Chiral Induced Spin Selectivity as a Spontaneous Intertwined Order

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Chiral induced spin selectivity (CISS) describes efficient spin filtering by chiral molecules. This phenomenon has led to nanoscale manipulation of quantum spins with promising applications to spintronics and quantum computing, since its discovery nearly two decades ago. However, its underlying mechanism still remains mysterious for the required spin-orbit interaction (SOI) strength is unexpectedly large. Here we report a multi-orbital theory for CISS, where an effective SOI emerges from spontaneous formation of electron-hole pairing caused by many-body correlation. This mechanism produces a strong SOI to the order of tens of melectronvolts which could support the large spin polarization observed in CISS at room temperature. One central ingredient of our theory is the Wannier functions of the valence and conduction bands correspond respectively to one- and two-dimensional representation of the spatial rotation symmetry around the molecule elongation direction. The induced SOI strength is found to decrease when the band gap increases. Our theory may provide important guidance for searching other molecules with CISS effects.

Introduction.— Atomic spin-orbit coupling is a relativistic quantum effect that originates from the fundamental quantum electrodynamics of electrons orbiting around the nucleus. It is established that heavier atoms tend to have stronger spin-orbit couplings as the effective coupling strength increases with the atomic number \( Z \) as \( Z^2 \) \([1, 2]\). Consequently, material research aiming for strong spin-orbit effects has been mainly focusing on materials composed of heavy atoms \([2]\).

However, a surprisingly large spin-orbit effect has been found in chiral organic and biological molecules mainly composed of carbon atoms in the study of chiral induced spin selectivity \([3]\), which has received enormous research efforts in the last decade \([4\text{–}18]\). This fascinating phenomenon has been observed in a broad range of chiral molecules, from DNA \([3, 6]\) and protein \([8]\), to \(\alpha\)-helical peptides \([11, 14]\) and helicene \([12, 16]\), using a number of different experimental setups such as photoelectron transmission \([3, 6]\), transport \([7, 12, 14]\), and electrochemistry measurements \([19]\). It has far reaching implications in the fundamental understanding of important biological processes such as protein-folding and biorecognition \([4]\). The experimental observations imply a generic underlying mechanism of spin filtering by chiral molecules, that demands a theoretical explanation. Previous theoretical studies have shown that the experimental features can be reproduced by proper tight-binding models with a strong SOI \([20\text{–}22]\), although the intrinsic spin-orbit interaction in these molecules mainly containing carbon atoms are too weak to accommodate the observed large spin polarization, for example up to 60% using chiral peptide molecules \([14]\). The essential question that remains outstanding is how the unexpectedly strong SOI emerges \([23]\) beyond the conventional consideration of the quantum electrodynamics.

In the study of atomic Bose-Einstein condensates in optical lattices, a quantum fluctuation enabled spin-
molecule intertwined order has been proposed in theory [24] to explain the observed spin-momentum locking in a hexagonal lattice without bare SOI [25]. This mechanism has been further generalized to a multi-orbital Bose-Einstein condensate, where a spontaneous spin angular-momentum intertwined order is shown to occur in a metastable state for spinor bosons residing on excited bands of a square lattice [26]. These theoretical studies are inspiring, suggesting that spin-orbit coupled effects could emerge from many-body correlation even in complete absence of single-particle SOI, although the bosonic theories do not apply to the description of electrons bearing CISS in chiral molecules.

In this work, we assume a rotation symmetry around the molecule elongation direction (to be refereed as $z$ below), and consider a setup where electronic valence and conduction bands correspond to a one- and two-dimensional representation of the symmetry group, respectively. The Wannier functions then have $s$ and $p$-orbital character (Fig. 1). Through field theory analysis and renormalization group calculation, we show the electron correlation causes a strong instability towards forming electron-hole pairs that spontaneously break the spin SU(2) and reflection symmetries with preservation of time-reversal. This spontaneous spin-orbit intertwined order gives rise to a strong SOI having a first-quantization form

$$\lambda_{so} \hat{\sigma}_z \otimes \hat{L}_z / 2, \quad (1)$$

with $\hat{\sigma}_z$ the spin Pauli-$z$ operator and $\hat{L}_z$ the angular momentum operator for $p$-orbitals in the conduction band. Considering a band gap $\Delta$, $s$- and $p$-orbital tunnelings, $t_s$ and $t_p$, and their interactions including density-density interaction $U$, Hund’s rule coupling $J$, and Josephson coupling $J'$ (see Methods), the induced SOI strength is given as

$$\lambda_{so} = \Delta / 2 - \sqrt{\Delta^2 / 4 + (U - J - J')^2} |\phi|^2 / 2, \quad (2)$$

with $|\phi|$ the amplitude of the spin-orbit intertwined order parameter estimated to be $|\phi| = 0.25 \times (U - J - J')/(t_s + t_p)$ in the small band gap limit. Taking an example of $t_s + t_p = 1 \text{ eV}$, $U - J - J' = 0.5 \text{ eV}$ and $\Delta = 0$, the induced SOI reaches 50 meV, which suffices for modeling the unexpectedly large CISS effects in chiral molecules [23]. The strength of the induced SOI decreases when the band gap is increased. We expect this result would strongly contribute to the understanding of spin-selective processes in biology [4].

