Theory of orbital d_*-vector in a two-band spin-singlet superconductor: application to nematic superconductivity

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We study even-parity spin-singlet orbital-triplet pairing states in a two-band superconductor. An orbital d_*-vector is introduced to characterize such pairings, in analogy to the spin d_s(k)-vector describing spin-triplet pairings in 3He superfluid. Naively, one might think the double degeneracy of orbitals would be lifted by orbital hybridizations due to the crystal field splitting or electron-electron repulsive interactions, then spin-singlet orbital-dependent pairings may be severely suppressed. However, we demonstrate that these pairings are not excluded in real materials and a corresponding orbital d_*-vector could be stabilized along certain axis in orbital subspace. Even more remarkably, the interplay between the many-body interaction induced nematic order and the superconducting order leads to the establishment of a nematic orbital d_*-vector, which gives rise to the coexistence of nematicity and superconductivity. The generalization to a single-band superconductor with two valleys (e.g. honeycomb lattice with two sublattices) is also discussed. The nematic superconductivity in both FeSe and magic-angle twisted bilayer graphene might be interpreted within our theory framework. Moreover, the complex orbital d_*-vector spontaneously breaks time-reversal symmetry (TRS), which may induce the TRS-breaking orbital-polarization, analogous to the spin magnetism.

Introduction.— The unconventional superconductivity remains an important topic in condensed matter physics. Other than the well-known d-wave pairing state in single-orbital high-temperature cuprate superconductors (SCs), the multi-orbital electronic systems may also support a rich phase diagram hosting orbital-dependent pairing states. These have been studied in a wide variety of multi-band systems, including Sr2RuO4, iron-based SCs, Cu-doped Bi2Se3, half-Heusler compounds. Moreover, spontaneous rotational symmetry breaking in SCs is another crucial topic that has attracted tremendous interest when many-body electron-electron interactions are appreciable. Among them, both spontaneous rotational symmetry breaking (i.e., nematic SC) and time-reversal symmetry (TRS) breaking are of special interest.

The orbital-dependent pairing in a spin-singlet SC with two orbitals {1, 2}, for example, c_{1,1} (k)c_{2,1} (−k) − [↑↑↓↓], may play a crucial role in unconventional SCs. Compared to spin-triplet pairings, spin-singlet orbital-triplet pairings have been much less explored since such pairings are usually considered to be energetically unfavorable. This is partly due to the common belief that the double degeneracy of the two orbitals might be lifted by orbital hybridizations so that orbital-dependent pairings would be severely suppressed under crystal field splittings or electron-electron repulsive interactions. One of the aims of this work is concerned with the possible existence of spin-singlet orbital-dependent pairings.

To address the problem, we adopt an orbital d_*-vector formalism to describe the spin-singlet orbital-dependent pairings in analogy to the spin d_s(k)-vector of spin-triplet SCs. The crystal field splitting of these two orbitals gives rise to the orbital hybridization depicted by a g_*-vector. We find that the presence of the g_*-vector generally suppresses the superconductivity with orbital d_*-vector except for d_s(k) ≈ g_*-vector. Within the mean-field theory for electron-electron repulsive interactions, the nematic order develops for T < Tnem, which contributes to the total orbital hybridization, gtot = g_o + g_nem. This leads to the stabilization of the nematic orbital d_*-vector for d_s(k) ≈ gtot-vector, which implies the coexistence of nematicity and superconductivity. The potential application of our theory to nematic SCs in both FeSe and magic-angle twisted bilayer graphene is also discussed. Furthermore, we also investigate the spontaneous TRS-breaking pairings by using the Ginzburg-Landau (GL) theory.

Spin d_*-vector in spin-triplet SCs.— We start with a brief review of single-band spin-triplet SCs. The spin-triplet pairing potential is generally given by Δ(k) ∝ (d_s(k) · σ)[iσ2], where σ are Pauli matrices in the spin subspace. Due to the Fermi statistics, the spin d_s(k)-vector has to satisfy d_s(k) = −d_s(−k). In the 3He superfluid, the d_s-vector can rotate freely due to the spin-rotation symmetry. However, in noncentrosymmetric SCs, the spin d_s(k)-vector is usually pinned along a certain crystal axis, since superconductivity is suppressed only for d_s(k) ≈ g_*-vector, where g_*-vector represents the Rashba spin-orbit coupling (SOC). Besides,
there is intrinsic spontaneous spin-polarization induced by the non-unitary pairing, \( d_a(\mathbf{k}) = k_z (1, -i\eta_0, 0) \) with real \( \eta_0 \). Fig. 1(a) shows the spin expectation value of the Cooper pairs \( (M_s \propto \mathbf{d}_s \times \mathbf{d}_s^*) \), where \( \mathbf{d}_s \) is the creation operator of Cooper pairs. \( \sigma_2 \) is indices for spins and \( a, b \) are for orbitals \{1, 2\}. The spin-singlet pairing function \( \Delta^{a,b}_{s_1 s_2}(\mathbf{k}) = f(\mathbf{k})M_{a,b}(\sigma_2 s_1 s_2) \) consists the angular form factor \( f(\mathbf{k}) \) and \( M_{a,b} \) in the orbital channel. The spin-singlet pairings are not mixed with spin-triplet pairings in the absence of SOC. In analogy to spin-triplet SCs, we then use an orbital \( \mathbf{d}_a(\mathbf{k}) \)-vector for the spin-singlet orbital-dependent pairing potential \[3\].

\[
\mathcal{H}_\Delta = \sum_{\mathbf{k}} \sum_{s_1 s_2 a b} \Delta^{a,b}_{s_1 s_2}(\mathbf{k}) F^\dagger_{s_1 s_2 a b}(\mathbf{k}) + \text{h.c.},
\]

where \( F_{s_1 s_2 a b}(\mathbf{k}) = c_{s_1 a}(\mathbf{k}) c_{s_2 b}^\dagger(-\mathbf{k}) \) is the creation operator of Cooper pairs, \( s_1, s_2 \) are indices for spins and \( a, b \) are for orbitals \{1, 2\}. The spin-singlet pairing function \( \Delta^{a,b}_{s_1 s_2}(\mathbf{k}) = f(\mathbf{k})M_{a,b}(\sigma_2 s_1 s_2) \) consists the angular form factor \( f(\mathbf{k}) \) and \( M_{a,b} \) in the orbital channel. The spin-singlet pairings are not mixed with spin-triplet pairings in the absence of SOC. In analogy to spin-triplet SCs, we then use an orbital \( \mathbf{d}_a(\mathbf{k}) \)-vector for the spin-singlet orbital-dependent pairing potential \[3\],

\[
\Delta^{ab}_{s_1 s_2}(\mathbf{k}) = \frac{1}{\beta} \sum_{\omega_n} \sum_{s_1' a' s_2' b'} V_{s_1' a' s_2' b'}(\mathbf{k}, \mathbf{k}') \times

\left[ G_{c}(\mathbf{k}', i\omega_n) \Delta_{c}(\mathbf{k}) G_{h}(\mathbf{-k}', i\omega_n) \right]_{s_1' a' s_2' b'}.
\]

where \( \beta = 1/k_BT \), \( G_{c}(\mathbf{k}, i\omega_n) = -G_{h}(\mathbf{k}, i\omega_n) \) is the Matsubara Green’s function for electrons with \( \omega_n = (2n + 1)\pi/\beta \). We expand the attractive interactions as \( V_{s_1 s_2 a b}(\mathbf{k}, \mathbf{k}') = -\frac{1}{\epsilon_0} \sum_{n_1 n_2} \epsilon_1 n_1 \epsilon_2 n_2 d_{s_1 s_2 a b}(\mathbf{k}) \cdot \epsilon_1 n_1 \epsilon_2 n_2 d_{s_1 s_2 a b}(\mathbf{k}) \cdot \tau \sigma_1 \sigma_2 d_{s_1 s_2 a b}(\mathbf{k}) \), where \( \epsilon_0 > 0 \). Here \( \Gamma \) labels the irreducible representation with \( m \)-dimension of crystalline groups. To the leading order of \( \lambda_k k^2 / \mu \), the equation for \( T_c \) reads (see details in Sec. (C. 1) of the SM [65]),

\[
\ln \left( \frac{T_c}{T_F} \right) = \int_{\mathbb{B}} d\Omega C_0(T_c) \left( |\mathbf{d}_a|^2 - |\mathbf{d}_a \cdot \mathbf{g}_o|^2 \right),
\]

where \( T_F \) is the Fermi temperature, \( \epsilon_0 = \mathbf{g}_o \cdot \mathbf{d}_a(\mathbf{k}) \) are normalized vectors. Here we take \( \int_{\mathbb{B}} d\Omega |\mathbf{d}_a|^2 = 1 \). And \( C_0(T) = \text{Re}\psi^0(\frac{1}{2}) - \psi^0(\frac{1}{2} + \frac{1}{T_F T}) \) is the digamma function. Since \( C_0(T_c) \leq 0 \) and it monotonically decreases as \( \lambda_k \) increases, the right-hand side of Eq. (5) suppresses \( T_c \) in general. However, \( \mathbf{d}_a \parallel \mathbf{g}_o \) can lead to \( T_c = T_{ct} \) for any value of \( \lambda_k \), which indicates that the orbital \( \mathbf{d}_a \)-vector that is parallel with \( \mathbf{g}_o \) is unaffected by the orbital hybridizations. Choosing \( \mathbf{g}_o(\mathbf{k}) = (2k_x k_y, 0, k_z - k^2_0) \),
the numerical results are shown in Fig. 2. The black line confirms that $T_c$ is unaffected as $\lambda_o k_F^2/k_B T_C$ increases for $d_\perp(k) = (2k_x k_y, 0, k_x^2 - k_y^2)$, which is the unconventional $A_{1g}$ pairing. However, for other $d_o$-vectors are severely suppressed. This is similar to spin-triplet SCs, where the $A_{1g}$-type $d_o$-vector could exist in SCs with two active orbitals that are not fully degenerate. This is also in agreement with the results from Eq. (2) of the SM [65].

