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 reaching the energy scale of room temperature, which could support the large spin polarization observed in CISS. 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.

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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^4 [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 [4–18]. This fascinating phenomenon has been observed in a broad range of chiral molecules, from DNA [3,6] and protein [8], to α-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, which demands a theoretical explanation. Previous theoretical studies have shown that a sufficiently strong SOI is required for tight-binding models to reproduce the experimental features [20–23], although the intrinsic spin orbit interaction in these molecules mainly containing carbon atoms is 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 [24] beyond the conventional consideration of the quantum electrodynamics.

In the study of atomic Bose-Einstein condensates, a quantum fluctuation enabled spin-momentum intertwined order has been proposed in theory [25] to explain the observed spin-momentum locking in a hexagonal optical lattice without bare SOI [26]. 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 [27]. These theoretical studies are inspiring, suggesting that spin-orbit coupling could emerge from many-body correlation even in complete absence of single-particle SOI, although the bosonic theories do not apply to electrons bearing CISS in chiral molecules.

In this work, we assume a rotation symmetry around the molecule elongation direction (to be refereed to 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

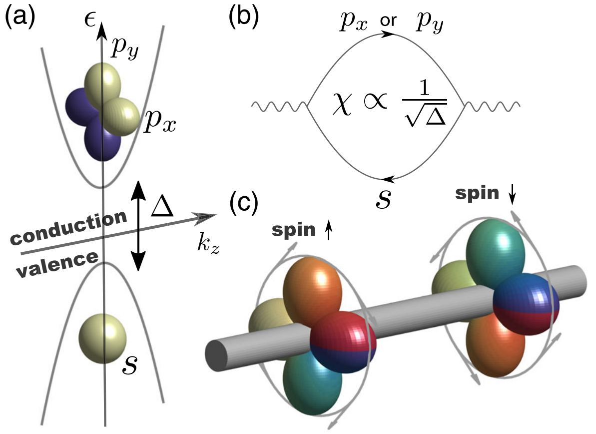

FIG. 1. Electron-hole paring in the three-orbital model. (a) Illustration of the three orbital (s, p_x, and p_y) model. The valence and conduction bands have s- and p-orbital character, respectively. The band gap is Δ. (b) The Feynman diagram that leads to the divergent susceptibility towards forming electron-hole pairing across the valence and conduction bands. (c) Sketch of the spin-orbit intertwined order with the spin and angular-momentum of the electron-hole pair spontaneously coupled.

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

$$\lambda_{\rm so}\hat{\sigma}_z \otimes \hat{L}_z/2, \tag{1}$$

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 Δ, 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' [28], the induced SOI strength is given as

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

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 $t_s + t_p = 2$ eV, $U - J - J' = 1$ eV and $\Delta = 0$—the parameter choice corresponds to an interaction estimate [28] for a right-handed peptide 3_{10} helix with a triplet-paired band structure [31]—the induced SOI reaches 0.1 eV, which suffices for modeling the unexpectedly large CISS effects in chiral molecules that persist up to room temperature [24]. 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),

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

Here $\nu = s$, p_x, or p_y index the orbitals, and α the spin degrees of freedom; P_ν represents the parity, i.e., $+$ and $-$ for s and p orbitals, respectively; m_s and $m_{p_x} = m_{p_y} \equiv m_p$ the effective mass associated with the motional dynamics of the valence and conduction bands along the molecular elongation direction. The field operators $\hat{\psi}_{\nu\sigma}$ incorporate the low energy degrees of freedom of electrons moving in a molecule near the band edge. For example, in modeling a right-handed peptide helix with a triplet-paired band structure [31], our three-orbital model corresponds to an effective description of electronic properties of the molecule, with s and p orbitals representing the quantized transverse modes. Considering a carbon atomic chain, our theory corresponds to the Fermi energy lying in between the sp-hybridized σ^* bond and π 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, $\chi^0_{\rm sp} = \partial_h\langle\hat{\psi}^\dagger_{s\alpha}\hat{\psi}_{p\alpha'} + \text{H.c.}\rangle = \sqrt{(m_s^{-1} + m_p^{-1})/2\Delta}$, considering a fictitious infinitesimal perturbation $\Delta H = -h\int dz[\hat{\psi}^\dagger_{s\alpha}\hat{\psi}_{p\alpha'} + \text{H.c.}]$. 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. In order to have spin-orbit coupled effect from the divergent electron-hole pairing, the idea here is to form a quantum superposition between the s orbital and the finite angular momentum $p_x \pm ip_y$ state that preserves time-reversal symmetry. This would then provide the required effective spin-orbit coupling. To form an orbital superposition in this channel, we need to assume that 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. Such physics does not occur in the previous study of correlation effects in a single-band setting [23].