Our theory starts from a field theory description of the three-orbital system (Fig. 1),

$$\hat{H}_0 = \int dz \sum_{\nu, \alpha} P_\nu \psi_{\nu \alpha} (z) \left[ \frac{\hbar^2 \partial_z^2}{2m_\nu} - \Delta \right] \psi_{\nu \alpha} (z). \quad (3)$$

Here $\nu = s, p_x$, or $p_y$ index the orbitals, and $\alpha$ the spin degrees of freedom; $P_\nu$ represents the parity, i.e., $+$ and $-$ for $s$- and $p$-orbitals, respectively; $m_\nu$ and $m_{p_x} = m_{p_y} = m_p$ the effective mass associated with the motional dynamics of the valence and conduction bands along the molecular elongation direction. The field operators $\psi_{\nu \sigma}$ incorporates the low energy degrees of freedom of electrons moving in a molecule near the band edge. Taking a carbon atomic chain as one example, our theory corresponds to the Fermi energy lying in between the $sp$-hybridized $\sigma^*$-bond and $\pi$-bond. We remark that the $s$-orbital in our model may represent a $p_z$ orbital in the molecule as well, which obeys the same symmetry under the spatial rotation around the $z$ direction.

An immediate consequence of the field theory is that it develops strong susceptibility towards electron-hole pairing at low temperature even without interaction. This is described by a response function, $\chi_{sp}^0 = \partial_h \langle \psi_{\nu \alpha}^\dagger \psi_{\nu \alpha'} \rangle = \sqrt{(m_{s-1}^0 + m_{p-1}^0)/2\Delta}$, considering a fictitious infinitesimal perturbation $\Delta H = -h \int dz \left[ \hat{\psi}_{\nu \alpha}^\dagger \hat{\psi}_{\nu \alpha'} + \text{H.c.} \right]$. This response has a divergent behavior $1/\sqrt{\Delta}$ for a small band gap, which is caused by the interplay of the logarithmic divergence of the Feynman diagram (Fig. 1) and the van Hove singularity in one dimensional density of states.

The divergence in the above response function indicates important many-body effects in the system. Having spin SU(2) and spatial rotation symmetries, the three interaction terms including density-density interaction $U$, Hund’s rule coupling $J$, and Josephson coupling $J'$ (see Methods) are the only allowed local interactions between the $s$- and $p$-orbitals. Due to the multi-orbital complexity of our model, there are 21 independent channels in the particle-hole pairing function, $G_{\nu \alpha, \nu' \alpha'} = \langle \psi_{\nu \alpha}^\dagger \psi_{\nu' \alpha'} \rangle$. According to symmetry properties, we group all the particle-hole pairings into the following channels,

$$\hat{\mathcal{G}}_{j^s, m_s, j^p, m_p} = (-)^{q+1} \sum_{\alpha \alpha'} i^{2 \alpha} C^1_{\alpha \alpha'} \left( -\alpha, \alpha' \right)_{j^s, m_s} \psi_{\alpha a} \psi_{q+m_p \alpha'} \quad (4)$$

where the $C^1_{\alpha \alpha'}$ matrix contains Clebsch-Gordon coefficients, and $\psi_{\alpha a}$ is a field operator in angular momentum basis defined by $|\psi_{\alpha a} \rangle \equiv \frac{1}{\sqrt{2}} (\psi_+ \pm i \psi_-) / \sqrt{2}$, $\psi_0 = \psi_\alpha$. The symmetry properties of these operators are listed in Table 1.
TABLE I. Symmetry properties of electron-hole pairings. We have introduced quantum numbers $j_s$ and $m_t$ according to the spin SU(2) symmetry, with $j_s$ equal to 0 and 1 labeling singlet and triplets. Under a spatial rotation around the $z$ direction by an angle $\theta$, the operators transform as $B_{j_s,m_t,q} \rightarrow B_{j_s,m_t,q} e^{im_{t} \theta}$, determined by the quantum number $m_t$. Under time-reversal symmetry (TRS) transformation, we have $B_{j_s,m_t,q} \rightarrow (-1)^{l_{s}/2} B_{j_s,m_t,q}$. The even/odd sign of TRS listed here is determined for an operator $\hat{\theta}$ according to whether its corresponding Hermitian observable $\hat{\theta}^{\dagger} + \hat{\theta}$ (with $\hat{\theta}$ an arbitrary complex number) is TRS even or odd. The operators with $m_t = 0, -1, -2$ are not listed here due to the constraint that $B_{j_s,m_t,q}^{\dagger} = (-1)^{m_{t}+m_{s}} B_{j_s,-m_t,-m_s,q_{t}+m_{t}}$.

