Odd-Parity Superconductivity in the Vicinity of Inversion Symmetry Breaking in Spin-Orbit-Coupled Systems

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We study superconductivity in spin-orbit-coupled systems in the vicinity of inversion symmetry breaking. We find that, because of the presence of spin-orbit coupling, fluctuations of the incipient parity-breaking order generate an attractive pairing interaction in an odd-parity pairing channel, which competes with the s-wave pairing. We show that Coulomb repulsion or an external Zeeman field suppresses the s-wave pairing and promotes the odd-parity superconducting state. Our work provides a new mechanism for odd-parity pairing and opens a route to novel topological superconductivity.

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Over the last few years, the search for unconventional superconductors has received a new impetus from the study of topological phases of matter. After early works on superfluid Helium-3 [1] and recent developments on topological insulators, it has been theoretically established [2] that superconducting states can be classified by their topological properties. Unlike conventional s-wave superconductors, topological superconductors are predicted to harbor exotic quasiparticle excitations on the boundary. There is currently intensive effort searching for topological superconductivity in naturally occurring materials (see for example, Refs. [3–14]), though definitive experimental evidence is still lacking. For the majority of superconductors that are time-reversal and inversion symmetric, the single most important requirement for being topologically nontrivial is that the pairing order parameter must be odd under spatial inversion [3,15], e.g., p- or f-wave. This rekindles interest in finding odd-parity superconductors.

The parity of the pairing order parameter is tied with its spin. In the absence of spin-orbit coupling, odd-parity pairing is spin-triplet and vice versa. It has long been known from studies on superfluid He [16] that triplet pairing can be induced by enhanced ferromagnetic spin fluctuations in the vicinity of ferromagnetic instability. This mechanism for triplet pairing, if realized in the solid-state, can lead to a topological superconductor [17], analogous to the topological superfluid He.

In this Letter, we propose an alternative mechanism for odd-parity pairing in the vicinity of nonmagnetic, inversion-symmetry-breaking phases in spin-orbit-coupled systems. In the presence of spin-orbit interaction, such parity-breaking orders directly couple to an electron’s spin texture on the Fermi surface [18]. As a result, the fluctuations of an incipient parity-breaking order, which we call “parity fluctuations,” generate an effective interaction that is strongly momentum and spin dependent. Without assuming any special features of the Fermi surface, we show on general ground that this effective interaction is attractive in both the s-wave and an odd-parity pairing channel. Moreover, the pairing interactions in the two channels are found to be of the same order of magnitude, and in several cases, roughly equal. We show that either Coulomb interaction or Zeeman field suppresses the s-wave pairing and promotes the odd-parity superconducting state on the border of parity-breaking order. Finally, we propose the pyrochlore oxide Cd₂RE₂O₇ and doped SrTiO₃ heterostructures as candidate systems where odd-parity superconductivity mediated by parity fluctuations may be realized.

In this work, we consider parity-breaking orders that are time-reversal invariant and carry zero momentum. Such order may originate from an unstable odd-parity phonon or the electron-electron interaction. The order parameter can be represented by a Hermitian fermion bilinear operator \( \hat{Q} \) with the same symmetry, which takes the form

\[
\hat{Q} = \sum_{\mathbf{k},\alpha\beta} \Gamma_{\alpha\beta}(\mathbf{k}) \hat{c}_{\mathbf{k}\alpha}^\dagger \hat{c}_{\mathbf{k}\beta}, \quad \text{with} \quad \Gamma^\dagger(\mathbf{k}) = \Gamma(\mathbf{k}). \tag{1}
\]

Here \( \alpha, \beta \) are pseudospin indices for the two degenerate states at every \( \mathbf{k} \). In spin-orbit-coupled systems, these states are not spin eigenstates but remain degenerate in the presence of time-reversal (\( \Theta \)) and inversion (\( P \)) symmetry [19]. For simplicity of notation, we have chosen an Ising-type parity-breaking order in Eq. (1). Vector and high-rank tensor orders are described by a multiplet of Hermitian operators denoted by \( \hat{Q}^\dagger \); these orders will be encountered later.