The divergence in the 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' [28] are the only allowed local interactions between the s and p orbitals. Because of the multi-orbital complexity of our model, there

TABLE I. Symmetry properties of electron-hole pairings. We have introduced quantum numbers j_s and m_s 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 θ, the operators transform as $B_{j_s,m_s,m_l;q} \to B_{j_s,m_s,m_l;q}e^{im_l\theta}$, determined by the quantum number m_l. Under time-reversal symmetry (TRS) transformation, we have $B_{j_s,m_s,m_l;q} \to (-)^{j_s+1}(-)^{m_l+m_s}B_{j_s,-m_s,-m_l;-q}$. The even or odd sign of TRS listed here is determined for an operator $\hat{\mathcal{O}}$ according to whether its corresponding Hermitian observable $h\hat{\mathcal{O}} + h^*\hat{\mathcal{O}}^\dagger$ (with h an arbitrary complex number) is TRS even or odd. The operators with $m_l = 0, -1, -2$ are not listed here due to the constraint that $B^\dagger_{j_s,m_s,m_l;q} = (-)^{m_l+m_s+1}B_{j_s,-m_s,-m_l;q+m_l}$.

j_s, m_s, m_l	Operators	SU(2)	Parity	TRS
0,0,0	$\hat{\mathcal{B}}_{0,0,0;0}, (\hat{\mathcal{B}}_{0,0,0;+1} + \hat{\mathcal{B}}_{0,0,0;-1})/\sqrt{2}$ $(\hat{\mathcal{B}}_{0,0,0;+1} - \hat{\mathcal{B}}_{0,0,0;-1})/\sqrt{2}$	Singlet	Even	Even Odd
0,0,1	$(\hat{\mathcal{B}}_{0,0,1;0} + \hat{\mathcal{B}}_{0,0,1;-1})/\sqrt{2}$ $(\hat{\mathcal{B}}_{0,0,1;0} - \hat{\mathcal{B}}_{0,0,1;-1})/\sqrt{2}$	Singlet	Odd	Even Odd
0,0,2	$\hat{\mathcal{B}}_{0,0,2;-1}$	Singlet	Even	Even
$1, m_s \in \{0, \pm 1\}, 0$	$\hat{\mathcal{B}}_{1,m_s,0;0}, (\hat{\mathcal{B}}_{1,m_s,0;+1} + \hat{\mathcal{B}}_{1,m_s,0;-1})/\sqrt{2}$ $(\hat{\mathcal{B}}_{1,m_s,0;+1} - \hat{\mathcal{B}}_{1,m_s,0;-1})/\sqrt{2}$	Triplet	Even	Odd Even
$1, m_s \in \{0, \pm 1\}, 1$	$(\hat{\mathcal{B}}_{1,m_s,1;0} + \hat{\mathcal{B}}_{1,m_s,1;-1})/\sqrt{2}$ $(\hat{\mathcal{B}}_{1,m_s,1;0} - \hat{\mathcal{B}}_{1,m_s,1;-1})/\sqrt{2}$	Triplet	Odd	Odd Even
$1, m_s \in \{0, \pm 1\}, 2$	$\hat{\mathcal{B}}_{1,m_s,2;-1}$	Triplet	Even	Odd

are 21 independent channels in the particle-hole pairing function, $G_{\nu\alpha,\nu'\alpha'} = \langle \hat\psi^\dagger_{\nu\alpha}\hat\psi_{\nu'\alpha'}\rangle$. According to symmetry properties, we group all the particle-hole pairings into the following channels,

$$\hat{\mathcal{B}}_{j_s,m_s,m_l;q} = (-1)^{q+1}\sum_{\alpha\alpha'} i^{2\alpha} C_{\frac{1}{2}\frac{1}{2}}(-\alpha,\alpha'|j_s,m_s)\hat\psi^\dagger_{q\alpha}\hat\psi_{q+m_l\alpha'}, \tag{4}$$

where the $C_{(1/2)(1/2)}$ matrix contains Clebsch-Gordon coefficients, and $\hat\psi_{q\alpha}$ is a field operator in angular momentum basis defined by $[\hat\psi_\pm \equiv \mp(\hat\psi_x \pm i\hat\psi_y)/\sqrt{2}, \hat\psi_0 \equiv \hat\psi_s]$. The symmetry properties of these operators are listed in Table I.

