.27-28 Our work paves a new avenue to search for
unconventional superconductivity in 2D centrosymmetric
SCs.

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Appendix A: Landau-Ginzburg free energy and linearized self-consistent gap equation

In this section, we review the formalism for Landau-Ginzburg free energy and linearized self-consistent gap equation23,97, which will be used in the main text. We may start from the interacting Hamiltonian

$$H = H_0 + H_V = \sum_{\mathbf{p}', \alpha \beta} \tilde{c}_{\mathbf{p}'}^\dagger (H_0)_{\alpha \beta} \tilde{c}_{\mathbf{p}' \beta} + \frac{1}{2} \sum_{\mathbf{p}, \mathbf{q}, \alpha \beta \gamma} V_{\alpha \beta \gamma \delta}(\mathbf{p}, \mathbf{p}', \mathbf{q}) \tilde{c}_{\mathbf{p} \alpha}^\dagger \tilde{c}_{\mathbf{p} \alpha} \tilde{c}_{\mathbf{p}' \beta}^\dagger \tilde{c}_{\mathbf{p}' \beta} \tilde{c}_{\mathbf{p} \gamma} \tilde{c}_{\mathbf{p} \gamma},$$

where the $H_0$ term is for single-particle Hamiltonian and the $V$ term is for interaction. We may consider the path integral formalism of the BdG Hamiltonian in the imaginary time, given by

$$Z = \int \mathcal{D} \psi \mathcal{D} \bar{\psi} e^{-S},$$

where the action is given by

$$S = \int_0^\beta d\tau \left( \sum_{\mathbf{p}, \alpha} \bar{\psi}_{\mathbf{p} \alpha} (\partial_\tau - \mu) \psi_{\mathbf{p} \alpha} - \hat{H}(\mathbf{c}, \bar{\mathbf{c}}) \right).$$

In the path integral formula, $c$ and $\bar{c}$ are the Grassmann field for the operators $c$ and $\bar{c}$ with $\bar{c}(\beta) = -\bar{c}(0)$ and $c(\beta) = c(0)$.
Let us assume the symmetry group for the single-particle Hamiltonian $H_0$ as $G$ and we can decompose the interaction term into the representation matrices (denoted as $\gamma_{ij}$, of the group $G$, where $i$ labels the representation and $\mu$ labels the dimension of the representation $i$. Let's define

\[
\tilde{B}_{ij}(q) = \sum_{p, \alpha, \beta} \tilde{c}_{\frac{q}{2}+p, \alpha}(\gamma_{ij}(p))_{\alpha \beta} \tilde{c}_{\frac{q}{2}+p, \beta},
\]

\[
B_{ij}(q) = \sum_{p, \alpha, \beta} c_{\frac{q}{2}+p, \alpha}(\gamma_{ij}^v(p))_{\alpha \beta} c_{\frac{q}{2}+p, \beta},
\]

\[
V_{ij}(p, p', q) = -V_i(q)(\gamma_{ij}(p))_{\alpha \beta} (\gamma_{ij}^v(p'))_{\alpha \beta},
\]

where the minus sign for $V_{ij}(p, p', q)$ keeps $V_i(q) > 0$ for attractive interactions. The interaction term can be written as

\[
H_V = \frac{-1}{2} \sum_{q, \mu} V_i(q) \tilde{B}_{ij}(q) B_{ij}(q).
\]

Since

\[
\exp \left\{ \int \frac{d\tau}{2} \sum_{q, \mu} V_i(q) \tilde{B}_{ij}(q) B_{ij}(q) \right\} = \frac{1}{Z_0} \int D\Delta^* D\Delta \exp \left\{ -\frac{1}{2} \int d\tau \sum_{q, \mu} \left( \frac{1}{V_i} \Delta_{ij}(q) + \tilde{B}_{ij}(q) B_{ij}(q) \right) \right\}
\]

with $Z_0 = \int D\Delta^* D\Delta \exp \left\{ -\int d\tau \sum_{q, \mu} \Delta_{ij}(q) \right\}$, we may apply Hubbard-Stratonovich transformation to the above action and obtain

\[
Z = \frac{1}{Z_0} \int D\Delta^* D\Delta D\tilde{\sigma} C D\tilde{c} \exp \left\{ -\frac{1}{2} \int d\tau \left( \sum_{p, \alpha, \beta} \tilde{c}_{\alpha \beta}((\partial_{\mu} - \mu)\delta_{\alpha \beta} + (H_0)_{\alpha \beta})c_{\alpha \beta} + \frac{1}{2} \sum_{q, \mu} \left( \frac{1}{V_i} \Delta_{ij}(q) + \tilde{B}_{ij}(q) B_{ij}(q) \right) \right) \right\},
\]

where $\Delta_{ij}$ is superconducting order parameter (gap function). This action only contain fermion bilinear terms and thus we can integrate out the electrons $c$ and $\tilde{c}$. We write the resulting path integral as $Z = \int D\Delta^* D\Delta \exp \left\{ -S_{\text{eff}} \right\}$ with the effective action $S_{\text{eff}} = \int d\tau L_{\text{eff}}$. Here the Lagrangian is expand as a function of the order parameters $\Delta$ and $\Delta^*$ with the form

\[
L_{\text{eff}} = L_2 + L_4,
\]

where $L_2$ is for the second order term and $L_4$ is for the fourth order term. The second order term $L_2$ is given by

\[
L_2 = \frac{1}{2} \sum_{q, \mu} \frac{1}{V_i} \Delta_{ij}(q) \Delta_{ij}(q) - \frac{1}{2} \sum_{q, \mu, j'} \Delta_{ij}(q) \chi_{ij}(q) \Delta_{ij}(q),
\]

where superconductivity susceptibility $\chi^{(2)}$ is given by

\[
\chi_{ij}(q) = -\frac{1}{\beta} \sum_{j, \mu, \nu} \text{Tr} \left( \gamma_{ij}(q) G_{v}(p + \frac{q}{2}, i\omega_n) G_{v}(p - \frac{q}{2}, i\omega_n) \right).
\]

Here the single-particle Green function is defined as $G_{v}(p, i\omega_n) = (i\omega_n - H_0(p))^{-1}$ for electrons and $G_{h}(p, i\omega_n) = (i\omega_n + H_0(-p))^{-1}$ for holes for our interacting Hamiltonian in the Matsubara frequency space. The fourth order term $L_4$ is given by

\[
L_4 = \frac{1}{4} \sum \Delta_{ij}(q_1) \Delta_{ij}(q_2) \chi^{(4)}_{ij}(q_1, q_2) \Delta_{ij}(q_3) \Delta_{ij}(q_4) \delta_{q_1+q_2=q_3+q_4},
\]

where

\[
\chi^{(4)}_{ij}(q_1, q_2) = \frac{1}{\beta} \sum_{p, \mu, \nu} \text{Tr} \left( \gamma_{ij}(q_1) G_{v}(p, i\omega_n) G_{v}(p, i\omega_n) \gamma_{ij}^v(q_1) G_{v}(p, i\omega_n) \right).
\]

Here we have neglected the spatial dependence (no $q$ dependence) in the fourth order superconductivity susceptibility $\chi^{(4)}$ and only focus on the uniform case for this term.

The superconducting order parameter is determined by minimizing Ginzburg-Landau free energy, which can be determined by considering $\frac{d^2}{d\Delta^2} = 0$. Near the critical temperature, the order parameter can be regarded a perturbation and in this case, we only need to consider the second order term and obtain the linearized gap equation

\[
\Delta_{ij}(q) = V_i \sum_{j'} \chi_{ij}(q) \Delta_{ij}(q).
\]

for the pairing in the representation $i$. The linearized gap equation will be used to study the critical temperature of our system.
Appendix B: Green function and superconductivity susceptibility

To solve the linearized gap equation, it is essential to calculate Matsubara Green functions and superconductivity susceptibility. In this part, we consider the Hamiltonian of bilayer TMD (Eq. (1) in the main text) with Zeeman coupling, given by

\[ H = \xi_k + \epsilon \beta s \cdot \tau z + \tau z + gB \cdot s. \]  

where \( \xi_k \) is the kinetic energy, \( \beta \) term describes Ising spin-orbit coupling (SOC), \( \tau \) term is for the hybridization between two layers and the last term is for Zeeman coupling with the magnetic field \( B \). The eigen-energy of the above Hamiltonian is given by

\[ \epsilon_{ext} = \xi_k + \lambda \sqrt{D_0^2 + g^2B^2 + 2sD_1} \]  

where \( D_0 = \sqrt{\beta^2 + \tau^2} \) and \( D_1 = \sqrt{\beta^2B^2 + \tau^2g^2B^2} \). The corresponding Matsubara Green function for electrons can be written in a compact form as

\[ G_e(k, i\omega_n) = \sum_{\Delta s} \frac{p_{e,ext}^{\Delta s}}{i\omega_n - \xi_k - \lambda F_s} \]  

where \( F_s = \sqrt{D_0^2 + g^2B^2 + 2sD_1} \) and

\[ p_{e,ext}^{\Delta s} = \frac{1}{4} \left( 1 + \frac{\lambda}{F_s}(\epsilon \beta s \cdot \tau z + \tau z + gB \cdot s) \right) \left( 1 + \frac{s}{D_1}(\epsilon \beta s \cdot \tau z + g(B \cdot s)\tau z) \right). \]  

The hole Green function is

\[ G_h(k, i\omega_n) = \sum_{\Delta s} \frac{p_{h,ext}^{\Delta s}}{i\omega_n + \xi_k - \lambda F_s} \]  

where

\[ p_{h,ext}^{\Delta s} = \frac{1}{4} \left( 1 + \frac{\lambda}{F_s}(\epsilon \beta s \cdot \tau z - \tau z - gB \cdot s) \right) \left( 1 + \frac{s}{D_1}(\epsilon \beta s \cdot \tau z + g(B \cdot s)\tau z) \right). \]  