In order to generate effective SOI with no spin polarization, we shall consider a spin-orbit intertwined order that breaks spin SU(2) symmetry and preserves time-reversal symmetry. From Table I, the potential candidates are,

$$\hat{\theta}_{m_t} = \frac{1}{\sqrt{2}} \big[ B_{1,m_t,-1;0} - B_{1,m_t,1;1} \big],$$

and

$$\hat{\theta}_{m_s}^l = \frac{1}{\sqrt{2}} \big[ B_{1,m_s,1;0} - B_{1,m_s,0;1} \big],$$

whose parities are odd and even, respectively. They both satisfy the symmetry requirement, but the parity even operators represent pairings within $s$ or $p$-bands, which do not benefit from the divergent susceptibility in our model. We thus focus on analyzing the parity odd spin-orbit intertwined operators $\hat{\theta}_{m_t}$. We emphasize here that this symmetry channel would not be possible in a single-orbital model, where a local triplet order necessarily breaks time-reversal symmetry.

Considering a weak Josephson coupling $J'$, the susceptibility towards forming an order of $\hat{\theta}_{m_t}$ is,

$$\chi_{SO} = \frac{\chi_{0}^{0}}{1 - (U - J)\chi_{0}^{0}}$$

under random phase approximation (see Methods). The divergence in the susceptibility for the non-interacting model at zero temperature persists to finite band gap and finite temperature, provided that $U > J$. This requirement is satisfied considering a typical situation for electrons in a molecule, that the density-density interaction is repulsive and the Hund’s rule coupling is ferromagnetic. Without Josephson coupling, i.e., $J' = 0$, a degenerate channel $\chi_{m_t}^{0} \equiv \chi_{0}^{0} [B_{1,m_t,-1;0} + B_{1,m_t,1;1}] / \sqrt{2}$ which would break time reversal symmetry yields a susceptibility of an identical strength. A negative Josephson coupling $J' < 0$ would break this degeneracy, and make the time reversal symmetric pairing more favorable. Even without Josephson coupling, the presence of orbital motion induced Zeeman splitting also selects the time reversal symmetric pairing over the asymmetric case.

Since all parity odd pairing channels in our theory potentially have a large susceptibility due to the divergence in $\chi_{m_t}^{0}$, we further go beyond the random phase approximation, and carry out a systematic submission of Feynman diagrams using a scheme of renormalization group flow equation, which incorporates the intertwined scatterings among different channels [27]. The results for the susceptibility of forming the spin-orbit intertwined order in Eq. 6 are shown in Fig. 2. It is confirmed that for $J < 0$ and $J' < 0$ the susceptibility for $\hat{\theta}_{m_t}$ diverges at finite temperature (Fig. 2a). The divergence requires the interaction energy to conquer the band gap barrier of the $sp$-orbital pairing. We also find that having a weak antiferromagnetic coupling in $J$ still supports divergent susceptibility in $\hat{\theta}_{m_t}$ channel (Fig. 2b). For a large enough positive $J$, this susceptibility is no longer divergent, but instead meets a strong suppression at low temperature (Fig. 2c), which is due to a divergent susceptibility in a different channel, $\delta_{0,0,1,q}^{0}$ (Supplementary Informa-
A sign change in the Josephson coupling also leads to a low-temperature suppression of $\hat{\Theta}_{m_{s}}$ pairing (Fig. 2 d), owing to a divergent susceptibility in the time-reversal odd channel of $[\hat{\Theta}_{1,m_{s},-1,0} + \hat{\Theta}_{1,m_{s},1,1}] / \sqrt{2}$ (Supplementary Information).

A divergence in the susceptibility $\chi_{SO}$ implies the correlation length of $(\hat{\Theta}_{m_{s}}^{+}(z) \hat{\Theta}_{m_{s}}^{-}(z'))$ reaches the size of the molecule. Taking mean field approximation, the operator $\hat{\Theta}_{m_{s}}$ acquires a finite expectation value, $\langle \hat{\Theta}_{m_{s}} \rangle \equiv \Phi_{m_{s}}$. The free energy of the order parameter takes an SU(3) symmetric form as shown in Methods. The pairings with different $m_{s}$ quantum numbers $0, \pm 1$ are degenerate, although they lead to two distinctive many-body states, analogous to the spin-1 polar and ferromagnetic superfluids in spinor condensate [28] or liquid Helium [29].