Next, we include $\Delta_o$ and investigate the coupling between $\Psi_s$ and $d_o$. Solving the coupled linearized gap equations up to $(\lambda_o k_F^2/\mu)^2$ order (see details in Sec. (C.2) of the SM [65]), we find that the results from Eq. (5) are still correct. Besides, the magnitude of orbital $d_o$-vectors might be determined as $d_o(k) = \Psi_s(k) g_o(k)$. It implies that $\Psi_s$ and $d_o$ belong to the same representation of crystalline groups.

We now explain Eq. (5) from the band picture. Within the band basis, the pairing potential in the orbital subspace becomes $\Delta_o(k) = U(k) \Delta_s(k) \tau_0 + \Delta_o(d_o(k) \cdot \tau) U(k)$, where $U(k)$ is the unitary matrix in the orbital subspace, $U(k) \Delta_s(k) \tau_0 = \Delta_o(d_o(k) \cdot \tau) U(k)$, which is the diagonal part of $[\Delta_o d_o(k) \cdot \tau] U(k) = \text{Diag}[E_+(k), E_-(k)]$, with $E_\pm(k) = \epsilon(k) \pm \lambda_o g_o(k)$. Then, we decompose the orbital $d_o$-vector, $d_o(k) = d_o(\perp) g_o(k) + d_o(\parallel)$, where $d_o(\perp) = d_o(\perp) \cdot g_o(k)$ and $d_o(\parallel) \cdot g_o(k) = 0$. We find that the $d_o(\perp)$-part gives rise to the inter-orbital pairing, while the $d_o(\parallel)$-part leads to the inter-band pairing (see Sec. (D) in the SM [65]). If the band splitting is much larger than the pairing gap $(\lambda_o k_F^2 \gg \Delta_o)$, the inter-band pairing is not energetically favorable in the weak-coupling pairing limit. It means that the inter-band pairing will be severely suppressed if we increase the orbital hybridization $\lambda_o$, consistent with Eq. (5) and results in Fig. 2.

Applications to nematic superconductivity. – Next, we study the stability of orbital $d_o$-vectors against on-site inter-orbital repulsive interaction, $H_{\text{int}} = \epsilon_i n_i(r) n_{i\sigma}(r)$, where $n_i$ is electron density operator for the $i$-atomic orbital. Using the mean-field theory, $C_n$ ($n > 2$) is spontaneously broken down to $C_2$ once $\langle \hat{g}_o(\theta) \rangle$ is non-zero. Therefore, the nematic orbital $d_o$-vector satisfies $d_o \parallel g_{\text{tot}}$ leads to the nematic superconductivity.
where \( g_{\text{int},1}(k) = \text{Re}[\Phi(k)] \) and \( g_{\text{int},2}(k) = -\text{Im}[\Phi(k)] \). In this case, TRS is \( T = i\tau_1\sigma_2 K \) and IS is \( T = \tau_1\sigma_0 K \).
The \( \mathbf{d}_s \)-vector is manifested as \( \mathbf{d}_s = (d_1(k), id_2(k), 0) \) with \( d_1(k) = d_1(-k) \) and \( d_2(k) = -d_2(-k) \). Both \( d_1(k) \) and \( d_2(k) \) are real to preserve TRS. As for the interaction-induced \( g_{\text{int},1} \), \( T \) and IS require \( g_{\text{int},1}(k) = g_{\text{int},1}(-k) \) and \( g_{\text{int},2}(k) = 0 \). By symmetry, there are two general possibilities. One is \( g_{\text{int},1}(k) = 1 \), so \( C_3 \) is preserved, and it describes the charge-density-wave order. The other one is \( g_{\text{int},1}(k) \notin \{k_xk_y, k_x^2 - k_y^2\} \) that spontaneously breaks \( C_3 \) down to \( C_2 \), forming a nematic order.

We next discuss the existence of superconductivity and inter-valley couplings, by replacing the \( g_{\alpha} \)-vector with the interaction-induced \( g_{\text{int},1} \) in Eq. (3). As a result, Eq. (3) is still applicable. For example, the Ising SCs in gated MoS\(_2\) \([72, 73]\) is characterized by \( \mathbf{d}_0(k) = (1,0,0) \), which apparently coexists with charge-density-wave order, \( g_{\text{int},1}(k) = (1,0,0) \). Our results may explain the experimental observation in the Pd-doped 2H-TaSe\(_2\) in Ref. [74]. On the other hand, the nematic inter-valley coupling is represented as \( g_{\text{int},1}(k) = \alpha_0 + 2\alpha_1k_xk_y + \alpha_2(k_x^2 - k_y^2) \), which requires that \( \mathbf{d}_s(k) = (\alpha_0 + 2\alpha_1k_xk_y + \alpha_2(k_x^2 - k_y^2), 0, 0) \) (see Sec. (F) in the SM [65]), which might help to understand the experimental observation for nematic superconductivity in magic-angle magic-angle graphene \([53, 54]\). From the viewpoint of our theory, it may be a \((s+d)\)-wave inter-valley pairing state. The system is fully gapped if the \( s\)-wave gap is dominant \( (\alpha_0 \gg \sqrt{\alpha_1^2 + \alpha_2^2}) \), otherwise, it is a \( d\)-wave dominant nodal SC \( (\alpha_0 \ll \sqrt{\alpha_1^2 + \alpha_2^2}) \). In the above analysis, the layer dependence has been neglected \([75, 76]\), then a more careful study is left for future work. Besides, the non-zero valley polarization \( (g_{\text{int},3} \neq 0) \) is also symmetry-allowed and would generally suppress the superconductivity.

To proceed, we briefly discuss the difference between our theory and the previous studies \([34]\) for nematic SCs. One example a pairing state belonging to a 2D irreducible representation (Irrep), e.g., the \( E\)-pairing in Cu or Nd-doped Bi\(_2\)Se\(_3\) \([77, 81]\) and UPt\(_3\) \([82, 83]\). A real order parameter vector \((\Delta_{E,1}, \Delta_{E,2})\) spontaneously breaks \( C_3 \), leading to nematic superconductivity. Alternatively, a nematic SC can be formed by mixing two different 1D-Irrep-pairing channels. In FeSe \([84, 86]\), the nematic order breaks the \( C_3 \) down to \( C_2 \), which mixes the \( s\)-wave and \( d\)-wave pairing channels. However, \( T_c \) of the \((s+d)\) orbital-independent pairing state could be generally affected with increasing nematicity. In our theory, the \((s+d)\)-like nematic \( \mathbf{d}_s \)-vector coexists with the nematic order, so \( T_c \) is almost unaffected with increasing nematicity. Therefore, it helps to distinguish our results from previous proposals in experiments, where one may use the chemical or physical pressures to tune the nematicity and measure \( T_c \) as a function of pressure \([87]\).

**Spontaneous TRS-breaking orbital-polarization.**—Next, we study spontaneous TRS-breaking and its consequences for a two-band SC with \( \{d_{xz}, d_{yz}\}\)-orbitals. \( T = \tau_1\sigma_0 \) constrains the orbital \( \mathbf{d}_s \)-vector to be \((d_1\Psi_0^s(k), 0, d_3\Psi_0^s(k))\) for Eq. (2). Under \( C_n \) \((T)\), the orbital \( \mathbf{d}_s \) transforms as \( \mathbf{d}_s \to e^{i2\pi n/\sigma}\mathbf{d}_s \) \((\mathbf{d}_s \to -\mathbf{d}_s)\). Choosing \( |d_1|^2 + |d_3|^2 = 1 \), the set of superconducting order parameters are given by \( \{\Delta_x, \Delta_y, \mathbf{d} \equiv (d_1, 0, d_3)\} \). Furthermore, the orbital orderings can be characterized by \( M_y \propto \sum_{k,\sigma} \langle c_{\sigma a}^\dagger(k) \tau_{0b} c_{\sigma b}(k) \rangle \) (details in Sec. (G) of the SM [65]). The total GL free energy preserving the \( U(1) \times T \times C_n \times \mathcal{I} \) symmetries is constructed as,

\[
\mathcal{F}[\Delta_x, \Delta_y, \mathbf{d}, M_y] = \mathcal{F}_1 + \mathcal{F}_2 + \mathcal{F}_3 + \mathcal{F}_4,
\]

where \( \mathcal{F}_1 = \frac{1}{2} \alpha_1(T)|\Delta_0|^2 + \frac{1}{2} \beta_1 |\Delta_0|^4 + \frac{1}{2} \alpha_2(T)|\Delta_x|^2 + \frac{1}{2} \beta_2|\Delta_x|^4 + \frac{1}{2} \alpha_3 |\Delta_y|^2 + \frac{1}{2} \alpha_4 |\Delta_y|^4 + \frac{1}{2} \alpha_5 |\Delta_x| |\Delta_y| \), determining the gap strengths by using \( \alpha_1(1,T) = \alpha_0(1,T/T_c^{1.2} - 1) \) and \( \alpha_3^2, \alpha_5 > 0, \beta_1 > 0 \), which determines \( \mathbf{d} \). In addition, there are two possibilities to achieve the spontaneously TRS-breaking states, which are described respectively by \( \mathcal{F}_3 \) and \( \mathcal{F}_4 \).