Now we only focus on the uniform case (\( q = 0 \)) and may substitute the form of electron and hole Green functions into superconductivity susceptibility to obtain

\[ \chi^{(2)}_{\nu \nu,00}(0) = -\frac{1}{\beta} \sum_{\Delta s, e, \nu, \nu', h, e, \nu} \sum_{o_n} \frac{1}{i\omega_n - \xi_k - \lambda F_s} \left( \frac{1}{i\omega_n + \xi_k - \lambda F_s} \right) \text{Tr} \left( \gamma_\nu \gamma_\nu' p_{\nu,ext}^{e,\Delta s} p_{\nu',ext}^{h,\Delta s} \right). \]  

Since \( \text{Tr} \left( \gamma_\nu \gamma_\nu' p_{\nu,ext}^{e,\Delta s} p_{\nu',ext}^{h,\Delta s} \right) \) does not depend on \( i\omega_n \) and \( k \), we can first integrate out \( \sum_{o_n} \frac{1}{i\omega_n - \xi_k - \lambda F_s} \frac{1}{i\omega_n + \xi_k - \lambda F_s} \). This integral can be further simplified. One can show that the singular behavior of the above expression comes from the term \( \lambda = -\lambda' \) when \( D_0 \) is large. Therefore, we consider the case with \( \lambda = -\lambda' = - \) (the case with \( \lambda = -\lambda' = + \) is similar). In this case, we define

\[ \chi_{0,0} = -\frac{1}{\beta} \sum_{\Delta s, o_n, k} \frac{1}{i\omega_n - \xi_k + F_s} \frac{1}{i\omega_n + \xi_k - F_s} \]  

and direct calculation shows that

\[ \chi_{0,0} = N_0 \ln \left( \frac{2\gamma D_0}{\pi k T} \right) + N_0 \psi \left( \frac{1}{2} \right) - \frac{N_0}{2} \left( \psi \left( \frac{1}{2} - \frac{i}{2\pi k T} (F_s - F_s') \right) + \psi \left( \frac{1}{2} + \frac{i}{2\pi k T} (F_s - F_s') \right) \right) \]  

where \( N_0 \) is density of states at the Fermi energy, \( \gamma = 1.57 \), \( \omega_D \) is Debye frequency, which is chosen to be 10meV in our calculation, and \( \psi \) is the di-gamma function. Therefore, we have \( \chi_{0,0'} = \chi_{0,-} = N_0 \ln \left( \frac{2\gamma D_0}{\pi k T} \right) = \chi_0 \) and \( \chi_{0,-} = \chi_{0,-} = \chi_0 - \delta \chi \) with

\[ \delta \chi = \frac{N_0}{2} \left[ \psi \left( \frac{1}{2} - \frac{i}{2\pi k T} (F_s - F_s') \right) + \psi \left( \frac{1}{2} + \frac{i}{2\pi k T} (F_s - F_s') \right) - 2\psi \left( \frac{1}{2} \right) \right] \approx C \left( \frac{1}{2\pi k T} \right)^2 (F_s - F_s')^2 \]  

where \( C \) is a constant and needs to be determined from the experiment. The critical temperature is then obtained as

\[ T_c \approx \frac{\beta C}{\pi k T} \left( \frac{D_0^2}{D_0' \omega_D} \right)^{1/2} \]  

which can be compared with the experimental results.
where $C = -\psi^{(2)}(\frac{3}{2}) \approx 16.83$. With the above expression, the superconductivity susceptibility is simplified as

$$\chi^{(2)}_{ij\mu\nu} = \sum_{\xi,\xi'} \chi_{0,\xi\xi'} Tr \left( \gamma^\xi_{ij} \left\{ \rho_{\mu}^e \gamma_{\xi \xi'} \rho_{\nu}^e \right\}_{\xi \xi'} \right)$$  \hspace{1cm} (B11)

for $\lambda = -\lambda' = -\lambda$. To obtain the gap equation, we need further to decompose the pairing interaction into different representations. The pairing interaction is introduced as $\tilde{H}_V = \frac{1}{2} \sum_{\mu'} \rho^e_{\mu} \sum_{\xi,\xi'} V^{\xi\xi'}_{\eta\eta'} \gamma^\xi_{\eta\eta'} \rho^e_{\mu'}$. For simplicity, we assume the interaction is momentum independent (on-site interaction) and only consider the following non-zero $V^{\eta\eta'}_{\xi\gamma\gamma'}$: (1) intra-layer interaction

$$V^{\gamma\gamma'}_{\xi\xi} = -V^{\gamma\gamma'}_{\xi\xi} = -U_0$$  \hspace{1cm} (B12)

and (2) inter-layer interaction

$$V^{\gamma\gamma'}_{\xi\xi} = -V^{\gamma\gamma'}_{\xi\xi} = -V_{\xi\xi} = -V_0,$$  \hspace{1cm} (B13)

where $\tilde{\sigma}$ and $\tilde{\eta}$ reverse the value of $\sigma$ and $\eta$. Here we introduce additional minus sign before $U_0$ and $V_0$ so that the attractive interaction is defined for $U_0 > 0$ and $V_0 > 0$. Next we decompose the interaction into different channels with the form

$$V^{\xi\xi'}_{\eta\eta'} = -\sum_{\mu,\mu'} V_{\mu\mu'} \left(\gamma^\xi_{\eta\eta'} \gamma^\mu_{\xi\xi'}\right).$$  \hspace{1cm} (B14)

where $\gamma$ matrices label different representation matrices. For the $D_{3d}$ group, $s_\gamma$ and $s_\tau_\gamma$ (labeled as $\gamma_{A_{1g},1}$ and $\gamma_{A_{1g},2}$) belong to the $A_{1g}$ representation, $s_\tau_\gamma$ (labeled as $\gamma_{A_{1u},1}$) belongs to $A_{1u}$, $s_\tau_{\gamma'}$ (labeled as $\gamma_{A_{2u}}$) belongs to $A_{2u}$, and $s_{\tilde{\tau}_\gamma}$ (labeled as $(\gamma_{E_u},1, \gamma_{E_u},2)$) to $E_u$. More explicitly, $\gamma_{A_{1g},1}$ and $\gamma_{A_{1g},2}$ describe intra-layer and inter-layer singlet pairings ($c_{\uparrow}c_{\downarrow} + c_{\downarrow}c_{\uparrow}$ and $c_{\uparrow}c_{\downarrow} - c_{\downarrow}c_{\uparrow}$), $\gamma_{A_{1u}}$ corresponds to intra-layer singlet pairings with opposite phases between two layers ($c_{\uparrow}c_{\downarrow} - c_{\downarrow}c_{\uparrow}$), $\gamma_{A_{2u}}$ is for inter-layer singlet pairing ($c_{\uparrow}c_{\downarrow} - c_{\downarrow}c_{\uparrow}$) and $\gamma_{E_u}$ gives inter-layer triplet pairing ($c_{\uparrow}c_{\downarrow} - c_{\downarrow}c_{\uparrow}$). With these matrices, we can compare matrix elements of interactions in Eq. (B14) with those in Eq. (B12) and Eq. (B13) and obtain $V_{A_{1g},1} = V_{A_{1u},1} = \frac{U_0}{2}$ and $V_{A_{1g},2} = V_{A_{2u},1} = V_{E_u,1} = V_{E_u,2} = \frac{V_0}{2}$.

Next we need to evaluate the element $Tr \left( \gamma^\xi_{ij} \left\{ \rho_{\mu}^e \gamma_{\eta\eta'} \rho_{\nu}^e \right\}_{\eta\eta'} \right)$ and discuss the gap equation in different representations separately.

(1) $A_{1g}$ representation ($s_\gamma, s_\tau_\gamma$)

Since we have two possible representation matrices for the $A_{1g}$ representation, the gap function can be expanded as $\Delta_{A_{1g}} = \Delta_{A_{1g},1}s_\gamma + \Delta_{A_{1g},2}s_\tau_\gamma$. We can compute $\chi^{(2)}_{A_{1g},\mu\nu}$ directly and only keep terms up to the second order in $B$. The superconductivity susceptibility is given by

$$\chi_{A_{1g},11} = 4(\chi_0 - \delta \chi)$$  \hspace{1cm} (B15)

$$\chi_{A_{1g},12} = \chi_{A_{1g},21} = 2(\chi_0 - \delta \chi) \left( - \frac{t}{D_0} - \frac{t^2gB^2}{D_0D_1^2} \right) + 2\chi_0 \left( - \frac{t}{D_0} + \frac{t^2gB^2}{D_0D_1^2} \right)$$  \hspace{1cm} (B16)

$$\chi_{A_{1g},22} = 2\chi_0 \left( \frac{t^2}{D_0} - \frac{t^2gB^2}{D_1} \right) \left( \frac{B_0^2 + B_1^2}{D_0} \right) + 2(\chi_0 - \delta \chi) \left( \frac{t^2}{D_0} + \frac{t^2gB^2}{D_1} \right) \left( \frac{B_0^2 + B_1^2}{D_0} \right),$$  \hspace{1cm} (B17)

where $\chi_{A_{1g},11}$ is for $s_\gamma$, $\chi_{A_{1g},22}$ is for $s_\tau_\gamma$ and $\chi_{A_{1g},12}$ and $\chi_{A_{1g},21}$ describe the coupling between $s_\gamma$ and $s_\tau_\gamma$. Since $V_{A_{1g},1} = \frac{U_0}{2}$ and $V_{A_{1g},2} = \frac{V_0}{2}$, we obtain a set of linear equations

$$\Delta_{A_{1g},1} = \frac{U_0}{2} \chi_{A_{1g},11} \Delta_{A_{1g},1} + \chi_{A_{1g},12} \Delta_{A_{1g},2},$$  \hspace{1cm} (B18)

$$\Delta_{A_{1g},2} = \frac{V_0}{2} \chi_{A_{1g},21} \Delta_{A_{1g},1} + \chi_{A_{1g},22} \Delta_{A_{1g},2}.$$  \hspace{1cm} (B19)
The corresponding eigen-state of $A_{1g}$ pairing satisfies

$$\Delta_{A_{1g}}^{(2)} = -\frac{V_0 t}{U_0 D_0}\Delta_{A_{1g},1}. \quad (B20)$$

Therefore in case $t \ll D_0$, the $\Delta_{A_{1g},1}$ part dominates.