Further considering a circular motion induced Zeeman splitting for electrons [22], we have a perturbative coupling $\Delta H = \delta \int dz \left[ i\Psi_{\downarrow}^{\dagger} \hat{\Psi}_{\uparrow} - i\Psi_{\uparrow}^{\dagger} \hat{\Psi}_{\downarrow} + H.c. \right]$. Despite its insufficient strength to model CISS [22, 23], this term determines a spin quantization axis, and triggers an order $(\Phi_{\uparrow,1} \equiv \phi, \Phi_{0} = 0, \Phi_{\downarrow,1} = 0)$ to minimize to total free energy in our theory, from which a strong SOI emerges from electron correlation,

$$H_{SOI} = \frac{U - J^{'}}{4\sqrt{2}} \int dz \left[ i\phi \Psi^{T}(z) \sigma_{z} \otimes L_{-} \Psi(z) + H.c. \right]$$

with $\Psi \equiv [\Psi_{1,\uparrow}, \Psi_{0,\uparrow}, \Psi_{-1,\uparrow}, \Psi_{1,\downarrow}, \Psi_{0,\downarrow}, \Psi_{-1,\downarrow}]^{T}$, $\sigma_{z}$ and $L_{-}$ the standard spin-1/2 and spin-1 angular momentum matrices (see Methods).

This correlation induced coupling breaks spin SU(2) and reflection symmetries with the time reversal symmetry unbroken. Through a unitary transformation into the quasi-particle basis, the induced coupling in the conduction band takes a more standard SOI form given in Eq. (2).

We remark here that Mermin-Wegner theorem does not forbid the long-range order formation in our setup as the continuous symmetries of spatial rotation and the spin SU(2) are all weakly broken considering the real geometry of a chiral molecule and the circular motion induced Zeeman splitting.

**Discussion.**— We have developed a novel quantum mechanism for strong SOI to emerge from many-body correlation effect. This provides an alternative origin for SOI, other than the fundamental quantum electrodynamics, which is particularly crucial to the understanding of the large CISS observed in chiral organic molecules, where the bare SOI is too weak for modeling the experimental observation. Our theory may provide important guidance for future searching of other chiral molecules with CISS, and potentially contributes to the fundamental understanding of spin-selective biological processes.

Since SOI plays an important role in topological physics in general, we expect the mechanism of corre-
loration induced SOI may also shed light on engineering of topological devices such as Majorana quantum computing qubits and also neutral-atom based quantum simulations of topological physics where the bare SOI is absent.

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Methods

Tight binding model. The tight binding Hamiltonian corresponding to the field theory in Eq. (3) is

\[ H_0 = \frac{\Delta}{2} \sum_{j\alpha} \left[ c_{p_j,\alpha,j}^\dagger c_{p_j,\alpha,j} + c_{p_j,\alpha,j}^\dagger c_{p_j,\alpha,j} - c_{s\alpha,j}^\dagger c_{s\alpha,j} \right] 
+ t_\parallel \sum_{j\alpha} \left[ c_{s\alpha,j}^\dagger c_{s\alpha,j+1} + c_{s\alpha,j}^\dagger c_{s\alpha,j-1} - 2c_{s\alpha,j}^\dagger c_{s\alpha,j} \right] 
- t_p \sum_{j\alpha,\nu=\nu_1,\nu_2} \left[ c_{\nu\alpha,j}^\dagger c_{\nu\alpha,j+1} + c_{\nu\alpha,j}^\dagger c_{\nu\alpha,j-1} - 2c_{\nu\alpha,j}^\dagger c_{\nu\alpha,j} \right], \]  

(9)

with \( j \) the site index of the tight binding model, \( t_\parallel \) and \( t_p \) the nearest neighboring tunneling of the \( s \)- and \( p \)-orbitals, and \( c_{\nu\alpha} \) the lattice annihilation operator associated with \( \nu_\alpha \). The energy dispersion of the tight binding model is \( \epsilon_{\nu}(k) = 2t_\nu (1 - \cos k) P_\nu \). The tunneling parameters relate to the effective mass in the field theory as \( m_{\nu}^{-1} = 2t_\nu / \hbar^2 \), taking the lattice constant as a length unit.