\[
\mathcal{F}_3 = b_1 \Delta_x^2 \Delta_y^2 + b_2 |\Delta_x|^2 |\Delta_y|^2 + \mathcal{F}_4 = \gamma_0(T)(\mathbf{d} \times \mathbf{d}^*) + i \gamma_1 M_y \cdot (\mathbf{d} \times \mathbf{d}^*) + \mathcal{F}_4.
\]

The \( \mathcal{F}_3 \) term in Eq. (9) helps to develop a relative phase difference between \( \Delta_x \) and \( \Delta_y \) of being \( \pm \pi/2 \) when \( b_1 = 0 \) and \( b_2 \geq 0 \) \([88]\). As for the \( b_2 < 0 \) case, the TRS-breaking is caused solely by the \( \mathcal{F}_4 \) term in Eq. (10). For example, \( \gamma_0(T) = \gamma_0(T/T_c^* - 1) \) and \( T_c^* < T_c \), where \( T_c^* \) is the critical temperature for the spontaneous TRS-breaking inside the superconducting states. When \( T < T_c^* \), the orbital \( \mathbf{d}_s \)-vector becomes complex, then it generates the orbital orderings as \( M_y = -\tau_1\sigma_y M_y (\mathbf{d} \times \mathbf{d}^*) \), of which only the \( y \)-component breaks TRS, as illustrated in Fig. 4(a). More precisely, \( M_y^y \sim \sum_{k,\sigma} (\hat{n}_{\sigma,++}(k) - \hat{n}_{\sigma,--}(k)) \). Here we define \( |z| = |1 + i2| \) for complex orbitals, thus \( M_y^y \neq 0 \) indicates the TRS-breaking orbital-polarization (OP).

We next solve the Bogoliubov-de-Gennes Hamiltonian, \( \mathcal{H}_{\text{BdG}}[\mathbf{E}_n(k)] = \mathcal{E}_n(k) [\mathbf{E}_n(k)] \) with \( \mathcal{E}_n(k) = (\nu_{n_{d_{xz},+}^x}^n \nu_{d_{xz},-}^n, \nu_{d_{yz},+}^n \nu_{d_{yz},-}^n) \) of the quasi-particle spectrum is plotted in Fig. 4(b), where two distinct gaps appear. Then, we calculate the atomic orbital-polarized density of states (DOS), \( D_n(E) = \frac{1}{2} \sum_{\sigma, n, k} |u_{\sigma, n}^k|^2 \delta (E - \mathcal{E}_n(k)) \), where
$u_{\alpha \sigma} = u_0^{\alpha \sigma} - i k u_0^{\alpha \sigma}$, with $\kappa = \pm$ for $d_{xz} \pm id_{yz}$ orbitals, and $\delta(x)$ is the delta function. The numerical results are shown in Fig. 3(c), confirming a two-gap feature due to the spontaneous breaking of TRS, compared with the quasi-particle spectrum in Fig. 3(b). Moreover, $D_+ \neq D_-$ at finite energy clearly indicates that the DOS is orbital-polarized, which is consistent with the GL analysis. The orbital-spin conversion would lead to the spin-polarized DOS $89$.

**Conclusion.**—To summarize, we have studied a phenomenological theory in a two-band spin-singlet SC. We adopt the orbital $d_\sigma$-vector to describe the spin-singlet orbital-dependent pairing states, in analogy to the spin $d_\sigma$-vector of spin-triplet SCs. We demonstrate that a orbital $d_\sigma$-vector that is parallel with $g_{\sigma}$-vector for orbital hybridizations is possible to exist in crystals. Remarkably, $d_\sigma$-vectors could even coexist with many-body interaction-induced nematic orders or charge-density-wave orders when $d_\sigma \propto g_{\text{tot}} = g_{\sigma} + g_{\text{nem}}$ (or $g_{\text{int}}$). Our theory discovers the important role of nematic orders in nematic SCs, and provides a possible understanding to nematic superconductivity in both FeSe and magic-angle twisted bilayer graphene. Based on the GL theory, a complex $d_\sigma$-vector that spontaneously breaks TRS may lead to the TRS-breaking orbital-polarization. Our work will motivate more theoretical and experimental efforts to search for spin-singlet orbital-triplet SCs.

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Supplementary materials for “Theory of orbital $d_o$-vector in a two-band spin-singlet superconductor: application to nematic superconductivity”

This supplemental material includes

- **Appendix A**: the discussion for different definitions of the orbital $d_o$-vector;
- **Appendix B**: the classification of spin-singlet pairing states with $C_n$ and TRS;
- **Appendix C**: the stability of orbital $d_o$-vector under crystal fields, derive Eq. (5) in the main text;
- **Appendix D**: the weak-coupling theory to understand Eq. (5) in the main text from the band picture;
- **Appendix E**: the application to nematic superconductor: FeSe superconductor;
- **Appendix F**: the application to nematic superconductor: single-layer graphene superconductor;
- **Appendix G**: the definition of spin and orbital magnetizations ($M_s$ and $M_o$).

**Appendix A: Two definitions for the orbital $d_o$-vector**

In the main text, we take the general pairing potential of a two-orbital SC,

$$\hat{\Delta}_{tot}(k) = (\Delta_s \Psi_s(k) \tau_0 + \Delta_o (d_o(k) \cdot \tau))(i\sigma_2),$$  \hspace{1cm} (A1)

where $\Delta_s$ and $\Delta_o$ are pairing strengths in orbital-independent and orbital-dependent channels, respectively. Here $\tau$ are Pauli matrices acting on the orbital subspace and $\tau_0$ is a 2-by-2 identity matrix. In the absence of band-splitting caused by spin-orbital couplings, the gap function on the Fermi surface is

$$\Delta(k) = \sqrt{|\Delta_s|^2 \Psi_s^2(k) + |\Delta_o|^2 |d_o(k)|^2 \pm |q_o|},$$  \hspace{1cm} (A2)

where $q_o = i|\Delta_o|^2(d_o^*(k) \times d_o(k)) + \text{Re}[\Delta_o^* \Delta_o d_o(k)]$. This expression is mathematically similar to the superconducting gap of non-unitary spin-triplet SCs.

At this point, it is a good place to comment on the other possible way to defining the orbital $d_o$-vector. Different from the one used in the main text, this definition groups the pairing term into orbital-singlet and orbital-triplet parts. In the form of Eq. (A1), $\Psi_s(k)$ and $d_o^{1,3}(k)$ are even in $k$, but $d_o^2(k)$ is odd in $k$ due to Fermi statistics. By regrouping the terms based on the parity in $k$, we have

$$\hat{\Delta}(k) = [\Delta_o d_o^2(k) \tau_0 + (i\Delta_o d_o^1(k), \Delta_s \Psi_s(k), i\Delta_o d_o^3(k)) \cdot \tau](\tau_2 i\sigma_2),$$  \hspace{1cm} (A3)

which contains $d_o \cdot \tau$ with the new $d_o$-vector redefined in terms of the original amplitudes and form factors. In this form, the first part is odd in $k$, which is the orbital-singlet part, and the second part is even in $k$ and gives orbital-triplet state. Table I gives a detailed comparison between the two definitions of the orbital $d_o$-vector. The spin $d_o$-vector is also presented for completeness. It shows that the definition of orbital $d_o$-vector used in the main text is more convenient to discuss the spontaneous TRS-braking pairing states.

**Appendix B: Classification of spin-singlet pairing states with $C_n$ and TRS**

In this section, we classify the possible spin-singlet pairing states constrained by $C_n$ about $z$-axis and TRS. The pairing potential $\hat{\Delta}(k)$ transforms under the rotation $C_n$ as

$$C_n \hat{\Delta}_J(k) C_n^T = e^{i\frac{2\pi}{n}J} \hat{\Delta}_J(C_n^{-1}k),$$  \hspace{1cm} (B1)

where $J$ labels the irreducible representations of the $C_n$ point group. For example, $J = 0$ is for $A$ representation and $J = 2$ is for $B$ representation. Firstly, the TRS requires the coexistence of $\Delta_J$ and $\Delta_{-J}$ with equal weight. If the rotation symmetry $C_n$ is further imposed, then $J$ and $-J$ have to be equivalent modulo $n$, i.e. $J \equiv -J \mod n$. The results for the basis functions of $\Psi_s(k)$ and $d_o(k)$ are summarized in Table II. Here, the $k_z$-dependent pairing symmetries are also presented for completeness. However, such pairings are neglected in the main text where we mainly focus on 2D systems.