For a non-zero but weak $B$, we have $\delta \chi \ll \chi_0$ and the critical temperature is close to $T_{c,0,A_{1g}}$. The correction to the critical temperature can be obtained by substituting (B20) into (B18) and is determined by

$$N_0 \ln \left( \frac{T_{c,A_{1g}}}{T_{c,0,A_{1g}}} \right) = \frac{2 U_0 + V_0 \frac{t^2}{D_0^2} + \frac{t^4 B^2}{D_1^2 D_0^4}}{2 U_0 + 2 V_0 \frac{t^2}{D_0^2}} \delta \chi. \quad (B21)$$

For an in-plane magnetic field ($D_1 = t B_0$), we have

$$\ln \left( \frac{T_{c,A_{1g}}}{T_{c,0,A_{1g}}} \right) \approx -2 C \left( \frac{1}{2 \pi k T_{c,0,A_{1g}}} \right)^2 \frac{t^2 \gamma^2 B_0^2}{D_0^2}, \quad (B22)$$

while for an out-of-plane magnetic field ($D_1 = \beta \parallel B_z$), we have

$$\ln \left( \frac{T_{c,A_{1g}}}{T_{c,0,A_{1g}}} \right) \approx -2 C \left( \frac{1}{2 \pi k T_{c,0,A_{1g}}} \right)^2 \frac{\gamma^2 B_z^2}{D_0^2}. \quad (B23)$$

Comparing the critical temperatures for in-plane magnetic fields and out-of-plane magnetic fields, we find that the paramagnetic effect for in-plane magnetic fields is much weaker than that for out-of-plane magnetic fields due to the factor $\frac{1}{D_0^2}$, which will lead to the high in-plane critical magnetic field and is consistent with the experimental observations in bilayer TMD materials.

(2) $A_{1u}$ pairing $(s, \tau_s)$

The superconductivity susceptibility is given by

$$\chi^{(2)}_{A_{1u}} = 4 \chi_0 \frac{\beta_{4s}^2}{D_0^2} \delta \chi \left( \frac{2 \beta_{4s}^2}{D_0^2} + \frac{\beta_{3s}^2 \gamma^2 B_z^2 - \gamma^2 B_z^2}{D_1^2} + \frac{D_0^2 \gamma^2 B_z^2 - \beta_{4s}^2 \gamma^2 B_z^2}{D_0^2 D_1^2} + \frac{t^4 \gamma^2 B_z^2}{D_0^2 D_1^2} \right) \quad (B24)$$

and the corresponding gap equation is $\Delta_{A_{1u}} = \frac{U_0}{\chi^{(2)}_{A_{1u}}} \Delta_{A_{1u}}$. At zero magnetic field $B = 0$ and $\delta \chi = 0$, we find

$$k T_{c,0,A_{1u}} = \frac{2 \gamma \omega_D}{\pi} \exp \left( - \frac{D_0^2}{2 U_0 \beta_{4s}^2} \right). \quad (B25)$$

In a weak magnetic field and in the limit $t \ll \beta_{4s}$,

$$\ln \left( \frac{T_{c,A_{1u}}}{T_{c,0,A_{1u}}} \right) = - \left( 2 + \frac{1}{D_1} \left( \frac{2 \beta_{4s}^2 \gamma^2 B_z^2 - \gamma^2 B_z^2}{D_1^2} - \frac{D_0^2 \gamma^2 B_z^2 - \beta_{4s}^2 \gamma^2 B_z^2}{D_0^2 D_1^2} + \frac{t^4 \gamma^2 B_z^2}{D_0^2 D_1^2} \right) \right) \frac{\delta \chi}{4}. \quad (B26)$$

Thus, for out-of-plane magnetic fields,

$$\ln \left( \frac{T_{c,A_{1u}}}{T_{c,0,A_{1u}}} \right) = -2C \left( \frac{1}{2 \pi k T_{c,0,A_{1u}}} \right)^2 \frac{\gamma^2 B_z^2}{D_0^2}, \quad (B27)$$

and for in-plane magnetic fields,

$$N_0 \ln \left( \frac{T_{c,A_{1u}}}{T_{c,0,A_{1u}}} \right) = -C \left( \frac{1}{2 \pi k T_{c,0,A_{1u}}} \right)^2 \frac{t^4 \gamma^2 B_z^2}{\beta_{4s}^2 D_0^2}. \quad (B28)$$

We find a factor $\frac{t^4}{\beta_{4s}^2}$ for in-plane magnetic fields but not for out-of-plane magnetic fields.

(3) $A_{2u}$ pairing $(s, \tau_s)$

In this case, we find that $\chi^{(2)}_{A_{2u}} = 0$ and thus no superconducting phase is possible for this representation.

(4) $E_u$ pairing $(\{\tau_y, s, \tau_s\})$
Since the $E_u$ pairing are two dimensional, we can write down a gap equation for each component. However, since they are related to each other by symmetry, we expect two components share the same $T_c$. Therefore, we only consider $\tau_y$ part here. The superconductivity susceptibility is

$$
\chi_{E_u}^{(2)} = 4\chi_0 \beta_{so}^2 D_0 - \left(2 \beta_{so}^2 D_0 + \frac{1}{D_1^2} (\beta_{so}^2 + D_0^2) g^2 B_z^2 - 2 r^2 g^2 B_z^2 \right) \delta \chi.
$$

(B29)

At zero magnetic field, we have

$$
kT_{c0,E_u} = \frac{2\gamma_D}{\pi} \exp \left(-\frac{D_0^2}{2N_0V_0\beta_{so}^2} \right).
$$

(B30)

For a weak magnetic field and $t \ll \beta_{so}$, we have

$$
N_0 \ln \left( \frac{T_{c,E_u}}{T_{c0,E_u}} \right) = -\left(2 - \frac{D_0^2 g^2}{D_1^2} \right) \left(1 + \frac{\beta_{so}^2}{D_0^2} \right) \frac{r^2 g^2 B_z^2}{4} \delta \chi.
$$

(B31)

For out-of-plane magnetic fields,

$$
\ln \left( \frac{T_{c,E_u}}{T_{c0,E_u}} \right) = 0,
$$

(B32)

and for in-plane magnetic fields,

$$
\ln \left( \frac{T_{c,E_u}}{T_{c0,E_u}} \right) = -C \left(\frac{1}{2\pi kT_{c0,E_u}}\right)^2 \left(1 + \frac{\beta_{so}^2}{D_0^2} \right) \frac{r^2 g^2 B_{||}^2}{4} \delta \chi.
$$

(B33)

Therefore, we find Zeeman coupling will not reduce the $T_c$ for out-of-plane magnetic fields and the contribution of in-plane magnetic fields has a factor of $\left(1 + \beta_{so}^2/D_0^2\right) \beta_{so}$. This is because the $E_u$ pairing corresponds to the interlayer equal spin pairing.

Appendix C: Ginzburg-Landau free energy

In the above, we have presented our derivations and results of the linearized gap equation for bilayer TMD materials. In this section, we will construct the Ginzburg-Landau free energy for our system. In particular, this approach will allow us to study inhomogeneous superconductivity when we consider the orbital effect of magnetic fields.

The Ginzburg-Landau free energy is given by Eqs. (A9)-(A13), in which one needs to evaluate superconductivity susceptibility $\chi^{(2)}$ and $\chi^{(4)}$. We have computed $\chi^{(2)}$ for $q = 0$, which can be directly applied to Landau free energy, in the last section for the linearized gap equation. In this part, we need to further include the $q$ dependence in order to discuss the gradient term in the Landau free energy.

With a finite $q$, the superconductivity susceptibility $\chi^{(2)}$ is changed to

$$
\chi_{ij,\mu\nu}^{(2)}(q) = -\frac{1}{\beta} \sum_{s,s',\lambda',\lambda} \sum_{\omega_{m},\epsilon_{k}} \frac{1}{i\omega_{\lambda} - \xi_{k+q}^{\lambda'}} - D_{\lambda} \lambda_{\lambda'} \lambda_{\lambda'} - \xi_{k-\lambda}^{\lambda'} + \lambda' D_{\lambda}
$$

\( \text{Tr} \left( \gamma^{s}_{\mu} \gamma^{s'}_{\nu} B^{p}_{\lambda} \right) \).