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

$$\hat{\mathcal{O}}_{m_s} \equiv \frac{1}{\sqrt{2}}[\mathcal{B}_{1,m_s,-1;0} - \mathcal{B}_{1,m_s,-1;1}], \tag{5}$$

and

$$\hat{\mathcal{O}}'_{m_s} \equiv \frac{1}{\sqrt{2}}[\mathcal{B}_{1,m_s,0;1} - \mathcal{B}_{1,m_s,0;-1}], \tag{6}$$

whose parities are odd and even, respectively. They both satisfy the symmetry requirement, but the parity even operators represent parings 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 $\hat{\mathcal{O}}_{m_s}$. 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 $\hat{\mathcal{O}}_{m_s}$ is

$$\chi_{\mathrm{SO}} = \frac{\chi^0_{sp}}{1-(U-J)\chi^0_{sp}} \tag{7}$$

under random phase approximation [28]. The divergence in the susceptibility for the noninteracting 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 ($J < 0$ in our notation). Without Josephson coupling, i.e., $J' = 0$, a degenerate channel—$[\mathcal{B}_{1,m_s,-1;0} + \mathcal{B}_{1,m_s,-1;1}]/\sqrt{2}$ 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. In the case of Josephson coupling being negligible, as is expected for elongated chiral molecules (see Supplemental Material [28]), the presence of orbital motion induced Zeeman splitting selects the time reversal symmetric over the asymmetric pairing.

Since all parity odd pairing channels in our theory potentially have a large susceptibility due to the divergence in χ^0_{sp}, 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 [32]. The results for the susceptibility of forming the spin-orbit intertwined order in Eq. (6) are shown in Fig. 2. It is confirmed that having J and J'