Taking spin SU(2) and rotation symmetries, the local electron interaction between \( s \)- and \( p \)-orbitals involves a density-density term,

\[ U \sum_j C_{\nu,j}^\dagger C_{\nu,j} C_{s,j} C_{s,j} \]

the Hund’s rule coupling,

\[ J \sum_{\nu=\nu_1,\nu_2} C_{\nu,j}^\dagger \sigma \nu \sigma C_{s,j}^\dagger \sigma C_{s,j} \]

and a Josephson coupling,

\[ J' \sum_j \left( \left( c_{\nu\alpha,j}^\dagger c_{s\alpha,j}^\dagger + c_{\nu\alpha,j}^\dagger c_{s\alpha,j} \right) c_{s\alpha,j} c_{s\alpha,j} + H.c. \right), \]

where we have introduced compact notation, \( C_{\nu,j} \equiv [c_{\nu\alpha,j}^\dagger, c_{\nu\alpha,j}]^T \). The intra-orbital interaction within the valence band or the conduction band is not included here because only the inter-band interactions contribute strongly to the divergent susceptibility considering the divergence in \( \chi_{0}^{\parallel} \).

Renormalization group flow. In the calculation of susceptibility, we use the scheme of renormalization group flow to carry out a systematic resubmission of higher order Feynman diagrams, in order to capture the complex intertwined scatterings among different channels in our theory. Having SU(2) symmetry, the one particle irreducible (1PI) four point function

\[ \Gamma_{\nu_1,\alpha_1,\nu_2,\alpha_2,\nu_3,\alpha_3,\nu_4,\alpha_4} = \langle \psi_{\nu_1,\alpha_1} \psi_{\nu_2,\alpha_2} \phi_{\nu_3,\alpha_3} \phi_{\nu_4,\alpha_4} \rangle_{1PI}, \]  

(10)

takes a restricted form

\[ \Gamma_{\nu_1,\alpha_1,\nu_2,\alpha_2,\nu_3,\alpha_3,\nu_4,\alpha_4} = V_{\nu_1,\nu_2,\nu_3,\nu_4} \delta_{\alpha_1,\alpha_2} \delta_{\alpha_3,\alpha_4} - V_{\nu_1,\nu_3,\nu_2,\nu_4} \delta_{\alpha_1,\alpha_3} \delta_{\alpha_2,\alpha_4} \]  

(11)

This function takes real values according to the time-reversal symmetry. Further considering rotation symmetry, the nonzero ones are \( V_{\nu_1,\nu_2} = V_{\nu_2,\nu_1} \), \( V_{\nu_3,\nu_4} = V_{\nu_4,\nu_3} \), \( V_{\nu_1,\nu_2} = V_{\nu_2,\nu_1} \), \( V_{\nu_3,\nu_4} = V_{\nu_4,\nu_3} \), \( V_{\nu_1,\nu_2} = V_{\nu_2,\nu_1} \), \( V_{\nu_3,\nu_4} = V_{\nu_4,\nu_3} \). These four point functions are obtained by solving a renormalization group flow equation [27],

\[ \partial_{\nu} V_{\nu_1,\nu_2,\nu_3,\nu_4} = \sum_{\nu'\nu''} \left\{ -\Pi_{\nu'\nu''}^{pp}, V_{\nu_1,\nu_2,\nu_3,\nu_4} V_{\nu_1,\nu_2,\nu_3,\nu_4} + 2\Pi_{\nu''}^{ph}, V_{\nu_1,\nu_2,\nu_3,\nu_4} V_{\nu_1,\nu_2,\nu_3,\nu_4} 
- \Pi_{\nu''}^{ph}, V_{\nu_1,\nu_2,\nu_3,\nu_4} V_{\nu_1,\nu_2,\nu_3,\nu_4} + V_{\nu_1,\nu_2,\nu_3,\nu_4} V_{\nu_1,\nu_2,\nu_3,\nu_4} \right\} \]  

(12)
Here \( l \) is a parameter running from 0 from \(+\infty\), and \( \Pi \) represents derivatives \( \Lambda \partial_\Lambda \) of the functions,

\[
\Pi^p_{vv'}(\Lambda) = \int \frac{dk}{2\pi} \frac{n_f(\epsilon_v(k)) - n_f(\epsilon_{v'}(k))}{\epsilon_v(k) + \epsilon_{v'}(k)} \left[ \Theta_c(\epsilon_v(k))\Theta_c(\epsilon_{v'}(k)) \right]
\]

\[
\Pi^{ph}_{vv'}(\Lambda) = \int \frac{dk}{2\pi} \frac{n_f(\epsilon_v(k)) - n_f(\epsilon_{v'}(k))}{\epsilon_v(k) - \epsilon_{v'}(k)} \left[ \Theta_c(\epsilon_v(k))\Theta_c(\epsilon_{v'}(k)) \right]
\]

where \( n_f(\epsilon) \) is the Fermi-Dirac distribution function, and \( \Theta_c(\epsilon) = \frac{|\epsilon|}{\epsilon + \Lambda} \), with \( \Lambda = \Lambda_0 e^{-l} \), introduced to continuously integrate out high energy modes in the renormalization group flow. For the initial condition at \( l = 0 \), \( \Lambda_0 \) is set to be much larger than the bandwidth, and the four point functions are initialized as \( V_{xxx} = V_{xxy} = V_{xys} = V_{yyx} = 0 \), \( V_{xxs} = V_{xyy} = V_{yxs} = V_{yys} = U - J \), with others initialized at 0 in absence of intraband interaction. The generated intra-band interactions in the renormalization group flow are kept in our calculation.