(C1)

There is no momentum dependence in $\text{Tr} \left( \gamma^{s}_{\mu} \gamma^{s'}_{\nu} B^{p}_{\lambda} \right)$ and thus, we only need to consider $\xi_{k+q}^{\lambda'}$. We may treat $q$ as a small number and expand it as $\xi_{k+q}^{\lambda'} = \xi_{k}^{\lambda'} \pm \frac{1}{2} \epsilon_{k} \cdot q$ up to the linear term in $q$. Since we have already discussed the Zeeman coupling, we will neglect this term in the discussion below for simplicity ($F_s = D_0$). In this case, we define

$$
\chi_{0,\lambda'\lambda}(q) = -\frac{1}{\beta} \sum_{\omega_{m},\epsilon_{k}} \frac{1}{i\omega_{\lambda} - \xi_{k+q}^{\lambda'}} - D_{\lambda} \lambda_{\lambda'} - \xi_{k-\lambda}^{\lambda'} + \lambda' D_{\lambda},
$$

(C2)

which is independent of $s$ and $s'$. $\chi_{ij,\mu\nu}^{(2)}(q) = \sum_{s,s',\lambda',\lambda} \chi_{0,\lambda'\lambda}(q) \text{Tr} \left( \gamma^{s}_{\mu} \gamma^{s'}_{\nu} B^{p}_{\lambda} \right)$. Furthermore, we only focus on $\lambda = -\lambda'$ and in this case,

$$
\chi(q) = \delta(q) = N_0 \ln \left( \frac{2\gamma_D}{\pi kT} \right) + N_0\psi\left(\frac{1}{2}\right) - \frac{N_0}{8\pi} \int d\Omega_{k} \left( \psi \left( \frac{1}{2} - \frac{i}{2\pi kT} \hbar v_{f} \epsilon_{k} \cdot q \right) + \psi \left( \frac{1}{2} + \frac{i}{2\pi kT} \hbar v_{f} \epsilon_{k} \cdot q \right) \right).
$$

(C3)
where $\nu_f$ is the Fermi velocity, $e_k$ is the unit vector of the momentum $k$ and $\Omega_k$ is the solid angle for the $k$ integral. By expand the digamma function and perform the $\Omega_k$ integral, we obtain

$$\chi_0(q) = N_0 \ln \left( \frac{2\nu \omega K}{\pi kT} \right) - \frac{N_0 C}{6} \left( \frac{\hbar}{2 m kT} \right)^2 q^2. \quad (C4)$$

From the above equation, we can construct the second order term $L_2$ in the Landau free energy. We will next discuss the pairing in each representation, separately. The $T$ in the above expression should be replaced by the $T_{c0}$ for the corresponding pairing.

For the fourth order term, we need to evaluate $\chi^{(4)}$, which is written as

$$\chi^{(4)}_{\hat{p}, \hat{p}, \hat{p}^*, \hat{s}, \hat{s}^*} = \frac{1}{\beta} \sum \sum \sum \frac{i \omega_n - \xi_k - \Lambda_1 D_0 i \omega_n + \xi_k - \Lambda_2 D_0 i \omega_n - \xi_k - \Lambda_3 D_0 i \omega_n + \xi_k - \Lambda_4 D_0}{1} \text{Tr} \left( \gamma_{\hat{p}, \hat{p}} \gamma_{\hat{s}, \hat{s}} \gamma_{\hat{p}^*, \hat{p}^*} \gamma_{\hat{s}^*, \hat{s}^*} \right). \quad (C5)$$

We again consider the most singular part of the momentum-frequency space integral, which is contributed from $\Lambda_1 = \Lambda_3 = -$ and $\Lambda_2 = \Lambda_4 = +$. Therefore, we have

$$\chi^{(4)}_{\hat{p}, \hat{p}, \hat{p}, \hat{s}, \hat{s}} = \frac{7N_0 \zeta(3)}{8(\pi kT)^2} \sum \text{Tr} \left( \gamma_{\hat{p}, \hat{p}} \gamma_{\hat{s}, \hat{s}} \gamma_{\hat{p}^*, \hat{p}^*} \gamma_{\hat{s}^*, \hat{s}^*} \right), \quad (C6)$$

where $\zeta(x)$ is the zeta-function.

(1) $A_{1g}$ pairing

Direct calculation of superconductivity susceptibility almost recovers our previous results with the replacement of $\chi_0$ by $\chi_0(q)$. Therefore, we have $\chi_{A_{1g},11} = 4\chi_0(q)$, $\chi_{A_{1g},12} = \chi_{A_{1g},21} = -\frac{2}{T_{c0}}\chi_0(q)$ and $\chi_{A_{1g},22} = \frac{4}{T_{c0}}\chi_0(q)$. Therefore, the second order term of Landau free energy is written as

$$L_{2,A_{1g}} = \frac{1}{2} \sum_q \left( \Delta_{A_{1g}}^+ \Delta_{A_{1g}}^- \right) \text{Tr} \left( \frac{1}{\Lambda_{A_{1g},11} - \Lambda_{A_{1g},12}} - \frac{1}{\Lambda_{A_{1g},21} - \Lambda_{A_{1g},22}} \right) \left( \Lambda_{A_{1g},11} \Lambda_{A_{1g},22} \right)$$

$$= \sum_q \left( \Delta_{A_{1g},11} \Delta_{A_{1g},22} \right) \left( \frac{1}{\Lambda_{A_{1g},11} - \Lambda_{A_{1g},12}} - \frac{1}{\Lambda_{A_{1g},21} - \Lambda_{A_{1g},22}} \right) \left( \Lambda_{A_{1g},11} \Lambda_{A_{1g},22} \right) \quad (C7)$$

One can take the derivative of $L_{2,A_{1g}}$ with respect to $\Delta_{A_{1g},1}$ and recover the linearized gap equation. Since the $\Delta_{A_{1g},1}$ pairing dominates when $r \ll B_{st}$, we may substitute $\Delta_{A_{1g},2}$ with $\Delta_{A_{1g},1}$ and obtain

$$L_{2,A_{1g}} = \int d\mathbf{r} \left( \mathcal{K}_{A_{1g}} \left| \nabla \Delta_{A_{1g},1}(\mathbf{r}) \right|^2 + \mathcal{A}_{A_{1g}} \left| \Delta_{A_{1g},1}(\mathbf{r}) \right|^2 \right) \quad (C8)$$

with $\mathcal{K}_{A_{1g}} = \frac{N_0 C}{3} \left( \frac{\hbar}{2 e k T} \right)^2 \left( 1 + \frac{V_{D_s}^2}{D_0^2} \right)^2$ and $\mathcal{A}_{A_{1g}} = 2N_0 \left( 1 + \frac{V_{D_s}^2}{D_0^2} \right)^2 \ln \left( \frac{T_{c0,A_{1g}}}{T_{c0,A_{1g}}} \right) \approx 2N_0 \left( 1 + \frac{V_{D_s}^2}{D_0^2} \right)^2 \left( T_{c0,A_{1g}} - T_{c0,A_{1g}} \right)$ when the temperature is close to $T_{c0,A_{1g}}$. We further label $C_{A_{1g}} = 2N_0 \left( 1 + \frac{V_{D_s}^2}{D_0^2} \right)^2$, and $\mathcal{A}_{A_{1g}} = C_{A_{1g}} \ln \left( \frac{T_{c0,A_{1g}}}{T_{c0,A_{1g}}} \right)$. The fourth order term can also be computed directly with the electron and hole Green functions, and we obtain

$$L_{4,A_{1g}} = \int d\mathbf{r} \left( B_{A_{1g}} \left| \Delta_{A_{1g},1}(\mathbf{r}) \right|^2 \right), \quad (C9)$$

where $B_{A_{1g}} = \frac{7N_0 \zeta(3)}{8(\pi kT)^2}$. Since we only concerns the temperature close to the critical temperature, $T$ in $\mathcal{K}$ and $B$ can be replaced by $T_{c0,A_{1g}}$. Eqs. (C8) and (C9) together form the Landau free energy for the $A_{1g}$ pairing.

For the out-of-plane magnetic field, the orbital effect can be taken into account by replacing $-i \nabla \to -i \nabla + \frac{2\pi^2}{e} \mathbf{A}$, where $\mathbf{A}$ is the gauge potential. The orbital effect of in-plane magnetic fields will be discussed in details later.

(2) $A_{1u}$ pairing

The second order term in the Landau free energy for the $A_{1u}$ pairing is given by

$$L_{2,A_{1u}} = \int d\mathbf{r} \left( \mathcal{K}_{A_{1u}} \left| \nabla \Delta_{A_{1u},1}(\mathbf{r}) \right|^2 + \mathcal{A}_{A_{1u}} \left| \Delta_{A_{1u},1}(\mathbf{r}) \right|^2 \right), \quad (C10)$$

where $\mathcal{K}_{A_{1u}} = \frac{N_0 C}{3} \left( \frac{\hbar}{2 e k T} \right)^2 \frac{D_s^2}{D_0^2} \mathcal{A}_{A_{1u}} = \frac{2N_0 \nu D_s^2}{D_0^2} \ln \left( \frac{T_{c0,A_{1u}}}{T_{c0,A_{1u}}} \right) \approx \frac{2N_0 \nu D_s^2}{D_0^2} \left( T_{c0,A_{1u}} - T_{c0,A_{1u}} \right)$ and $C = \frac{2N_0 \nu D_s^2}{D_0^2}$.
The fourth order term is given by

\[ L_{4,A_{1u}} = \int d\mathbf{r} \left( B_{A_{1u}} |A_{4u}(\mathbf{r})|^4 \right), \]  

(\text{C11})

where \( B_{A_{1u}} \equiv \frac{7N_a(0)\mu^4}{8n_F^2D_f^2} \).