When inversion symmetry is also present, it leads to the following constraints for different orbital basis,

1.) If the inversion symmetry is $I = \tau_0 \sigma_0$, two atomic orbitals have the same parity, it require that $d_o^2 = 0$.

2.) If the inversion symmetry is $I = \tau_1 \sigma_0$, two atomic orbitals have opposite parities, it require that $d_o^1 = 0$.

3.) If the inversion symmetry is $I = \tau_1 \sigma_0$, two orbitals are the valley indexes, it require that $d_o^3 = 0$. 

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TABLE I. Comparison between the two possible definitions of the orbital $d_o$-vector in spin-singlet SCs, together with the spin $d_s$-vector of spin-triplet SCs. The parity properties are obtained from Fermi statistics. The TRS row gives the transformation properties in order to preserve TRS. Both the atomic orbital polarization (AOP) and the spin polarization (SP) take the same form in terms of their respective $d$-vectors.

| $C_n$  | $J = -J \ (\text{mod } n)$ | $\Psi_s(k) = \Psi_s(-k)$ | $d_s^1(k) = d_s^1(-k)$ | $d_s^2(k) = -d_s^2(-k)$ | $d_s^3(k) = d_s^3(-k)$ |
|-------|-----------------|-----------------|-----------------|-----------------|-----------------|
| $n = 2$ | $J = 0$ | $1, k_x^2 + k_y, k_x k_y$ | $k_x, k_x k_y$ | $k_x, k_x k_y$ | $k_x, k_x k_y$ |
| | $J = 1$ | $k_x, k_x k_y$ | $k_x, k_x k_y$ | $k_x, k_x k_y$ | $k_x, k_x k_y$ |
| $n = 3$ | $J = 0$ | $1, k_x^2 + k_y, k_x k_y$ | $E_g$ representation | $k_x$ | $E_g$ representation |
| | $J = 2$ | $k_x^2 + k_y, k_x k_y$ | $k_x, k_x k_y$ | $k_x, k_x k_y$ | $k_x, k_x k_y$ |
| $n = 4$ | $J = 0$ | $1, k_x^2 + k_y, k_x k_y$ | $k_x^2 - k_y, k_k$ | $k_x, k_x k_y$ | $k_x, k_x k_y$ |
| | $J = 2$ | $k_x^2 + k_y, k_x k_y$ | $k_x, k_x k_y$ | $k_x, k_x k_y$ | $k_x, k_x k_y$ |
| $n = 6$ | $J = 0$ | $(k_x + i k_y)^2, (k_x - i k_y)^2$ | $E_u$ representation | $k_x^3 - 3 k_y k_x^2, 3 k_y^2 k_x - k_x^3$ | $E_g$ representation |
| | $J = 3$ | $(k_x + i k_y)^2, (k_x - i k_y)^2$ | $E_u$ representation | $k_x^3 - 3 k_y k_x^2, 3 k_y^2 k_x - k_x^3$ | $E_g$ representation |

TABLE II. Classification of spin-singlet pairing potentials. Here we consider a spin-singlet two-orbital SC with $\{d_{xz}, d_{yz}\}$-orbitals. Based on the n-fold rotation symmetry $C_n$ about z-axisd and TRS, we have $J = -J \ (\text{mod } n)$, which leads to all the pairing channels with orbital-independent $\Psi_s(k)$ and orbital-dependent $d_o(k)$-vector in Eq. (2) of the main text.

Note: For $J = 0$ pairing subspace of $C_3$, the $\{d_s^1(k), d_s^3(k)\}$ forms a two-dimensional $E_g$ representation, where the basis functions are $(k_x^2 + k_y, k_x k_y)$ and $(k_x, k_x k_y)$. For $J = 0$ pairing subspace of $C_6$, the $\{d_s^1(k), d_s^3(k)\}$ forms a two-dimensional $E_u$ representation, where the basis functions are $(k_x^2, k_y)$. For $J = 3$ pairing subspace of $C_6$, the $\{d_s^1(k), d_s^3(k)\}$ forms a two-dimensional $E_g$ representation, where the basis functions are $(k_x^3 - 3 k_y k_x^2, 3 k_y^2 k_x - k_x^3)$.

Appendix C: The stability of orbital $d_o$-vector under crystal fields

In this section, we firstly solve this problem only for orbital $d_o$-vectors, giving rise to the first-order approximated results. Then, we consider the coupling between orbital-independent pairing ($\Psi_s(k)$-part in Eq. (A1)) and orbital-dependent pairing ($d_o(k)$-part in Eq. (A1)), and study the second-order approximated results.

1. First-order approximated results

In this subsection, we derive Eq. (3) in the main text, to show the possible existence of the orbital $d_o$-vector under crystal fields (orbital hybridizations). The general $k \cdot p$ normal Hamiltonian considered in the main text reads,

$$H_0(k) = \epsilon(k)\tau_0\sigma_0 + \lambda_o(g_o(k) \cdot \tau)\sigma_0,$$

where the electronic basis is made of $\{1, 2\}$-orbitals $\Psi^1_k = (c_{1,1}^\dagger(k), c_{1,1}^\dagger(k), c_{1,2}^\dagger(k), c_{1,2}^\dagger(k)), \epsilon(k) = (k_x^2 + k_y^2)/2m - \mu$ is the band energy measured relative to the chemical potential $\mu, \lambda_o$ represents the orbital hybridization and $g_o(k) = (g_1(k), g_2(k), g_{3}(k)).$ The TRS $\mathcal{T} = i\sigma_2\tau_0K$ requires $g_{1,3}(k) = g_{1,3}(-k)$ and $g_2(k) = -g_2(-k).$ It leads that

$$g_o(k) \cdot \tau = [g_o(-k) \cdot \tau]^*.$$

Besides, we set $\lambda_o > 0$ without loss of generality. The Matsubara Green’s function for electrons is $G_e(k, i\omega_n) = [i\omega_n - H_0(k)]^{-1}$ and that for holes is $G_h(k, i\omega_n) = -G^*_e(k, i\omega_n)$. Here $\beta = 1/k_BT$ and $\omega_n = (2n + 1)\pi/\beta$ with $n$
integer. Therefore,

\[ G_e(k, i\omega_n) = \frac{P_-(k)}{i\omega_n - \epsilon(k) + \lambda_o |g_o(k)|} + \frac{P_+(k)}{i\omega_n - \epsilon(k) - \lambda_o |g_o(k)|} = G_e^-(k, i\omega_n)P_-(k) + G_e^+(k, i\omega_n)P_+(k), \quad (C3) \]

\[ G_h(-k, i\omega_n) = \frac{P_-(k)}{i\omega_n + \epsilon(k) - \lambda_o |g_o(k)|} + \frac{P_+(k)}{i\omega_n + \epsilon(k) + \lambda_o |g_o(k)|} = G_h^-(k, i\omega_n)P_-(k) + G_h^+(k, i\omega_n)P_+(k), \quad (C4) \]

where \( P_\pm(k) = \frac{1}{2} (\pm \hat{g}_o(k) \cdot \tau) \) with \( \hat{g}_o(k) = g_o(k)/|g_o(k)| \). Here \( G_e^+(k, i\omega_n) = \frac{1}{i\omega_n - \epsilon(k) + \lambda_o |g_o(k)|} \) and \( G_h^+(k, i\omega_n) = \frac{1}{i\omega_n + \epsilon(k) + \lambda_o |g_o(k)|} \). We expand the attractive interactions as

\[ V_{s_1, a', s_2, b'}^{\alpha, \beta}(k, k') = -v_0 \sum_{\Gamma, m} [d_{\alpha, m}^{\Gamma}(k) \cdot \tau i\sigma_2]_{s_1 a, s_2 b}[d_{\beta, m}^{\Gamma}(k') \cdot \tau i\sigma_2]_{s_1'a', s_2'b'}, \quad (C5) \]

where \( v_0 > 0 \) and \( \Gamma \) labels the irreducible representation with \( m \)-dimension of crystalline groups. The coupling between orbital-dependent pairings and orbital-independent pairings will be discussed in detail later. The transition temperature \( T_c \) of orbital-dependent pairing channels is calculated by solving the linearized gap equation,

\[ \Delta_{s_1, s_2}(k) = -\frac{1}{\beta} \sum_{\omega_n} \sum_{s_1', a', s_2', b'} V_{s_1', a', s_2', b'}^{\alpha, \beta}(k, k') \times \left[ G_e(k', i\omega_n) \hat{\Delta}(k') G_h(-k', i\omega_n) \right]_{s_1'a', s_2'b'}, \quad (C6) \]

which is reduced to \( v_0 \chi(T) - 1 = 0 \) with the superconductivity susceptibility \( \chi(T) \) defined as,

\[ \chi(T) = \frac{1}{\beta} \sum_{k, \omega_n} \text{Tr} \left[ (d_o(k) \cdot \tau i\sigma_2)^\dagger G_e(k, i\omega_n)(d_o(k) \cdot \tau i\sigma_2)G_h(-k, i\omega_n) \right], \quad (C7) \]

\[ = -\frac{1}{\beta} \sum_{k, \omega_n} \sum_{\alpha, \beta} G_e^\alpha(k, i\omega_n)G_h^\beta(k, i\omega_n) \times \text{Tr} \left[ (d_o(k) \cdot \tau)^\dagger P_\alpha(k)(d_o(k) \cdot \tau)P_\beta(k) \right], \quad (C8) \]

where \( \alpha, \beta \in \{+, -\} \). Firstly, let us calculate the trace part. In the following calculation, we will use

\[ \text{Tr} \left[ (d_o(k) \cdot \tau)^\dagger P_+(k)(d_o(k) \cdot \tau)P_+(k) \right] + \text{Tr} \left[ (d_o(k) \cdot \tau)^\dagger P_-(k)(d_o(k) \cdot \tau)P_-(k) \right] \]

\[ = (d_o^\dagger(k) \cdot d_o(k)) + 2(d_o^\dagger(k) \cdot \hat{g}_o(k)) (d_o(k) \cdot \hat{g}_o(k)) - (d_o^\dagger(k) \cdot d_o(k)) (\hat{g}_o(k) \cdot \hat{g}_o(k)). \quad (C9) \]