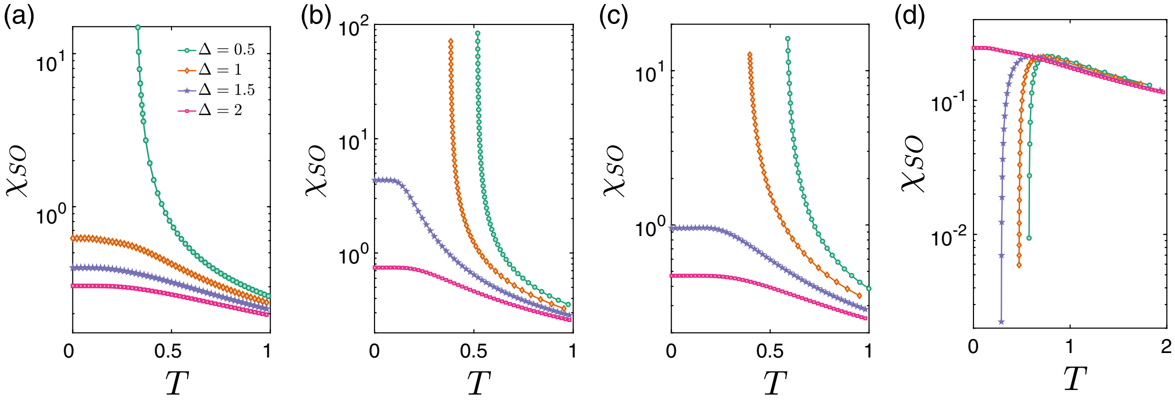

FIG. 2. Divergent susceptibility for a spin-orbit intertwined order with renormalization group calculation. The plots correspond to different choices of interaction strengths (U, J, J') and band gap (Δ) in the tight binding model [28]. The tunneling of the p-orbital electron, or one-half of bandwidth of the p band is set as the energy unit here. In (a),(b),(c),(d) we have $(U, J, J') = (2, 0, 0)$, $(U, J, J') = (2, -1, -0.5)$, $(2, 0.1, -0.5)$, and $(2, -1, 0.5)$, respectively, and set the s-band tunneling $t_s = 2$. With both of J and J' negligible (set to zero in this plot) in (a), a divergent susceptibility at finite temperature (T) in the time-reversal symmetric spin-orbit intertwined channel [Eq. (6)] is confirmed, which agrees with the analytical results from random phase approximation. In (b), we find that with both of J and J' being negative, the susceptibility in the spin-orbit intertwined channel is more strengthened, compared to (a). In (c), we find that a positive Hund's rule coupling $J > 0$ does not immediately cause suppression of the spin-orbit intertwined order. (d) Shows that a positive Josephson coupling $J' > 0$ does not support the time-reversal symmetric spin-orbit intertwined order.

vanishing, the susceptibility for $\hat{\mathcal{O}}_{m_s}$ diverges at finite temperature [Fig. 2(a)]. The chiral peptide 3_{10} helix corresponds to this case according to interaction strength estimate [28]. The divergence requires the interaction energy to conquer the band gap barrier of the sp-orbital pairing. The susceptibility is more strengthened for both J and J' being negative [Fig. 2(b)]. We also find that having a weak antiferromagnetic coupling in J still supports divergent susceptibility in $\hat{\mathcal{O}}_{m_s}$ channel [Fig. 2(c)]. For a large enough positive J, this susceptibility is no longer divergent, but instead meets a strong suppression at low temperature, which is due to a divergent susceptibility in a different channel, $\hat{\mathcal{B}}_{0,0,\pm1,q}$ [28]. A sign change in the Josephson coupling leads to a low-temperature suppression of $\hat{\mathcal{O}}_{m_s}$ pairing [Fig. 2(d)], owing to a divergent susceptibility in the time-reversal odd channel of $[\mathcal{B}_{1,m_s,-1;0} + \mathcal{B}_{1,m_s,-1;1}]/\sqrt{2}$ [28]. We expect that further considering scattering with additional orbitals present in molecules would renormalize J' to the negative side due to BCS channels [32], which would make the time-reversal symmetric spin-orbit intertwined order even more favorable.

A divergence in the susceptibility χ_{SO} implies the correlation length of $\langle \hat{\mathcal{O}}^{\dagger}_{m_s}(z)\hat{\mathcal{O}}_{m_s'}(z')\rangle$ reaches the size of the molecule. Taking mean field approximation, the operator $\hat{\mathcal{O}}_{m_s}$ acquires a finite expectation value, $\langle \hat{\mathcal{O}}_{m_s}\rangle \equiv \Phi_{m_s}$. The free energy of the order parameter takes an SU(3) symmetric form as shown in the Supplemental Material [28]. 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 [33,34] or liquid helium [35].

Further considering a circular motion induced Zeeman splitting for electrons [22], we have a perturbative coupling $\Delta H = \delta \int dz (i\psi^{\dagger}_{x\uparrow}\psi_{y\uparrow} - i\psi^{\dagger}_{x\downarrow}\psi_{y\downarrow} + \mathrm{H.c.})$. Despite its insufficient strength to model CISS [22,24], this term determines a spin quantization axis, and triggers an order $(\Phi_{+1} \equiv \phi, \Phi_0 = 0, \Phi_{-1} = 0)$ to minimize to total free energy in our theory, from which a strong SOI emerges from electron correlation,

$$H_{\mathrm{SOI}} = \frac{U - J - J'}{4\sqrt{2}} \int dz [i\phi \Psi^{\dagger}(z)\sigma_{+} \otimes L_{-}\Psi(z) + \mathrm{H.c.}], \tag{8}$$

with $\Psi \equiv [\psi_{1,\uparrow}, \psi_{0,\uparrow}\psi_{-1,\uparrow}, \psi_{1,\downarrow}, \psi_{0,\downarrow}\psi_{-1,\downarrow}]^{T}$, σ_{+} and L_{-} the standard spin-1/2 and spin-1 angular momentum matrices [28].

This correlation induced coupling breaks spin SU(2) and reflection symmetries with the time reversal symmetry unbroken. Through a unitary transformation into the quasiparticle basis, the induced coupling in the conduction band takes a more standard SOI form given in Eq. (2). The induced SOI strength for the quasiparticles reaches to the order of the energy scale of room temperature, and is sufficient to model the spin dependent transmission observed in chiral molecule experiments [24].

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 correlation 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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