The second order term and the fourth order term are given by

\[ L_{2,E_u} = \int d\mathbf{r} \sum_{\mu} \left( K_{E_u} \| \Delta_{E_u}(\mathbf{r}) \|^2 + A_{E_u} \| \Delta_{E_u}(\mathbf{r}) \|^2 \right), \]  

(\text{C12})

and

\[ L_{4,E_u} = \int d\mathbf{r} \left( B_{E_u,1} |\Delta_{E_u,1}(\mathbf{r})|^2 + |\Delta_{E_u,2}(\mathbf{r})|^2 \right) + B_{E_u,2}(\Delta_{E_u,1}^* \Delta_{E_u,2} - \Delta_{E_u,1} \Delta_{E_u,2}^*), \]  

(\text{C13})

where \( K_{E_u} \equiv \frac{N_a}{3} \left( \frac{\hbar v}{2m_F} \right)^2 \), \( A_{E_u} \equiv \frac{2N_a\mu^2}{D_f} n \left( \frac{T}{T_{\mu\mu}} \right) \approx \frac{2N_a\mu^2}{D_f} T - T_{\mu\mu}, C_{E_u} \equiv \frac{2N_a\mu^2}{D_f} B_{E_u,1} = \frac{7N_a(0)\mu^4}{8n_F^2D_f^2} B_{E_u,2} = -\frac{7N_a(0)\mu^4}{8n_F^2D_f^2} \) in the weak coupling limit. It is known that when \( B_{E_u,2} < 0 \), the nematic superconductivity with pairing function \( \Delta_{E_u,1}, \Delta_{E_u,2} = \Delta_{E_u}(\cos \theta, \sin \theta) \) will become stable. On the other hand, if \( B_{E_u,2} > 0 \), chiral superconductivity \( \Delta_{E_u,1}, \Delta_{E_u,2} = \Delta_{E_u}(1, i) \) will be realized.

Appendix D: The orbital effect of in-plane magnetic fields

In this part, we will discuss the orbital effect of in-plane magnetic fields, which turns out to be important for inducing inhomogeneous superconducting pairing. Normally, the orbital effect of in-plane magnetic fields is neglected in 2D systems because of quantum confinement along the out-of-plane direction. However, as we will show below, it will have an interesting consequence in 2D TMD materials due to the unique band structures.

Let us assume the magnetic field is along the \( x \) direction and the corresponding gauge potential is chosen as \( A = (0, -B_z, 0) \). The distance between two layers of TMD materials is taken as \( z_0 \) and the origin point is chosen at the center between two layers. Thus, in our Hamiltonian, we need to replace \( \xi_k \) by \( \xi_k = \frac{\hbar}{2m}(k_x^2 + (k_y - Q\tau_z)^2) - \mu \), where \( Q = \frac{eB_z}{2\hbar} \). We may expand \( \xi_{\pi} \) and keep only the first order term in \( B_z \). The resulting Hamiltonian is

\[ H = \xi_k - \hbar v_Q k_x \tau_z + \epsilon \beta_{so} s_z \tau_z + tr_s, \]  

(\text{D1})

with \( v_Q = \frac{eB_z}{2m} \) and the corresponding energy dispersion is given by

\[ \epsilon_{\pi}(\epsilon, k) = \xi_k + \lambda F_{\pi}(k_x), \]  

(\text{D2})

where \( F_{\pi} = \sqrt{(B_{so} - \epsilon \hbar v_Q k_x)^2 + t^2} \). The energy dispersion is shown in Fig. 1d in the main text for the limit \( t = 0 \).

The electron and hole Green functions are given by

\[ \mathcal{G}_e(\epsilon, k, i\omega_n) = \sum_{\Delta s} \frac{P^e_{\epsilon,s}(k_x)}{i\omega_n - \xi_k - \lambda F_{\pi}(k_x)}, \]  

(\text{D3})

\[ \mathcal{G}_h(\epsilon, k, i\omega_n) = \sum_{\Delta s} \frac{P^h_{\epsilon,s}(k_x)}{i\omega_n + \xi_k - \lambda F_{\pi}(k_x)}, \]  

(\text{D4})

where we have used \( \xi_{-k} = \xi_k \) and

\[ P^e_{\epsilon,s} = \frac{1}{4} \left( 1 + \frac{\lambda}{F_{\pi}(k_x)}(-\hbar v_Q k_x \tau_z + \epsilon \beta_{so} s_z \tau_z + tr_s) \right)(1 + ss_z), \]  

(\text{D5})

\[ P^h_{\epsilon,s} = \frac{1}{4} \left( 1 + \frac{\lambda}{F_{\pi}(k_x)}(-\hbar v_Q k_x \tau_z + \epsilon \beta_{so} s_z \tau_z - tr_s) \right)(1 + ss_z). \]  

(\text{D6})

Next we need to use the Green function and eigen-energy of the Hamiltonian to evaluate \( \chi^{(2)}(\mathbf{q}) \). We treat both \( \mathbf{q} \) and the magnetic field \( B_z \) (the corresponding \( Q \) and \( v_Q \)) as perturbations and expand superconductivity susceptibility up to the second order in \( \mathbf{q} \) and \( B_z \). In particular, in-plane magnetic fields break the \( D_{3d} \) symmetry, and thus can couple the pairings in different representations. Direct calculations show that the \( A_{1g} \) pairing is coupled to the \( A_{1u} \) pairing. Therefore, we first discuss the coupled \( A_{1g} - A_{1u} \) pairing and then consider \( E_u \) pairing.
\[ L = \frac{1}{2} \sum_q \left( \begin{array}{ccc}
\Delta_1^* & \Delta_2^* & \Delta_3^* \\
1 & -\chi_{11} & -\chi_{12} & -\chi_{13} \\
\chi_{11} & 1 & -\chi_{21} & -\chi_{22} \\
\chi_{12} & \chi_{21} & 1 & -\chi_{31} \\
\chi_{13} & \chi_{22} & \chi_{31} & 1 \\
\end{array} \right) \left( \begin{array}{c}
\Delta_1 \\
\Delta_2 \\
\Delta_3 \\
\end{array} \right) . \] (D7)

where \( V_{A_{1g}} = \frac{\hbar \omega}{2} \), \( A_{1u} = \frac{\hbar \omega}{2} \) and \( V_{A_{1u}} = \frac{\hbar \omega}{2} \). The superconductivity susceptibility in the above Landau free energy is given by

\[ \chi_{11} = 4N_0 \left( \ln \left( \frac{2\gamma_D \pi kT}{\hbar \omega} \right) \left( 1 - \frac{(\hbar \omega \gamma_f)^2 \pi^2}{4D_0^2 D_0^2} \right) - C \left( \frac{1}{2\pi kT} \right)^2 \left( \frac{(2\beta_{so}(\hbar \omega k_f)^2}{D_0^2 \hbar \omega} + \frac{(\hbar \omega)^2}{2} \right) \right) \] (D8)

\[ \chi_{12} = \chi_{21} = -4N_0 \left( \ln \left( \frac{2\gamma_D \pi kT}{\hbar \omega} \right) \left( 1 + \frac{\beta_{so}(\hbar \omega k_f)^2}{2D_0^4} \right) - C \left( \frac{1}{2\pi kT} \right)^2 \left( \frac{(2\beta_{so}(\hbar \omega k_f)^2}{D_0^2 \hbar \omega} + \frac{(\hbar \omega)^2}{2} \right) \right) \] (D9)

\[ \chi_{13} = \chi_{31} = 2N_0 \hbar \omega \gamma_f \left( \ln \left( \frac{2\gamma_D \pi kT}{\hbar \omega} \right) \left( \frac{(\hbar \omega k_f)^2}{2D_0^2} \right) - \frac{1}{2\pi kT} \right)^2 \left( \frac{(2\beta_{so}(\hbar \omega k_f)^2}{D_0^2 \hbar \omega} + \frac{(\hbar \omega)^2}{2} \right) \] (D10)

\[ \chi_{22} = 4N_0 \left( \ln \left( \frac{2\gamma_D \pi kT}{\hbar \omega} \right) \left( \frac{\beta_{so}^2}{D_0^2} \right) \left( 1 + \frac{1}{2\pi kT} \right)^2 \left( \frac{2\beta_{so}(\hbar \omega k_f)^2}{D_0^2 \hbar \omega} + \frac{(\hbar \omega)^2}{2} \right) \right) \] (D11)

\[ \chi_{23} = \chi_{32} = 2N_0 \hbar \omega \gamma_f \left( \ln \left( \frac{2\gamma_D \pi kT}{\hbar \omega} \right) \left( \frac{(\beta_{so}^2)(\hbar \omega k_f)^2}{2D_0^4} \right) \right) \left( \frac{(\hbar \omega k_f)^2}{2D_0^2} \right) \left( \frac{(2\beta_{so}(\hbar \omega k_f)^2}{D_0^2 \hbar \omega} + \frac{(\hbar \omega)^2}{2} \right) \] (D12)

\[ \chi_{33} = 4N_0 \left( \ln \left( \frac{2\gamma_D \pi kT}{\hbar \omega} \right) \left( \frac{(2\beta_{so}(\hbar \omega k_f)^2}{D_0^2 \hbar \omega} \right) \left( 1 - \frac{(\beta_{so}^2)(\hbar \omega k_f)^2}{D_0^4} \right) \right) - C \left( \frac{1}{2\pi kT} \right)^2 \left( \frac{2\beta_{so}(\hbar \omega k_f)^2}{D_0^2 \hbar \omega} + \frac{(\hbar \omega)^2}{2} \right) \] (D13)