And,

\[ \text{Tr} \left[ (d_o(k) \cdot \tau)^\dagger P_+(k)(d_o(k) \cdot \tau)P_-(k) \right] + \text{Tr} \left[ (d_o(k) \cdot \tau)^\dagger P_-(k)(d_o(k) \cdot \tau)P_+(k) \right] \]

\[ = (d_o^\dagger(k) \cdot d_o(k)) - 2(d_o^\dagger(k) \cdot \hat{g}_o(k)) (d_o(k) \cdot \hat{g}_o(k)) + (d_o^\dagger(k) \cdot d_o(k)) (\hat{g}_o(k) \cdot \hat{g}_o(k)). \quad (C10) \]

Therefore, we arrive at

\[ \text{Tr} \left[ (d_o(k) \cdot \tau)^\dagger P_\alpha(k)(d_o(k) \cdot \tau)P_\beta(k) \right] \]

\[ = \frac{1}{2} \left[ (d_o^\dagger(k) \cdot d_o(k)) \right. + i\alpha (d_o(k) \cdot (d_o^\dagger(k) \times \hat{g}_o(k))) \right. + i\beta (d_o^\dagger(k) \cdot (d_o(k) \times \hat{g}_o(k))) + \alpha\beta \left( 2(d_o^\dagger(k) \cdot \hat{g}_o(k)) (d_o(k) \cdot \hat{g}_o(k)) - (d_o^\dagger(k) \cdot d_o(k)) (\hat{g}_o(k) \cdot \hat{g}_o(k)) \right) \quad (C11) \]

Then we have

\[ \chi(T) = -\frac{1}{\beta} \sum_{k, \omega_n} \sum_{\alpha, \beta} G_e^\alpha(k, i\omega_n)G_h^\beta(k, i\omega_n) \left[ (d_o^\dagger(k) \cdot d_o(k)) + i\alpha (d_o(k) \cdot (d_o^\dagger(k) \times \hat{g}_o(k))) + i\beta (d_o^\dagger(k) \cdot (d_o(k) \times \hat{g}_o(k))) \right] \]

\[ + \alpha\beta \left( 2(d_o^\dagger(k) \cdot \hat{g}_o(k)) (d_o(k) \cdot \hat{g}_o(k)) - (d_o^\dagger(k) \cdot d_o(k)) (\hat{g}_o(k) \cdot \hat{g}_o(k)) \right). \quad (C12) \]

Next, we calculate the integration for \( \sum_{k, \omega_n} \) by using,

\[ \sum_{k, \omega_n} \rightarrow \frac{N_0}{4} \int_{-\omega_D}^{+\omega_D} de \int \int d\Omega \sum_{\omega_n}, \quad (C13) \]
where \( N_0 \) is the density of states at Fermi surface and \( \Omega \) is the solid angle of \( \mathbf{k} \) on Fermi surfaces. Then,
\[
- \frac{N_0}{\beta} \int_{-\omega_D}^{+\omega_D} \sum_{\omega_n} d\epsilon G^t_e(\mathbf{k}, i\omega_n) G^+_h(\mathbf{k}, i\omega_n) = - \frac{N_0}{\beta} \int_{-\omega_D}^{+\omega_D} \sum_{\omega_n} G^t_e(\mathbf{k}, i\omega_n) G^+_h(\mathbf{k}, i\omega_n) = \chi_0(T),
\]
where \( \chi_0(T) = N_0 \ln \left( \frac{2\epsilon_c \omega_D}{\pi \hbar^2 T} \right), \gamma = 0.57721 \cdots \) the Euler-Mascheroni constant and \( \omega_D \) the Deybe frequency. And,
\[
- \frac{N_0}{\beta} \int_{-\omega_D}^{+\omega_D} \sum_{\omega_n} d\epsilon G^t_e(\mathbf{k}, i\omega_n) G^+_h(\mathbf{k}, i\omega_n) = - \frac{N_0}{\beta} \int_{-\omega_D}^{+\omega_D} \sum_{\omega_n} G^t_e(\mathbf{k}, i\omega_n) G^+_h(\mathbf{k}, i\omega_n) = \chi_0(T) + N_0 \mathcal{C}_0(T),
\]
where \( \mathcal{C}_0(T) = \text{Re}[\psi^{(0)}(\frac{1}{2}) - \psi^{(0)}(\frac{1}{2} + \frac{\lambda_\omega}{2\pi \hbar^2 T})] \). Here \( \psi^{(0)}(z) \) is the digamma function. Now we calculate \( \chi(T) \),
\[
\chi(T) = \iint_S d\Omega \left\{ \chi_0(T) |\mathbf{d}_o \cdot \mathbf{g}_o|^2 + (\chi_0(T) + N_0 \mathcal{C}_0(T)) \left[ |\mathbf{d}_o|^2 - |\mathbf{d}_o \cdot \mathbf{g}_o|^2 \right] \right\},
\]
\[
= \chi_0(T) + N_0 \iint_S d\Omega \mathcal{C}_0(T) \left( |\mathbf{d}_o|^2 - |\mathbf{d}_o \cdot \mathbf{g}_o|^2 \right).
\]
In the calculation, we use normalized gap functions with \( \iint_S d\Omega |\mathbf{d}_o|^2 = 1 \). It leads to,
\[
\ln \left( \frac{T_c}{T_{ct}} \right) = \iint_S d\Omega \mathcal{C}_0(T) \left( |\mathbf{d}_o|^2 - |\mathbf{d}_o \cdot \mathbf{g}_o|^2 \right),
\]
where \( T_{ct} \) is \( T_c \) for \( \lambda_\omega = 0 \) case by solving \( v_0 \chi_0(T_{ct}) = 1 \). This is the Eq. (5) in the main text. In general, the right-hand side of Eq. (C30) suppresses \( T_c \). It clearly indicates that \( T_c \) would not be suppressed by orbital hybridization once \( \mathbf{d}_o \parallel \mathbf{g}_o \) for all \( \mathbf{k} \). So we conclude that the orbital \( \mathbf{d}_o \)-vector is possible to be stabilized in materials.

### 2. Second-order approximated results

In this subsection, we consider the coupling between orbital-independent pairing (\( \Psi_s(\mathbf{k}) \)-part in Eq. (A1)) and orbital-dependent pairing (\( \mathbf{d}_o(\mathbf{k}) \)-part in Eq. (A1)), and study the second-order approximated results for the above conclusion.

The attractive interaction is now decomposed as
\[
V_{s_1 a, s_2 b, s'_1 a', s'_2 b'}^{s_1 a, s_2 b}(\mathbf{k}, \mathbf{k}') = -v_0 [\mathbf{d}_o(\mathbf{k}) \cdot \mathbf{\tau} i \sigma_2]_{s_1 a, s_2 b} [\mathbf{d}_o(\mathbf{k}') \cdot \mathbf{\tau} i \sigma_2]_{s'_1 a', s'_2 b'} - v_1 [\Psi_s(\mathbf{k}) i \sigma_2]_{s_1, s_2} [\Psi_s(\mathbf{k}') i \sigma_2]_{s'_1, s'_2},
\]
where \( v_0 \) is the interaction strength in the orbital-dependent channel and \( v_1 \) is the interaction strength in the orbital-independent channel. And they belong to the same representation of symmetry groups, leading to the coupled linearized gap equation,
\[
\text{Det} \begin{pmatrix} v_0 \chi(T) - 1 & v_0 \chi_{os}(T) \\ v_1 \chi_{so}(T) & v_1 \chi_{s}(T) - 1 \end{pmatrix} = 0,
\]
where
\[
\chi_{os}(T) = - \frac{1}{\beta} \sum_{\mathbf{k}, \omega_n} \text{Tr} \left[ (\mathbf{d}_o(\mathbf{k}) \cdot \mathbf{\tau} i \sigma_2) G_e(\mathbf{k}, i\omega_n) \Psi_s(\mathbf{k}) i \sigma_2 G_h(-\mathbf{k}, i\omega_n) \right],
\]
\[
\chi_{so}(T) = - \frac{1}{\beta} \sum_{\mathbf{k}, \omega_n} \text{Tr} \left[ \Psi_s(\mathbf{k}) i \sigma_2 G_e(\mathbf{k}, i\omega_n) (\mathbf{d}_o(\mathbf{k}) \cdot \mathbf{\tau} i \sigma_2) G_h(-\mathbf{k}, i\omega_n) \right].
\]
It leads to
\[
(v_0 \chi(T) - 1)(v_1 \chi_{s}(T) - 1) - v_0 v_1 \chi_{so}(T) \chi_{os}(T) = 0,
\]
Considering the \( v_0 > v_1 \) case firstly, then, the bare \( T_c \) of orbital-dependent pairings are larger than that of orbital-independent pairings, we have
\[
v_0 \chi(T) - 1 - \frac{v_0 v_1 \chi_{so}(T) \chi_{os}(T)}{v_1 \chi_{s}(T) - 1} = 0,
\]
from which, we define the total superconductivity susceptibility as,
\[
\chi'(T) = \chi(T) + \frac{\chi_{os}(T)\chi_{os}(T)}{1/v_1 - \chi_s(T)}
\]  
(C24)
where \(\chi(T)\) has been calculated in the above subsection (see Eq. (C17)), and the second part is the second-order correction. After tracing out the spin degrees of freedom, we have \(\chi_{os}(T) = \chi_{os}(T)\). Following the same procedure as in the first-order case, we have