**Calculation of susceptibility.** In presence of an external perturbation \( \delta H = \int d\mathbf{r} \sum_{\nu \alpha} \delta h_{\nu \alpha} \psi^\dagger_{\nu \alpha} \psi_{\nu \alpha} \), the susceptibility is obtained from linear response theory,

\[
\chi_{\nu \alpha} = \frac{\partial \langle \psi^\dagger_{\nu \alpha} \psi_{\nu \alpha} \rangle}{\partial h_{\nu \alpha}}
\]

\[
\chi_{\nu \alpha} = \left[ -\delta_{\sigma_1 \sigma_2} \delta_{\sigma_3 \sigma_4} \delta_{v_{1s} v_{2s}} \delta_{v_{3s} v_{4s}} \Pi^{ph}_{\nu_1 \nu_2}\Pi^{ph}_{\nu_3 \nu_4} \right]
\]

where the self-energy corrections are neglected following a standard approximation in simplification of functional renormalization group flow [27]. The first term in Eq. (13) is the non-interacting part, and the specification to \( \nu \) and \( \alpha \) is neglected following a standard approximation in simplification of functional renormalization group flow [27]. The first term in Eq. (13) is the non-interacting part, and the specification to \( \nu \) and \( \alpha \) is neglected following a standard approximation in simplification of functional renormalization group flow [27].

The symbol \( \sum \) performs summation over the indices \( v_1 \sigma_1, v_2 \sigma_2, v_3 \sigma_3, \) and \( v_4 \sigma_4 \). The unitary transformation is determined according to the definition of \( \hat{\Phi} \) operators as,

\[
X_{j, m, n}^{m', n'}(q, q') = \sum U_{j, m, n}^{m', n'}(q, q') \chi_{s, s'}^{j, m, n} \chi_{s', s'}^{j, m, n'}
\]

with \( T \) the matrix corresponds to transformation of \( s \)- and \( p \)-orbitals into the angular momentum basis (Eq. (4)). The susceptibility towards the order formation in \( \hat{\Phi}_m \) channels is obtained as

\[
\chi_{SO} = \chi_{lm, s}^{l, s'}(0, 0) - \chi_{lm, s}^{l, s'}(0, 1) = -\Pi^{ph}_{\nu_1 \nu_2}(\Lambda_0) - [V_{xxx}(l \rightarrow +\infty) - V_{xxx}(l \rightarrow +\infty)] \Pi^{ph}_{\nu_1 \nu_2}(\Lambda_0)^2.
\]

The numerical results shown in the main text are obtained by solving the full flow equation. The analytic expression in Eq. 7 is obtained in the framework of renormalization group flow by solving the flow equation keeping the divergent \( (\sim \Delta^{-1/2}) \) contributions only, yielding \( \hat{\Phi}_m = -\Pi^{ph}_{\nu_1 \nu_2} V^{2}_{xxx} \).

**Free energy of the spin-orbit intertwined order.** Having a diverging correlation length in \( \langle \hat{\Phi}_m^{\dagger}(z) \hat{\Phi}_{m'}(z') \rangle \) implies the order acquires an expectation value \( \langle \hat{\Phi}_m \rangle = \Phi_m \), which is a three-component complex field. From SU(2) symmetry, the associated Ginzburg Landau free energy reads as

\[
F = -r \Phi^{\dagger} \Phi + c_0 (\Phi^{\dagger} \Phi)^2 + c_2 (\Phi^{\dagger} \tilde{L} \Phi) \cdot (\Phi^{\dagger} \tilde{L} \Phi) + O(\Phi^{\dagger} \Phi^3),
\]

with \( L_x, L_y, L_z \) the standard spin-1 representation matrix of SU(2) group. To relate to our Fermionic theory, the phenomenological couplings \( r, c_0 \) and \( c_2 \) are calculated with a Hubbard-Stratonovich transformation as

\[
r = \int \frac{dk}{2\pi} \frac{|J + J' - U|^2}{2\pi \epsilon_p(k) - \epsilon_s(k)},
\]