The critical temperature can be obtained by minimizing the above Landau free energy, but this is quite complicated. Therefore, we can consider the following simplifications. We consider the limit \( t \ll \beta_{so} \), in which the \( \Delta_1 \) pairing will dominate over \( \Delta_2 \) for the \( A_{1g} \) pairing. Therefore, we can substitute \( \Delta_2 \) by \( \Delta_2 = -\frac{\gamma_D}{u_0 D_0} \Delta_1 \) and obtain the Landau free energy

\[ L = \frac{1}{2} \sum_q \left( \begin{array}{ccc}
\Delta_1^* & \Delta_2^* & \Delta_3^* \\
\frac{1}{2} & \frac{1}{2} & \frac{1}{2} \\
-\chi_{11} & -\chi_{12} & -\chi_{13} \\
\chi_{11} & 1 & -\chi_{21} \\
\chi_{12} & \chi_{21} & 1 \\
\chi_{13} & \chi_{22} & \chi_{31} \\
\end{array} \right) \left( \begin{array}{c}
\Delta_1 \\
\Delta_2 \\
\Delta_3 \\
\end{array} \right) . \] (D14)

where

\[ \chi_{11} = \chi_{11} + \frac{V_0^2}{U_0 D_0^2} \chi_{22} \frac{V_0}{U_0 D_0} (\chi_{12} + \chi_{21}) = 4N_0 \left( 1 + \frac{V_0^2}{U_0 D_0} \right) \ln \left( \frac{2\gamma_D \pi kT}{\hbar \omega} \right) + \mathcal{P}(v_D, q) \] (D15)

\[ \chi_{13} = \chi_{31} = \chi_{13} - \frac{V_0}{U_0 D_0} \chi_{23} = q(v_D, q) \] (D16)

\[ \chi_{33} = \chi_{33} = 4N_0 \frac{\beta_{so}^2}{U_0 D_0} \ln \left( \frac{2\gamma_D \pi kT}{\hbar \omega} \right) + \mathcal{R}(v_D, q). \] (D17)

Here

\[ \mathcal{P} = 4N_0 \left( \ln \left( \frac{2\gamma_D \pi kT}{\hbar \omega} \right) \frac{(\hbar \omega k_f)^2}{4D_0^2} \left( 1 + \frac{V_0^2}{U_0 D_0} \right) \left( 1 + \frac{(\beta_{so}^2)(\hbar \omega k_f)^2}{D_0^4} \right) \right) + \mathcal{Q}(v_D, q) \] (D18)

\[ \mathcal{Q} = 2N_0 \hbar \omega \gamma_f \left( 1 - \frac{\gamma_D}{u_0 D_0} \right) \frac{\beta_{so}^2}{D_0^2} \left( 1 + \frac{V_0^2}{U_0 D_0} \right) \left( 1 + \frac{(\hbar \omega k_f)^2}{2D_0} \right) + \frac{2\gamma_D \hbar \omega \gamma_f \beta_{so}^2}{D_0^2} \left( 1 + \frac{V_0^2}{U_0 D_0} \right) + \frac{(\hbar \omega k_f)^2}{2D_0} \left( 1 + \frac{V_0^2}{U_0 D_0} \right) \right) \] (D19)

\[ \mathcal{R} = 4N_0 \left( \frac{2\gamma_D \hbar \omega \gamma_f \beta_{so}^2}{D_0^2} \left( 1 - \frac{\gamma_D}{u_0 D_0} \right) \left( 1 + \frac{(\beta_{so}^2)(\hbar \omega k_f)^2}{D_0^4} \right) \right) \left( 1 + \frac{(\hbar \omega k_f)^2}{2D_0} \right) \] (D20)
FIG. 3: The critical temperature as a function of momentum $q_y$ for (a) $B_x/B_N = 0.18$ and (b) $B_x/B_N = 0.37$. $B_N = \frac{2\pi T_c}{\gamma\omega_D}$.

The corresponding linearized gap equation is given by

$$
\begin{pmatrix}
\frac{2}{\nu_0} & 1 + \frac{\nu_0^2}{2\omega_D^2} & -P & -Q & \Delta_1 \\
-\nu_0 & \frac{2}{\nu_0} & -R & \Delta_3
\end{pmatrix}
\begin{pmatrix}
\Delta_1 \\
\Delta_3
\end{pmatrix}
= N_0 \ln \left( \frac{2\gamma \omega_D}{\pi kT} \right)
\begin{pmatrix}
1 + \frac{\nu_0^2}{\omega_D^2} & 0 & 0 \\
0 & \frac{\nu_0^2}{\omega_D^2} & 0
\end{pmatrix}
\begin{pmatrix}
\Delta_1 \\
\Delta_3
\end{pmatrix}
$$

By solving this generalized eigen-equation, one can obtain two solutions for the critical temperature $T_c$, which is a function of magnetic fields $B_x$ and momentum $q_y$. The true $T_c$ is obtained by maximizing the larger solution with respect to $q_y$. Fig. 3 reveal the eigen values of Eq. (D21) as a function of the momentum $q_y$ for different magnetic fields. One can clearly see that for $B_x/B_N = 0.18$, the maximum $T_c$ is located at $q_y = 0$ while for $B_x/B_N = 0.37$, the maximum $T_c$ is shifted to $q_y \approx 0.015$. With this type of calculation for different magnetic fields, one can extract the $T_c$ and $q_c$ as a function of magnetic fields, as shown in Fig. 2b and c in the main text. We find a transition between the conventional BCS state and the FFLO state. Furthermore, we can substitute the maximum $T_c$ and the corresponding eigen vector of $(\Delta_1, \Delta_3)^T$ back to the free energy (D14) and calculate the free energy and the gap function as a function of magnetic fields, as shown in Fig. 4. One can see a rapid change of the slope of free energy at the transition point. At the same time, there is a jump in the gap function. These features indicate a first-order transition in the current case. This transition will be further discussed in details below.
2. The decoupling limit \( t \to 0 \)

In this section, we first consider a simple limit \( t \to 0 \), in which we find \( D_0 = \beta_m \) and \( T_{c,0,Ai} = T_{c,0,Ai} = \frac{2\mu_0}{\pi k} \exp \left( -1/(2U_0 N_0) \right) = T_{c,0} \) and \( \mathcal{P} = \mathcal{R} \). In this case, the Landau free energy takes a simple form

\[
L = \frac{1}{2} \sum_q \left[ \left( \Delta^*_1 \right)^2 + \Delta^*_3 \right] \left( 4N_0 \ln \left( \frac{T_c}{T_{c,0}} \right) - \mathcal{P}(v_0, q) \right) \left( -Q(v_0, q) \right) \left( -Q(v_0, q) \right) \left( 4N_0 \ln \left( \frac{T_c}{T_{c,0}} \right) - \mathcal{P}(v_0, q) \right) \left( \Delta_1 \right),
\]

where

\[
\mathcal{P}(v_0, q) = -N_0 C \left( \frac{1}{2\pi k T} \right)^2 \left( 4(hv_q k_f)^2 + (hv_f q)^2 \right)
\]

\[
Q(v_0, q) = 4N_0 C(hv_f k_f) \left( \frac{1}{2\pi k T} \right)^2 (hv_q q_z)
\]

Since \( v_0 \approx B_z \), we find that \( \Delta_1 \) and \( \Delta_3 \) pairings are coupled to each other by a new term with the form \( \Delta^*_i B_{zi}, q_z \Delta_3 \). We notice a similar term \( \Delta^*_i B_{dzi}, \Delta_3 \) (i, j are two indices for axis) describes the electro-magnetic effect in non-centrosymmetric superconductors. Since \( \Delta_1 \) and \( \Delta_3 \) have opposite parities under inversion, this term does not break inversion symmetry, in sharp contrast to the term \( \Delta^*_i B_{dzi}, \Delta_3 \) in non-centrosymmetric superconductors.

According to the above Landau free energy, the critical temperature can be obtained by maximizing the following function

\[
4N_0 \ln \left( \frac{T_c}{T_{c,0}} \right) = \mathcal{P} + |Q|.
\]

By substituting the form of \( \mathcal{P} \) and \( Q \), we can re-write the above equation as

\[
\ln \left( \frac{T_c}{T_{c,0}} \right) = -C \left( \frac{1}{2\pi k T_{c,0}} \right)^2 \left( (hv_q q_z)^2 + (hv_f q)^2 \right).
\]

It is easy to see that when we choose \( q_z = 0 \) and \( |q| = \frac{2k_\text{vol}}{\nu_f} \), the right hand side of the above expression is maximized. The corresponding \( T_c \) is given by

\[
\ln \left( \frac{T_c}{T_{c,0}} \right) = 0,
\]

from which one can see there is no correction to \( T_c \). In contrast, for \( q = 0 \), we find

\[
\ln \left( \frac{T_c(q = 0)}{T_{c,0}} \right) = -C \left( \frac{\beta_m}{2\pi k T_{c,0}} \right)^2 \left( hv_f k_f \right)^2 /
\]

\[
\frac{\beta_m^2}{\beta_m^2}
\]

which is always smaller than the finite momentum pairing. Therefore, we conclude that under in-plane magnetic fields, the stable superconducting phase occurs for a non-zero \( q_z \), leading to the FFLO state. The stable \( q_z \) is given by \( |q_z| = q_c = \frac{2k_\text{vol}}{\nu_f} = \frac{eB_{zi}}{\mu_0} \). With flux quantum \( \phi_0 = \frac{h}{2e} \), we have \( \lambda_c = \frac{2\pi}{q_c} = \frac{2}\mu_0 \). Thus, the wave length of the FFLO state is determined by the corresponding area for magnetic flux quantum.

We notice that \( q_z = q_c \) and \( q_z = -q_c \) are two degenerate states. To see this, we perform a transformation \( \Delta_+ = \frac{1}{\sqrt{2}} (\Delta_1 + \Delta_3) \) and \( \Delta_0 = \frac{1}{\sqrt{2}} (\Delta_1 \Delta_3) \) and the corresponding Landau free energy is transformed as

\[
L = \frac{1}{2} \sum_q \left( \Delta^*_+ \right) \left( 4N_0 \ln \left( \frac{T_c}{T_{c,0}} \right) - \mathcal{P}(v_0, q) \right) \left( Q(v_0, q) \right) \left( 4N_0 \ln \left( \frac{T_c}{T_{c,0}} \right) - \mathcal{P}(v_0, q) \right) \left( \Delta_+ \right).
\]

Physically, \( \Delta_+ \) describes the pairing in the top layer and \( \Delta_- \) is for the pairing in the bottom layer. Thus, the diagonal form of the above Landau free energy just corresponds to the decoupling between two layers.