\[
\chi_{os}(T) = -\frac{2N_0}{\beta} \int_{-\omega_D}^{+\omega_D} dx \int_{S} d\Omega \sum_{\omega_n} \sum_{\alpha} \alpha G_n^\alpha(k, i\omega_n)G_n^\alpha(k, i\omega_n)[(d_o(k) \cdot g_o(k))\Psi_s(k)]
\]  
(C25)
which would vanish if \(\lambda_o = 0\), i.e. no orbital hybridization, based on the definitions of \(G_{\alpha/h}^+/-\), which in turn, reproduces the first-order calculation above. At non-zero, but small \(\lambda_o\) (\(\lambda_o k_F^2 < \mu\), \(\chi_{os}(T)\) will also be small but non-zero. For convenience of discussion, we define

\[
\delta(T, \lambda_o) = -\frac{N_0}{\beta} \int_{-\omega_D}^{+\omega_D} dx \int_{S} d\Omega \sum_{\omega_n} \sum_{\alpha} \alpha G_n^\alpha(k, i\omega_n)G_n^\alpha(k, i\omega_n) - G_n^\alpha(k, i\omega_n)G_n^\alpha(k, i\omega_n)[(d_o(k) \cdot g_o(k))\Psi_s(k)],
\]  
(C26)
where \(\delta(T, \lambda_o) \sim \lambda_o k_F^2 / \mu\) would vanish at leading order (see Eq. (C14)). Then we have

\[
\chi_{os}(T) = 2\delta(T, \lambda_o) \int_{S} d\Omega [(d_o(k) \cdot g_o(k))\Psi_s(k)].
\]  
(C27)
With this, the total superconductivity susceptibility in Eq. (C24) becomes

\[
\chi'(T) = \chi(T) + \frac{\chi_{os}(T)}{2N_0 \log(T/T_s)} = \chi(T) + \delta\chi(T).
\]  
(C28)
where \(\delta\chi(T)\) is the second-order correction due to the coupling between orbital-independent pairings (\(\Psi_s(k)\)-part in Eq. (A1)) and orbital-dependent pairing (\(d_o(k)\)-part in Eq. (A1)),

\[
\delta\chi(T) = \frac{2\delta^2(T, \lambda_o)}{N_0 \log(T/T_s)} \left( \int_{S} d\Omega \left| (d_o(k) \cdot g_o(k))\Psi_s(k) \right| \right)^2.
\]  
(C29)
Since we assumed \(\nu_0 > v_1\), i.e. \(T_{ct} > T_s\), then the actual transition temperature would be \(T_c \approx T_{ct} > T_s\), giving \(\log(T_c/T_s) > 0\). As a result, the correction to the susceptibility is positive: \(\delta\chi(T) > 0\). Then following the same procedure as in the previous section, we have

\[
\ln \left( \frac{T_c}{T_{ct}} \right) = \int_{S} d\Omega C_0(T_{ct}) \left( |d_o|^2 - |d_o \cdot g_o|^2 \right) + \delta\chi(T_{ct}) \frac{N_0}{N_0},
\]  
(C30)
where the first part is the first-order result (see Eq. (C30) or Eq. (5) in the main text; order as \(\mathcal{O}(\lambda_o k_F^2 / \mu)\)), and the second part is the second-order result (order as \(\mathcal{O}(\lambda_o k_F^2 / \mu)^2\)). Therefore, we conclude that,

- **The first part**: it determines the direction of orbital \(d_o\)-vector to be \(d_o \parallel g_o\). Because of \(C_0(T_{ct}) \leq 0\), once \(d_o \parallel g_o\) at any momentum \(k\), the first part vanishes.
- **The second part**: it determines the magnitude of orbital \(d_o\)-vector to be \(d_o \propto \Psi_s(k)g_o\). Thus, the second part becomes maximum, leading to the increasing of \(T_{ct}\) maximally.

Here we have used the fact,

\[
\left( \int_{S} d\Omega |\Psi_i(k)\Psi_j(k)| \right)^2 \leq 1, \quad \text{for any scalar } \Psi_i, \quad \int_{S} d\Omega |\Psi_i(k)\Psi_i(k)| = 1.
\]  
(C31)
And \(\left( \int_{S} d\Omega |\Psi_i(k)\Psi_j(k)| \right)^2 \) reaches 1 only when \(i = j\).
Next, we briefly discuss the case where \( v_0 < v_1 \). The same result can be similarly argued. In this case, the dominant pairing channel is the orbital-independent pairing \((T_s > T_d)\), which can induce the orbital \( \mathbf{d}_o \)-vector via their couplings. Similar to Eq. (C23), we define the total superconductivity susceptibility for orbital-independent pairing as

\[
v_1 \chi_s(T) - 1 - \frac{v_0 v_1 \chi_{so}(T) \chi_{as}(T)}{v_0 \chi(T) - 1} = 0, \tag{C32}
\]

which leads to

\[
x'(T) = x(T) + \frac{\chi_{so}(T) \chi_{as}(T)}{1/v_0 - \chi(T)} = N_0 \ln \left( \frac{2 \gamma \omega_0}{2 \pi k_B T} \right) + \frac{\chi_{as}^2(T)}{N_0 \ln(T_c/T_s) - \int_S d\Omega C_0(T_c)} \left( |\mathbf{d}_o|^2 - |\mathbf{d}_o \cdot \mathbf{g}_o|^2 \right), \tag{C33}
\]

thus,

\[
\ln \left( \frac{T_c}{T_s} \right) = 2 \delta^2(T_c, \lambda_o) \times \frac{\int_S d\Omega [ \mathbf{d}_o(k) \cdot \mathbf{g}_o(k) \Psi_s(k)]^2}{N_0 \ln(T_c/T_s) - \int_S d\Omega C_0(T_c)} \left( |\mathbf{d}_o|^2 - |\mathbf{d}_o \cdot \mathbf{g}_o|^2 \right) \geq 0, \tag{C34}
\]

here \( T_c \sim T_s > T_{ct} \) so that \( \ln(T_c/T_s) > 0 \). The correction is in order of \( \mathcal{O}(\lambda_o k_F^2/\mu^2) \). Therefore,

- **The denominator**: it determines the direction of orbital \( \mathbf{d}_o \)-vector to be \( \mathbf{d}_o \parallel \mathbf{g}_o \). Because of \( C_0(T_{ct}) \leq 0 \), then,
  \[ -\int_S d\Omega C_0(T_{ct}) \left( |\mathbf{d}_o|^2 - |\mathbf{d}_o \cdot \mathbf{g}_o|^2 \right) \geq 0, \]
  once \( \mathbf{d}_o \parallel \mathbf{g}_o \) at any momentum \( \mathbf{k} \), the denominator is positive and minimum.

- **The numerator**: it determines the magnitude of orbital \( \mathbf{d}_o \)-vector to be \( \mathbf{d}_o \propto \Psi_s(k) \mathbf{g}_o \). Thus, the numerator becomes maximum, leading to the increasing of \( T_{ct} \) maximally.

Therefore, according to both Eq. (C30) and Eq. (C34), we conclude that the orbital \( \mathbf{d}_o \)-vector that is parallel with orbital hybridization \( \mathbf{g}_o \)-vector could be generally stabilized in real materials. And we find that

\[
\mathbf{d}_o = \pm \Psi_s(k) \mathbf{g}_o, \tag{C35}
\]

which is shown in Eq. (6) in the main text. However, it has also a \( Z_2 \) phase \( \pm \), which can be pinned by taking higher order corrections into account.

**Appendix D: Formation of the pairing near Fermi surface in band picture**

Here, we provide another perspective on the pairing in orbital channel near the Fermi surface (FS) by looking at the total free energy of the system in band picture, where the pairing amplitude is treated perturbatively.