\[
c_0 = \frac{1}{2} \int \frac{dk}{2\pi} \frac{|J + J' - U|^4}{(\epsilon_p(k) - \epsilon_s(k))^2},
\]

\[c_2 = \frac{1}{2} \int \frac{dk}{2\pi} \frac{|J + J' - U|^4}{(\epsilon_p(k) - \epsilon_s(k))^2}.\]
In our theory, the coefficient $c_2$ vanishes for an accidental symmetry, causing an emergent SU(3) symmetry in the free energy. Then all states with the same order parameter amplitude $\sqrt{\Phi_0}\Phi$ are degenerate in energy, although there are two distinctive states analogous to the polar and ferromagnetic phases in spin-1 superfluids [28, 29]. The order parameter strength is then given by $\sqrt{\frac{1}{2\delta}}$, and its small band gap limit is given in the main text.

However the above degeneracy is lifted by considering a circular motion induced Zeeman splitting for electrons, which is described by a Hamiltonian

$$\Delta H = \delta \int dz \left[ i\psi_\uparrow^\dagger(z) \psi_\uparrow - i\psi_\downarrow^\dagger(z) \psi_\downarrow + H.c. \right].$$

This leads to a symmetry-breaking term in the Free energy,

$$\Delta F = -\delta \sum_{m_s} m_s |\Phi_{m_s}|^2 \int \frac{dk}{2\pi} |\epsilon_p(k) - \epsilon_i(k)|^2.$$  

Minimizing the total free energy leads to $\Phi_+ = \phi$, $\Phi_0 = \Phi_- = 0$ for $\delta > 0$. The remaining unbroken $U(1)$ symmetry, $\phi \rightarrow \phi e^{i\theta}$, corresponds to the rotation symmetry around the z direction.

Quasi-particle Hamiltonian. Having an order $\Phi_+ = \phi$, $\Phi_0 = \Phi_- = 0$, the bare electrons turn into dressed quasi-particles, whose Hamiltonian reads as $H_{QP} = H_0 + H_{SOI}$, with

$$H_{SOI} = -(U - J - J') \int dz \left[ \phi^* \hat{\sigma}^{\dagger}_{+1}(z) + H.c. \right].$$

Its explicit form in terms of $\tilde{\psi}$ operators is given in Eq. (8), where we have invoked spin-1/2 and spin-1 matrices

$$\sigma_+ = \begin{bmatrix} 0 & 2 \\ 0 & 0 \end{bmatrix}, \quad L_- = \begin{bmatrix} 0 & 0 \\ 2 & 0 \end{bmatrix}.$$ (22)

The eigenmodes describing quasi-particles are introduced through a unitary transformation,

$$\begin{bmatrix} \tilde{\psi}_{+\uparrow}(k) \\ \tilde{\psi}^\dagger_0(k) \\ \tilde{\psi}_{-\downarrow}(k) \end{bmatrix} = \begin{bmatrix} 1 & 0 & 0 \\ 0 & \cos(\vartheta_k/2)e^{i\varphi/2} & -\sin(\vartheta_k/2)e^{-i\varphi/2} \\ 0 & \sin(\vartheta_k/2)e^{i\varphi/2} & \cos(\vartheta_k/2)e^{-i\varphi/2} \end{bmatrix} \begin{bmatrix} \psi_{+\uparrow}(k) \\ \psi^\dagger_0(k) \\ \psi_{-\downarrow}(k) \end{bmatrix},$$ (23)

$$\begin{bmatrix} \tilde{\psi}_{+\downarrow}(k) \\ \tilde{\psi}^\dagger_0(k) \\ \tilde{\psi}_{-\uparrow}(k) \end{bmatrix} = \begin{bmatrix} \cos(\vartheta_k/2)e^{-i\varphi/2} & \sin(\vartheta_k/2)e^{i\varphi/2} & 0 \\ -\sin(\vartheta_k/2)e^{-i\varphi/2} & \cos(\vartheta_k/2)e^{i\varphi/2} & 0 \\ 0 & 0 & 1 \end{bmatrix} \begin{bmatrix} \psi_{+\downarrow}(k) \\ \psi^\dagger_0(k) \\ \psi_{-\uparrow}(k) \end{bmatrix},$$ (24)

with $\cos \vartheta_k = [\epsilon_p(k) - \epsilon_i(k)]/\sqrt{[\epsilon_p(k) - \epsilon_i(k)]^2 + 2(U - J - J')^2[\phi]^2}$, $\sin \vartheta_k = (U - J - J')\phi/\sqrt{[\epsilon_p(k) - \epsilon_i(k)]^2 + 2(U - J - J')^2[\phi]^2}$, $\varphi = \arg(-i\phi^*)$. We emphasize that the renormalized modes $\tilde{\psi}_{m,\alpha}$ still mainly carry their original spin moment and angular momentum.