Let’s assume \( B_z > 0 \) and if \( q_z > 0, Q > 0 \) and thus \( \Delta_+ \) is favored. For \( q_z < 0, Q < 0 \) and correspondingly, \( \Delta_- \) is favored. Since \( \Delta_0(q_c) \) and \( \Delta_0(-q_c) \) pairings are degenerate, the full real space expression of the FFLO state is given by

\[
\Delta(r) = \Delta_0(q_c)e^{i\mathbf{q}_z} + \Delta_0(-q_c)e^{-i\mathbf{q}_z}.
\]
In the above expression, the first term describes the helical phase in the top layer while the second term is for the helical phase in the bottom layer with opposite momentum.

The relative magnitude of \( \Delta_+(q_c) \) and \( \Delta_-(q_c) \) are determined by the fourth order term in Landau free energy. The general form of the fourth order term is

\[
L_4 = B_t \left( |\Delta_+(q_c)|^2 - |\Delta_-(q_c)|^2 \right)^2 + B_o \left( |\Delta_+(q_c)|^2 + |\Delta_-(q_c)|^2 \right)^2.
\]  

If \( B_o > 0 \), we need \( |\Delta_+| = |\Delta_-| \) to minimize the second term in the above expression. This corresponds to the stripe phase with its pairing amplitude oscillating in the real space. If \( B_t < 0 \), we have either \( |\Delta_+| = 0 \) or \( |\Delta_-| = 0 \), which corresponds to the helical phase, in which the phase oscillates while the amplitude persists.

Microscopically, the fourth order term can be computed from \( \text{(D6)} \). More specifically, the fourth order terms for \( A_{1g} \) and \( A_{1u} \) pairings are given by

\[
L_{4,A_{1g}} = \frac{7N_0\zeta(3)}{16(\pi k T_c)^2} \sum_{|q|} \Delta_1^*(q_1)\Delta_1^*(q_3)\Delta_1(q_2)\Delta_1(q_4)\delta_{q_1+q_3=q_2+q_4},
\]

\[
L_{4,A_{1u}} = \frac{7N_0\zeta(3)}{16(\pi k T_c)^2 D_0^2} \sum_{|q|} \Delta_3^*(q_1)\Delta_3^*(q_3)\Delta_3(q_2)\Delta_3(q_4)\delta_{q_1+q_3=q_2+q_4},
\]

where the summation \( |q| \) is for \( q_1, q_2, q_3, q_4 \). For the study of the FFLO state, it turns out that one also needs to take into account the coupling between the \( A_{1g} \) and \( A_{1u} \) pairing for the fourth order term, which is given by

\[
L_{4,A_{1g}-A_{1u}} = \frac{7N_0\zeta(3)}{16(\pi k T_c)^2 D_0^2} \sum_{|q|} \left( \Delta_1^*(q_1)\Delta_3^*(q_3)\Delta_1(q_2)\Delta_3(q_4) + \Delta_1^*(q_1)\Delta_3^*(q_1)\Delta_1(q_3)\Delta_3(q_4) + \Delta_1^*(q_1)\Delta_3^*(q_3)\Delta_1(q_2)\Delta_3(q_4) + \Delta_1^*(q_1)\Delta_3^*(q_1)\Delta_1(q_3)\Delta_3(q_4) \right) \delta_{q_1+q_3=q_2+q_4}.
\]

Collecting all the above terms, we find the fourth order term for the \( A_{1g} \) and \( A_{1u} \) pairings can be written as a compact form

\[
L_4 = \frac{7N_0\zeta(3)}{32(\pi k T_c)^2 D_0} \sum_{|q|,a=\pm} \left( \Delta_1^*(q_1) + a \frac{\beta_{1o}}{D_0} \Delta_3^*(q_1) \right) \left( \Delta_1(q_3) + a \frac{\beta_{1o}}{D_0} \Delta_3(q_3) \right) \delta_{q_1+q_3=q_2+q_4},
\]

In the limit \( t \to 0 \), the fourth order term is reduced to the form

\[
L_4 = \frac{7N_0\zeta(3)}{8(\pi k T_c)^2 D_0} \sum_{|q|,a=\pm} \Delta_1^*(q_1)\Delta_1^*(q_3)\Delta_1(q_2)\Delta_1(q_4) \delta_{q_1+q_3=q_2+q_4},
\]

which is also decoupled between the top and bottom layers. In combining with the form of \( L_2 \) in Eq. (6) in the main text, we conclude that the Landau free energy is decoupled between the top and bottom layers for the limit \( t \to 0 \).

We consider the fourth order term for \( \Delta_+(\pm q_c) \), which takes the form

\[
L_4 = \frac{7N_0\zeta(3)}{8(\pi k T_c)^2 D_0^2} \sum_{a=\pm} \left( |\Delta_+(q_c)|^4 + |\Delta_-(q_c)|^4 + 4|\Delta_+(q_c)|^2|\Delta_-(q_c)|^2 \right)
\]

Since only \( \Delta_+(q_c) \) and \( \Delta_-(q_c) \) are favored by the second order term, the fourth order term involving \( \Delta_+(q_c) \) and \( \Delta_-(q_c) \) is given by

\[
L_4 = \frac{7N_0\zeta(3)}{16(\pi k T_c)^2 D_0^2} \left( |\Delta_+(q_c)|^2 + |\Delta_-(q_c)|^2 \right)^2 + \left( |\Delta_+(q_c)|^2 - |\Delta_-(q_c)|^2 \right)^2.
\]

Compared with Eq. (D31), we find \( B_t = B_o = \frac{7N_0\zeta(3)}{16(\pi k T_c)^2 D_0^2} \). Since \( B_o > 0 \), we conclude that stripe phase will be favored when the temperature is close to \( T_c \).
3. Phase transition between the BCS superconducting state and the FFLO state

The phase diagram of the gap function as a function of magnetic fields and temperature is shown in the main text. Here we will present more analytical results and show that the transition between the uniform $A_{1g}$ pairing and the FFLO state is of the first order.

By solving the gap equation [D21], we can obtain the $T_c$ as a function of momentum $q$ and magnetic field $B$. Since the state with $q_x=0$ will always be favored, we focus on $q_y$ here. The critical temperature is an even function of $q_y$ and it can be expanded as $T_c(q_y) = T_{c0} + T_{c1}q_y^2 + T_{c2}q_y^4$, up to the fourth order in $q_y$, where $T_{c2} < 0$. The transition between uniform superconductivity and FFLO state occurs when $T_{c1}$ changes from negative to positive. Since the transition is tuned by magnetic field $B$, the parameter $T_{c1}$ takes the form $T_{c1} = S_1(B_x - B_{c0})$, where $S_1 > 0$ and $B_{c0}$ labels the critical magnetic field. For a positive $T_{c1}$, the maximum $T_c$ is achieved when $q_y = \pm \frac{S_1}{2T_{c2}}(B_{c0} - B_x)$ and the corresponding $T_c$ is $T_c(q_y) = T_{c0} - \frac{S_1^2}{4T_{c2}^2}(B_{c0} - B_x)^2$. In addition, $T_{c0}$ should be an even function of magnetic field, $T_{c0} = R_1 - R_2B^2$. We focus on the magnetic field around $B_{c0}$ and thus expand $T_{c0}$ as $T_{c0} = \hat{T}_{c0} - 2R_2B_{c0}\delta B$ with $\hat{T}_{c0} = R_1 - R_2B^2_{c0}$. Furthermore, the eigen-vector for the corresponding $T_c$ in Eq. [D21] is simplified as

$$\left(\frac{\Delta_1}{\Delta_3}\right) = \Delta_0 \begin{pmatrix} 1 \\ 0 \end{pmatrix}$$

(D39)

around $B_{c0}$ since the $A_{1g}$ pairing will always dominate and the amplitude $\Delta_0$ is to be determined. With these simplifications, we are able to evaluate the free energy analytically. The second order term is derived as

$$L_2 = 2N_0 \sum_{q_y} |\Delta_0q_y|^2 \ln \left(\frac{T}{T_{c0}}\right) \left(1 + \frac{V_0q^2}{U_0D_0}\right)^2.$$

(D40)

The main difference between zero momentum and finite momentum pairings lies in the fourth order term.

For zero momentum pairing ($T_{c1} < 0$), the fourth order term is given by

$$L_4 = \frac{7N_0\zeta(3)}{16\pi kT_{c0}^2} |\Delta_{0,0}|^4.$$

(D41)

By minimizing $L_2 + L_4$ for $q_y = 0$, we find the minimal free energy is given by

$$\mathcal{F}_{q_y = 0} = -\frac{16N_0(\pi kT_{c0})^2}{\zeta(3)} \left(\ln \left(\frac{T}{T_{c0}}\right)\right)^2 \left(1 + \frac{V_0q^2}{U_0D_0}\right)^4.$$

(D42)

as a function temperature $T$, which is assumed to be close to $T_{c0}$.