In the presence of orbital hybridization or nematic order, the Hamiltonian without pairing is given by

\[
\mathcal{H}_0 = (\epsilon_k - \mu) \tau_0 \sigma_0 + \lambda (\mathbf{g} \cdot \tau) \sigma_0, \tag{D1}
\]

where \( \lambda \) is taken to be positive. The degeneracy in the orbital channel will be lifted, whereas the spin channel still has the double-degeneracy. Effectively, the vector \( \mathbf{g} \) acts as a “Zeeman field” in the orbital space, and the pseudo-spin will be parallel or anti-parallel to the field for the two splitting levels. And we notice that

\[
[\mathbf{g} \cdot \tau, \mathcal{H}_0] = 0. \tag{D2}
\]

More precisely, the two eigenstates of \( \mathcal{H}_0 \) can be denoted by \( |E_\pm \rangle \equiv |\mathbf{g}; \pm \rangle \), where +/- means parallel/anti-parallel (eigenvalues of the symmetry operator \( \mathbf{g} \cdot \tau \)). Please note that \( \mathbf{g} = \mathbf{g}/|\mathbf{g}| \). And

\[
\mathcal{H}_0|E_\pm \rangle = E_\pm |E_\pm \rangle, \quad \text{with} \quad E_\pm = \epsilon_k - \mu \pm \lambda |\mathbf{g}(\mathbf{k})|. \tag{D3}
\]

with \( \lambda > 0 \). Setting \( E_\pm = 0 \), it gives rise to two FSs (labeled as FS\( \pm \)) with energy splitting as \( 2\lambda |\mathbf{g}(\mathbf{k})| \), which is approximately as \( \sim \lambda k_F^2 \) with respect to \( \epsilon_{k_F} = \mu \).

Now we consider the spin-singlet pairing part in the original basis,

\[
\mathcal{H}_\Delta = \sum_{\mathbf{k}} (c_{1,+}^\dagger(\mathbf{k}), c_{2,+}^\dagger(\mathbf{k})) [\Delta_s \Psi(\mathbf{k}) \tau_0 + \Delta_o (\mathbf{d}_o(\mathbf{k}) \cdot \tau)] (c_{1,+}(-\mathbf{k}), c_{2,+}(-\mathbf{k}))^T + \text{H.c.}. \tag{D4}
\]
The BdG Hamiltonian is then given by
\[ H_{\text{BdG}} = ((\epsilon_k - \mu)\tau_0 + \lambda(\mathbf{g} \cdot \mathbf{\tau})\sigma_0) \gamma_3 + [\Delta_s \Psi(k) + \Delta_o(d_o(k) \cdot \mathbf{\tau})] (i\sigma_2) \gamma_2, \]
where \( \gamma_i \) are the Pauli matrices in particle-hole channel.

Next, we consider weak-coupling limit (infinitesimal pairing strength, namely, \( \Delta_o \to 0 \)) and we use the band picture to study the pairing Hamiltonian. For this purpose, we rewrite \( \mathbf{H} \) as
\[ \mathbf{H}_0 = \sum_{k, s} E_r(k) c^\dagger_{\tau s}(k) c_{\tau s}(k). \]

where \( \tau = \pm \) is the band index and \( s \) is for spin. The unitary transformation matrix \( U(k) \) in the orbital subspace leads to the diagonalization of \( \mathbf{H}_0 \),
\[ U(k) [(\epsilon_k - \mu)\tau_0 + \lambda(\mathbf{g} \cdot \mathbf{\tau})] U(k) = \text{Diag}[E_+(k), E_-(k)], \]
where spin index has been dropped and the 2-by-2 \( U(k) \) can be expressed by the eigen-states of \( \mathbf{H}_0 \),
\[ U(k) = \{|E_+(k)\}, |E_-(k)\}. \]

Thus, \( U(k)^\dagger U(k) = 1 \). And the time-reversal symmetry requires that
\[ U(k) = (U(-k))^*. \]

Acting on the basis, we have
\[ (c^\dagger_{1, s}(k), c^\dagger_{2, s}(k)) = (c^\dagger_{1, s}(k), c^\dagger_{1, s}(k)) U(k) \]
where \( s \) is for spin. We then project the spin-singlet pairing Hamiltonian in Eq. (D4) into the band basis, thus, the spin-singlet pairing Hamiltonian becomes
\[ \mathbf{H}_\Delta = \sum_{k} \{ U(k) [\Delta_s \Psi(k) + \Delta_o(d_o(k) \cdot \mathbf{\tau})] U(k) \} (c^\dagger_{1, \uparrow}(-k), c^\dagger_{1, \downarrow}(-k))^T. \]

Here \( U(k) = (U(-k))^* \) has been used. Therefore, the spin-singlet pairing potential in the band basis becomes
\[ \Delta_{\text{band}}(k) = U(k) [\Delta_s \Psi(k) \tau_0 + \Delta_o(d_o(k) \cdot \mathbf{\tau})] U(k). \]

Thus, the orbital-independent pairings only lead to the intra-band pairings, while the orbital-dependent pairings can give rise to both intra-band and inter-band pairings.

First of all, we focus on the pure orbital \( d_o \)-vector (orbital-dependent pairings) by assuming \( \Delta_s = 0 \). To separate the intra-band pairings from the inter-band pairings, we decompose the orbital \( d_o \)-vector as
\[ d_o(k) = d_{\parallel}(k) \mathbf{g}(k) + d_{\perp}(k), \]
where \( d_{\parallel}(k) = d_o(k) \cdot \mathbf{g}(k) \) and \( d_{\perp}(k) \cdot \mathbf{g}(k) = 0 \). Thus,

- **The parallel component**: the \( d_{\parallel}(k) \mathbf{g}(k) \)-part commutes with \( \mathbf{g} \cdot \mathbf{\tau} \), hence only generates intra-band pairing for the two FSs. Therefore, the projected intra-band pairing Hamiltonian reads,
\[ \mathbf{H}_{\text{intra-band}, \Delta} = \sum_{k, \tau} (\tau \Delta_o d_{\parallel}(k) \mathbf{g}(k)) c_{\tau, \uparrow}(-k) c_{\tau, \downarrow}(-k) \] + H.c.. \]

Here \( \tau = \pm \) indicates that the intra-band pairing strengths on two FSs are opposite. And \( d_{\parallel}(k) = d_{\parallel}(-k) \), the intra-band pairing is a even-parity pairing.

- **The perpendicular component**: the \( d_{\perp}(k) \)-part would mix the two states \( |E_\pm\rangle \), then it only produces inter-band pairings. At a fixed \( k \), we now perform a rotation,
\[ O(3) \text{ rotation: } R_o \mathbf{g}_o R_o^\dagger = (0,0,g'_x) \text{ and } R_o d_o R_o^\dagger = (g'_x,g'_y,0) \]
at the same time, we perform a rotation in the orbital subspace

$$ SU(2) \text{ rotation: } R_\tau \tau_{x,y,z} R_\tau^\dagger = \tau'_{x,y,z} $$

Due to this rotation, the perpendicular components only couple $|E_+\rangle$ with $|E_-\rangle$. This proves that

$$ \langle E_-| \mathbf{d}_\parallel \cdot \tau | E_+ \rangle = 0. $$

(D17)

Here $|E_\tau\rangle$ are eigen-states of $\hat{g}_o \cdot \tau$. Thus, the projected inter-band pairing Hamiltonian reads,

$$ H_{\text{inter-band,} \Delta} = \sum_{\mathbf{k}, \tau} \Delta_{\tau, -\tau}(\mathbf{k}) [c_{\tau, \uparrow}(\mathbf{k})c_{-\tau, \downarrow}(-\mathbf{k}) - c_{-\tau, \downarrow}(\mathbf{k})c_{\tau, \uparrow}(-\mathbf{k})] + \text{H.c.} $$

(D18)

where $\Delta_{\tau, -\tau}(\mathbf{k}) = \Delta_o \langle E_\tau(\mathbf{k})| \mathbf{d}_\parallel(\mathbf{k}) \cdot \tau | E_-\tau(\mathbf{k}) \rangle$. In the limit $\lambda k_F^2 \gg \Delta_o$ (i.e., band splitting is much larger than the pairing gap), the inter-band pairing is not energetically favorable in the weak-coupling pairing theory (i.e., attractive interaction is infinitesimal small). It means the inter-band pairing will be severely suppressed if we increase the orbital hybridization $\lambda$, consistent with the calculation in the main text (see Fig. (2)).

From the above analysis, we conclude that the orbital $\mathbf{d}_o$-vector should be parallel with the orbital hybridization $\mathbf{g}$.