The induced quasi-particle Hamiltonian associated with the conduction band is then obtained to be

$$H_{SOI} = \int \frac{dk}{2\pi} \lambda_{so}(k) \left[ \tilde{\psi}^\dagger_{+\uparrow}(k) \tilde{\psi}_{+\uparrow}(k) + \tilde{\psi}^\dagger_{-\downarrow}(k) \tilde{\psi}_{-\downarrow}(k) - \tilde{\psi}^\dagger_{-\uparrow}(k) \tilde{\psi}_{-\uparrow}(k) - \tilde{\psi}^\dagger_{+\downarrow}(k) \tilde{\psi}_{+\downarrow}(k) \right],$$ (25)

with a momentum dependent SOI strength

$$\lambda_{so}(k) = [\epsilon_p(k) - \epsilon_i(k)]/2 - \sqrt{[\epsilon_i(k) - \epsilon_p(k)]^2}/4 + (J - U - J')^2[\phi]^2/2.$$ (26)

Near the band edge, the induced SOI strength further simplifies to the expression given in Eq. (2).
S-1. SYMMETRY PROPERTIES

In this supplementary section, we provide more details of our symmetry analysis. In the basis of $\psi_{q,\alpha}$, the anti-unitary time-reversal symmetry transformation ($T$) is represented as

$$T \psi_{q,\alpha} T^{-1} = i^{2\alpha} (-1)^q \psi_{-q,-\alpha}. \quad (S1)$$

$$T \psi_{q,\alpha}^\dagger T^{-1} = i^{2\alpha} (-1)^q \psi_{-q,-\alpha}. \quad (S2)$$

The spatial parity transformation ($P$) is represented as

$$P \psi_{q,\alpha} P^\dagger = (-1)^q \psi_{q,\alpha}. \quad (S3)$$

Under spatial rotation ($R_\theta$) around the $z$-axis, we have

$$R_\theta \psi_{q,\alpha} R_\theta^\dagger = \psi_{q,\alpha} e^{i\theta}. \quad (S5)$$

Then the corresponding symmetry properties of the composite operators $B_{j,m,m',q}$ are derived as listed in Table I in the main text.

![Graph](image)

**Fig. S1.** The susceptibility corresponding to the channel of $[B_{0,0,1,0} - B_{0,0,1,-1}] / \sqrt{2}$ with renormalization group calculation. The plots correspond to different choices of interaction strengths ($U, J, J'$) and band gap ($\Delta$) in the tight binding model (see Methods). The tunneling of the $s$-orbital electron, or one half of bandwidth of the $s$-band is set as the energy unit here. In (a, b, c, d), we have $(U,J,J') = (2, -1, -0.5), (2, 0.1, -0.5), (2, 1, -0.5),$ and $(2, -1, 0.5),$ respectively. This susceptibility remains non-divergent at low temperature in (a, b). In (c, d), we find a divergent susceptibility, which causes the strong suppression of spin-orbit intertwined order at low temperature shown in the main text.

S-2. SUSCEPTIBILITIES IN OTHER CHANNELS

In this supplementary section, we provide other relevant susceptibility channels which affect the spin-orbit intertwined order in the functional renormalization group flow. In Fig. S1, we provide the susceptibility corresponding to a time-reversal odd spin singlet channel,

$$[B_{0,1,0,0} - B_{0,1,-1,0}] / \sqrt{2}. \quad (S6)$$

In Fig. S1(a, b), this susceptibility does not diverge at low temperature, which then does not cause suppression of the spin-orbit intertwined order as shown in Fig. 2(a, b) in the main text. In Fig. S1(c, d), the susceptibility diverges at
low temperature, which causes the strong suppression of the spin-orbit intertwined order at low temperature as shown in Fig. 2(c, d).

In Fig. S2, we provide the susceptibility corresponding to a time-reversal odd spin triplet channel,

\[ \frac{\mathcal{B}_{1,m_s,1;0} + \mathcal{B}_{1,m_s,1;1}}{\sqrt{2}}. \] (S7)

This susceptibility is non-divergent in Fig. S2(a, b, c). In Fig. S2(d), this susceptibility diverges.

**FIG. S2.** The susceptibility corresponding to the channel of \[ \frac{\mathcal{B}_{1,m_s,1;0} + \mathcal{B}_{1,m_s,1;1}}{\sqrt{2}}. \] The parameter choices are the same as in Fig. S1. This susceptibility remains non-divergent at low temperature in a, b and c. In d, we find the susceptibility diverges.