On the other hand, for a non-zero $q_y$, the fourth order term is much more complicated. The $q_1, q_2, q_3$ and $q_4$ in Eq. [D35] can take the following six cases: (1) $q_1 = q_2 = q_3 = q_4 = q_1$; (2) $q_1 = q_2 = q_3 = q_4 = -q_1$; (3) $q_1 = q_2 = -q_3 = -q_4 = q_1$; (4) $q_1 = -q_2 = q_3 = -q_4 = q_1$; (5) $q_1 = -q_2 = -q_3 = q_4 = q_1$; (6) $q_1 = q_2 = -q_3 = -q_4 = q_1$. Furthermore, we only focus on the stripe phase, which has been confirmed in numerical calculations. Thus, we take $|\Delta_{0,-q}| = |\Delta_0q|$ and as a result, the full Landau free energy takes the form

$$L = 4N_0|\Delta_0|^2 \ln \left(\frac{T}{T_{c0}}\right) \left(1 + \frac{V_0q^2}{U_0D_0}\right)^2 + \frac{21N_0\zeta(3)}{8(\pi kT_{c0}(q_y))^2} |\Delta_0q|^4,$$

(D43)

the minimal of which gives the temperature dependence of the free energy

$$\mathcal{F}_{q_y = 0} = \frac{32N_0(\pi kT_{c0}(q_y))^2}{21\zeta(3)} \left(\ln \left(\frac{T}{T_{c0}}\right)\right)^2 \left(1 + \frac{V_0q^2}{U_0D_0}\right)^4.$$

(D44)

Now let’s fix the temperature $T$ smaller than $T_{c0}$ and study the phase transition by varying magnetic field $B$. The transition happens at $B_{c0}$ for the temperature at $T_{c0}$ but will shift a bit away when the temperature $T$ is lower than $T_{c0}$. To see that, we need to consider the limit of $q_y \to 0$. In this limit, $T_{c0}(q_y) \to T_{c0}$ and thus

$$\mathcal{F}_{q_y \to 0} = \frac{-32N_0(\pi kT_{c0}(q_y))^2}{21\zeta(3)} \left(\ln \left(\frac{T}{T_{c0}}\right)\right)^2 \left(1 + \frac{V_0q^2}{U_0D_0}\right)^4 = \frac{2}{3} \mathcal{F}_{q_y = 0} > \mathcal{F}_{q_y = 0}.$$

(D45)
Thus, at $B_c = B_{c0}$, the zero momentum pairing is more stable. The difference comes from the form of fourth order term when $q_y = 0$ and $q_x \neq 0$. To determine the critical magnetic field at $T$, we may expand the free energy around $B_{c0}$. With $B_c = B_{c0} + \delta B$, we find

$$F_{q_y=0} = -\frac{16N_0(\pi k)^2}{7\zeta(3)} \left( 1 + \frac{V_0q_y^2}{U_0D_0^2} \right)^2 \left( \ln \left( \frac{T}{T_{c0}} \right) \right)^2 T_{c0}^2 + 4R_2B_{c0}T_{c0}ln \left( \frac{T}{T_{c0}} \right) \left( 1 - \ln \left( \frac{T}{T_{c0}} \right) \right) \delta B$$

(D46)

and

$$F_{q_y=0} = -\frac{32N_0(\pi k)^2}{21\zeta(3)} \left( 1 + \frac{V_0q_y^2}{U_0D_0^2} \right)^2 \left( \ln \left( \frac{T}{T_{c0}} \right) \right)^2 T_{c0}^2 + 4R_2B_{c0}T_{c0}ln \left( \frac{T}{T_{c0}} \right) \left( 1 - \ln \left( \frac{T}{T_{c0}} \right) \right) \delta B$$

(D47)

up to the first order in $\delta B$. Thus, the critical magnetic field is determined by

$$\delta B_c = -\frac{\ln \left( \frac{T}{T_{c0}} \right) T_{c0}}{4R_2B_{c0} \left( 1 - \ln \left( \frac{T}{T_{c0}} \right) \right)}$$

(D48)

when $T \rightarrow T_{c0}$ and $\ln \left( \frac{T}{T_{c0}} \right)$ is treated as a small number.

At the $\delta B_c$, the first derivative of the free energy is given by

$$\frac{\partial F_{q_y=0}}{\partial \delta B} = -\frac{16N_0(\pi k)^2}{7\zeta(3)} \left( 1 + \frac{V_0q_y^2}{U_0D_0^2} \right)^2 4R_2B_{c0}T_{c0}ln \left( \frac{T}{T_{c0}} \right)$$

(D49)

and

$$\frac{\partial F_{q_y=0}}{\partial \delta B} = -\frac{32N_0(\pi k)^2}{21\zeta(3)} \left( 1 + \frac{V_0q_y^2}{U_0D_0^2} \right)^2 4R_2B_{c0}T_{c0}ln \left( \frac{T}{T_{c0}} \right)$$

(D50)

up to the first order in $\ln \left( \frac{T}{T_{c0}} \right)$. We find $\frac{\partial F_{q_y=0}}{\partial \delta B} \neq \frac{\partial F_{q_y=0}}{\partial \delta B}$ at $\delta B_c$, thus confirming the phase transition is of the first order nature.

4. FFLO/BCS Josephson junction

Next we will discuss the possible detection of the FFLO state. A Josephson junction between the FFLO state and a conventional superconductor is considered, as shown in Fig. 1d in the main text. The Josephson current in this system is given by

$$I_J = Im \left( \int dy \Psi^*_BCS(y)\Psi_{FFLO}(y) \right)$$

(D51)

where $\Psi_{FFLO}$ and $\Psi_{BCS}$ are the pair functions for the FFLO state and conventional superconductors. $t$ is the hopping parameter. We take the form $\Psi_{BCS} = \Psi_0e^{i\varphi}$, where $\varphi$ is the phase factor, and $\Psi_{FFLO} = \Psi_+e^{iqy} + \Psi_-e^{-iqy}$ according to the form of the gap function. With these forms of the pair functions, the Josephson current is found to be

$$I_J = \sum_{\alpha=\pm} I_{c,\alpha} \sin(\alpha q, L/2) \sin \varphi$$

(D52)

where the $y$-direction integral is taken from $-L/2$ to $L/2$ and thus the maximum supercurrent is

$$I_m = \left| \sum_{\alpha=\pm} I_{c,\alpha} \sin(\alpha q, L/2) \right|$$

(D53)

The maximum supercurrent will depend on the details of Josephson contact, and if we assume $I_{c,+} = I_{c,-} = I_{c0}$, we find

$$I_m = 2I_{c0} \frac{\sin(q, L/2)}{(q, L/2)}$$

(D54)

Since $q \propto B_c$, the maximum supercurrent reveals an interference pattern for in-plane magnetic fields, just like the Fraunhofer pattern in a normal Josephson junction under out-of-plane magnetic fields.30

For a large in-plane magnetic field with $q_L \ll 1$, $I_m$ will decrease to zero, but an additional magnetic field along the surface normal will lead to an asymmetric Fraunhofer pattern.15
5. $E_u$ pairing

Finally, we discuss the orbital effect of the $E_u$ pairing. Direct calculation shows that up to the second order in $v_Q$ and $q$, 

$$
\chi_{E_u,11} = \chi_{E_u,22} = 2N_0 \left( \ln \left( \frac{2\gamma Q D}{\pi k T} \right) \left( \frac{2 \beta_{tt}^2}{D_0^2} + \frac{t^4 - 5F_{tt}^2}{2D_0^2} \right) - C\beta_{tt}^2 \left( \frac{1}{2\pi k T} \right)^2 \left( \frac{2}{2} + \beta_{tt}^2 \right) \right) \tag{D55}
$$

and

$$
\chi_{E_u,12} = \chi_{E_u,21} = 0. \tag{D56}
$$

As a consequence, the corresponding Landau free energy still follows the standard form with the $T_c$ given by

$$
ln \left( \frac{T_{c, E_u}}{T_{c,0, E_u}} \right) = \frac{t^4 - 5F_{tt}^2}{4\beta_{tt}^2 D_0^2} \ln \left( \frac{2\gamma Q D}{\pi k T} \right) (hv_Q k_f)^2. \tag{D57}
$$

Appendix E: The orbital effect of out-of-plane magnetic fields

Finally, we would like to comment about the orbital effect of out-of-plane magnetic fields. We consider the form of Landau free energy in the real space as

$$
L = \int dr \left( K|\nabla \Delta(r)|^2 + A|\Delta(r)|^2 \right) \tag{E1}
$$

where $A = Cln \left( \frac{T}{T_{c,0}} \right)$. The orbital effect of magnetic fields is taken into account by $-i\nabla \to D = -i\nabla + \frac{2e}{h} A$, where the vector potential $A$ can be chosen as $A = (0, B_z, 0)$. The corresponding linearized gap equation is given by

$$
(K\Delta^2 + A) \Delta(r) = 0. \tag{E2}
$$

This is nothing but the Landau level problem, which can be solved exactly by introducing the boson operators

$$
a = \sqrt{\frac{\hbar}{4eB_z}} (\pi_x - i\pi_y),
$$

$$
a^\dagger = \sqrt{\frac{\hbar}{4eB_z}} (\pi_x + i\pi_y), \tag{E3}
$$

where $\pi_{x(y)} = -i\partial_{x(y)} + \frac{2e}{h} A_{x(y)}$. The above gap equation can be simplified as

$$
\left( \frac{4eB_z}{\hbar} K(a^\dagger a + \frac{1}{2}) + A \right) = 0. \tag{E4}
$$

The lowest eigen-energy of the above equation is

$$
\frac{2eB_z}{\hbar} K + A = 0, \tag{E5}
$$

leading to the correction

$$
ln \left( \frac{T_c}{T_{c,0}} \right) = \frac{2e}{\hbar} K \frac{B_z}{C}, \tag{E6}
$$

which is linear in $B_z$. Therefore, the $T_c$ correction from the out-of-plane magnetic fields is determined by the ratio $\frac{K}{C}$. By looking at the parameters in Landau free energy, we find for the representation $i \ (i = A_{1x}, A_{1y}, E_u)$, the ratio is given by $\frac{K}{C} = \frac{C}{6} \left( \frac{hv}{2\pi k T_{c,0}} \right)^2$.

Therefore, the correction is determined by $T_{c,0,i}$, and a weaker $T_{c,i}$ correction for the pairing with higher $T_{c,0,i}$. The stable superconducting phase will always be stable under an out-of-plane magnetic field.

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