To determine the magnitude of the orbital $\mathbf{d}_o$-vector, we turn on the orbital-independent pairing $\Delta_s \neq 0$. We also assume both pairing channels are small. Assuming the two FSs have DOS $N_{\pm}$ near the FS (ignoring the momentum-dependence if the FS is almost isotropic), then the total condensation energy per volume and per spin of the two intra-band pairings is given by

$$ \delta E = N_+ \sum_{\mathbf{k} \in \text{FS}_+} (\Delta_s \Psi_s(\mathbf{k}) + \Delta_o d_{\parallel}(\mathbf{k}))^2 + N_- \sum_{\mathbf{k} \in \text{FS}_-} (\Delta_s \Psi_s(\mathbf{k}) - \Delta_o d_{\parallel}(\mathbf{k}))^2, $$

with the approximation $\lambda k_F^2 \ll \mu$, $\delta E$ becomes

$$ \delta E = (N_+ + N_-) \sum_{\mathbf{k} \in \text{FS}} [\Delta_s^2 (\Psi_s(\mathbf{k}))^2 + \Delta_o^2 (d_{\parallel}(\mathbf{k}))^2] + 2(N_+ - N_-) \Delta_s \Delta_o \sum_{\mathbf{k} \in \text{FS}} (\Psi_s(\mathbf{k})d_{\parallel}(\mathbf{k})), $$

(D20)

where FS is for $\lambda \to 0$. And we see that in order to maximize the condensation energy, we require

$$ d_{\parallel}(\mathbf{k}) \propto \Psi_s(\mathbf{k}), \text{ if sign}[(N_+ - N_-)\Delta_s\Delta_o] = 1, $$

(D21)

$$ d_{\parallel}(\mathbf{k}) \propto -\Psi_s(\mathbf{k}), \text{ if sign}[(N_+ - N_-)\Delta_s\Delta_o] = -1, $$

(D22)

according to Eq. (C31). The results from the weak-coupling limit are the same as the calculations from linearized gap equations.

Appendix E: The coexistence of orbital $\mathbf{d}_o$-vector and nematic order

In this section, we discuss the interaction effects on the stability of orbital $\mathbf{d}_o$-vector for a two-band superconductor. A general density-density interaction, including both inter-band and intra-band terms, is,

$$ H_{\text{int}} = v_1(\hat{n}_{\uparrow\uparrow} + \hat{n}_{\downarrow\downarrow})(\hat{n}_{\uparrow\downarrow} + \hat{n}_{\downarrow\uparrow}) + v_2(\hat{n}_{\uparrow\uparrow}\hat{n}_{\downarrow\downarrow} + \hat{n}_{\uparrow\downarrow}\hat{n}_{\downarrow\uparrow}), $$

(E1)

where $\hat{n}$ is the electron density operator and $v_{1,2}$ are interaction strengths. With the following mean-field decomposition, we can define the nematic ordering that spontaneously breaks the rotational symmetry.

$$ H_{MF} = \hat{n}_{\uparrow\uparrow} \left( \frac{v_1}{2} \langle \hat{n}_2 \rangle + v_2 \langle \hat{n}_{\downarrow\downarrow} \rangle \right) + \hat{n}_{\downarrow\downarrow} \left( \frac{v_1}{2} \langle \hat{n}_2 \rangle + v_2 \langle \hat{n}_{\uparrow\uparrow} \rangle \right) + \hat{n}_{\uparrow\downarrow} \left( \frac{v_1}{2} \langle \hat{n}_1 \rangle + v_2 \langle \hat{n}_{\downarrow\uparrow} \rangle \right) + \hat{n}_{\downarrow\uparrow} \left( \frac{v_1}{2} \langle \hat{n}_1 \rangle + v_2 \langle \hat{n}_{\uparrow\downarrow} \rangle \right). $$

(E2)

For the purpose of our discussion, we assume there is no spin ferromagnetism, i.e. $\langle \hat{n}_{\alpha\uparrow} \rangle = \langle \hat{n}_{\alpha\downarrow} \rangle = \frac{1}{2} \langle \hat{n}_\alpha \rangle$, then the mean field Hamiltonian simplifies to

$$ H_{MF} = \hat{n}_1 \left( \frac{v_1}{2} \langle \hat{n}_2 \rangle + \frac{v_2}{2} \langle \hat{n}_1 \rangle \right) + \hat{n}_2 \left( \frac{v_1}{2} \langle \hat{n}_1 \rangle + \frac{v_2}{2} \langle \hat{n}_2 \rangle \right) \equiv \hat{n}_1 \Phi_1 + \hat{n}_2 \Phi_2. $$

(E3)
Then the nematic order parameter can be defined as

$$
\Phi \equiv \Phi_1 - \Phi_2 = \frac{1}{2} (v_2 - v_1) \left( \langle \hat{n}_1 \rangle - \langle \hat{n}_2 \rangle \right).
$$

Now the stage is set to define the total orbital hybridization as,

$$
g_{\text{tot}} = g_o + t_{\text{nem}} g_{\text{nem}}
$$

where $g_{\text{nem}} = (0, 0, \Phi)$ and $t_{\text{nem}} = \lambda_{\text{nem}}/\lambda_o$. Then, we apply Eq. (C30) to study the stability of orbital $d_o$-vector when nematic order develops above superconducting $T_c$. The results are summarized in Fig. 5.

**FIG. 5.** The coexistence of orbital $d_o$-vector and nematic order. In (a), each $t_{\text{nem}}$ corresponds to a particular form of the $d$-vector, which determines $T_c$ based on Eq. (C30). The three curves correspond to three different values for the nematic order $\Phi$. The $T_c$ is not suppressed by the nematic order as long as $d_o \parallel g_{\text{tot}}$, i.e. $t_{\text{nem}} = 1$. The $t_{\text{nem}} = 1$ case is further illustrated in (b), where it is shown that the magnitude of the nematic order does not change $T_c$ (up to the order of approximation made in Eq. (C30)). (c) shows non-zero nematic order breaks the original $C_4$ (red line) down to $C_2$ (blue line). Here $g_o = (k_x^2 - k_y^2, 0, 3k_x k_y)$.

**Appendix F: Application to single-layer graphene superconductor**

Based on the discussion in the main text, the nematic $d$-vector is characterized by $g_{\text{tot}} = (g_{\text{int,1}}, 0, 0)$, with

$$
g_{\text{int,1}} = 1 + 2t_1 k_x k_y + t_2 (k_x^2 - k_y^2).
$$

A closed Fermi surface (FS) can be parametrized by $k_F(\theta)$. By using Eq. (C30), the nematic $d_o$-vector represents an $(s+d)$-wave pairing states,

$$
\begin{cases}
  \text{s-wave dominant: } t_{1,2} \ll 1, \text{ fully gapped superconductors} \\
  \text{d-wave dominant: } t_{1,2} \gg 1, \text{ nodeless superconductors}
\end{cases}
$$

As a result, we have $|d_o| \sim |g_{\text{tot}}| = |1 + k_F(\theta) (t_1 \sin 2\theta + t_2 \cos 2\theta)|$. We see that the SC gap function can have nodes as long as $\sqrt{t_1^2 + t_2^2}$ is large enough. For graphene, the FS has $C_3$ symmetry, i.e. $k_F(\theta)$ has periodicity of $\frac{2}{3}\pi$, whereas $\sin 2\theta$ and $\cos 2\theta$ have periodicity of $\pi$, giving a periodicity of $2\pi$ to $|d_o|$, which completely breaks the $C_3$ symmetry of the system. Fig. 6 shows the $C_3$-breaking nematic orders from inter-valley scattering, one with nodal gap function, the other with nodeless gap function.
FIG. 6. The $C_4$-breaking nematic order from inter-valley scattering in graphene. $t_1 = 0, t_2 = 1.2$ for the nodal case (red) and $t_1 = 0, t_2 = 0.5$ for the fully gapped case (blue).

Appendix G: Spin and orbital magnetizations: $M_s$ and $M_o$

In this section, we show the definition of spin and orbital magnetizations at the mean-field level. The spin magnetization in orbital-inactive systems takes the form

$$M_s \propto \sum_{\mathbf{k}, s_1, s_2} \langle c_{s_1}^\dagger(\mathbf{k}) \sigma_{s_1 s_2} c_{s_2}(\mathbf{k}) \rangle,$$

which tells us the magnetic moments generated by spin polarizations. Similarly, the orbital magnetization in orbital-active system is given by

$$M_o \propto \sum_{\mathbf{k}, s, a, b} \langle c_{s,a}^\dagger(\mathbf{k}) \tau_{ab} c_{s,b}(\mathbf{k}) \rangle.$$

The different components of the orbital magnetization vector represents different orders in the SC ground state. More specifically, we have

$$M_o^x = \sum_{\mathbf{k}, s} \langle c_{s,d_x}^\dagger c_{s,d_y} + c_{s,d_y}^\dagger c_{s,d_x} \rangle,$$

$$M_o^y = -i \sum_{\mathbf{k}, s} \langle c_{s,d_x}^\dagger c_{s,d_y} - c_{s,d_y}^\dagger c_{s,d_x} \rangle = \frac{1}{2} \sum_{\mathbf{k}, s} \langle \hat{n}_{s,d_x} + i \hat{n}_{s,d_y} - \hat{n}_{s,d_x} - i \hat{n}_{s,d_y} \rangle,$$

$$M_o^z = \sum_{\mathbf{k}, s} \langle c_{s,d_x}^\dagger c_{s,d_z} - c_{s,d_z}^\dagger c_{s,d_x} \rangle.$$

We see that $M_o^{x,z}$ breaks the $C_4$ rotation symmetry and $M_o^y$ breaks TRS. In our work, we only consider the possibility of spontaneous TRS breaking, thus the $M_o^{x,z}$ will not couple to the superconducting order parameters, which are required to be invariant under $C_n$. Because $M_o^y$ breaks TRS so that it could be coupled to the superconducting order parameters, which spontaneously breaks TRS. This is one of the main results in our work,

$$(0, M_o^y, 0) \propto \mathbf{i} \mathbf{d}_o \times \mathbf{d}_o,$$

where the complex orbital $\mathbf{d}_o$-vector breaks TRS.