Different types of parity-breaking orders are classified by their transformation properties under crystal symmetry operations, which act on an electron’s spin and momentum jointly. Before proceeding to the symmetry analysis, we emphasize that the form factors for physical observables, such as \( \Gamma(\mathbf{k}) \) for \( \hat{Q} \), depend on the basis for the doubly degenerate energy band. For the purpose of this work, it is most convenient to choose the “manifestly covariant Bloch
basis” (MCBB) [18]. In this basis, the two-component electron operator \((c_{k_1}^\dagger, c_{k_2}^\dagger)\) transforms simply as a spinor field in \(k\) space under time reversal and crystal symmetry operation \(g \in O(3)\):

\[
\Theta c_{k_\alpha}^{\dagger} \Theta^{-1} = c_{\alpha'}^{\dagger} c_{-k_{\beta'}}, \tag{2}
\]

\[
g c_{k_\alpha}^{\dagger} g^{-1} = U_{\alpha\beta}(g) c_{k_\beta'}, \tag{3}
\]

where \(k^* = g k\) is the star of \(k\), \(e_{\alpha\beta}\) is the Levi-Civita symbol, and \(U(g)\) is the \(U(2)\) matrix that represents the action of \(g\) on the pseudospin, in the same way as it acts on the spin of a free electron.

It then follows from the symmetry transformation laws (2, 3) that the form factor \(\Gamma(k)\) of time-reversal-invariant and parity-breaking orders satisfies the condition

\[
\Gamma(k) = c \Gamma^*(-k) = -\Gamma(-k), \tag{4}
\]

and hence takes the general form [18]

\[
\Gamma(k) = d_k \cdot \sigma, \quad \text{with} \quad d_k = -d_{-k}, \tag{5}
\]

where \(\sigma = (\sigma^x, \sigma^y, \sigma^z)\) denotes Pauli matrices in pseudospin space. The \(d\)-vector field \(d_k\) defines the pseudospin splitting in the ordered state, whose magnitude and direction vary over the Fermi surface.

It may seem counterintuitive that nonmagnetic parity-breaking orders, such as structural distortion and orbital order, couple to an electron’s spin. As we show by example in the Supplemental Material [20], this remarkable fact is a general consequence of spin-orbit interaction in centrosymmetric systems. It will play a crucial role in mediating superconductivity in the vicinity of parity-breaking order.

In contrast, for spin-rotationally invariant systems, the above symmetry analysis implies that parity-breaking orders at zero momentum cannot couple directly to electrons on the Fermi surface, unlike the nematic order that is even parity [24,25]. Therefore, spin-orbit coupling is crucial for superconductivity mediated by odd-parity phonons or parity fluctuations.

In a system close to a parity-breaking instability, the effective interaction arising from the order parameter fluctuations is given by

\[
H_{\text{eff}} = \sum_q V_q \hat{Q}(q) \hat{Q}(-q), \tag{6}
\]

where \(\hat{Q}(q) = \hat{Q}^\dagger(-q)\) is the Fourier transform of the order parameter field in real space:

\[
\hat{Q}(q) = \frac{1}{2} \sum_{k, \alpha\beta} (\Gamma_{\alpha\beta}(k + q) + \Gamma_{\alpha\beta}(k)) c_{k+q, \alpha}^{\dagger} c_{k, \beta}. \tag{7}
\]

Within the random-phase approximation (RPA), \(V_q\) can be expressed in terms of the \(q\) dependent susceptibility:

\[
V_q = I/[1 + \chi(q)I]. \quad \text{\(V_q\) is enhanced and has a maximum at \(q = 0\) close to a \(q = 0\) instability. Restricting the effective interaction (6) to the Cooper pairing channel with zero total momentum, we obtain the pairing interaction

\[
H_p = \sum_{k, k'} V_{a'b'd'}(k, k') c_{k_{\alpha}}^{\dagger} c_{-k_{\beta}}^{\dagger} c_{-k'_{\gamma}} c_{k'_{\delta}}. \tag{8}
\]

Using (5), (6), and (7), we find the momentum- and pseudospin-dependent interaction vertex \(V_{a'b'd'}(k, k')\) is given by

\[
V_{a'b'd'}(k, k') = -\frac{1}{8} |V_{\text{const}}| \sum_{i,j} (d_k^i d_{k'}^j + d_k^j d_{k'}^i) (\sigma^i_{a'b'} \sigma^j_{\gamma\delta} - \sigma^i_{a'\gamma'} \sigma^j_{\beta\delta'}).
\]

(9)

To proceed, we expand \(V_{k\pm k}\) in the pairing interaction (9) in terms of spherical harmonics on the Fermi surface: \(V_{k\pm k} = V_0 \mp V_{y} \hat{q} \cdot \hat{k} + \ldots\). Below we consider the leading term \(V_0\). Despite the fact that \(V_0\) is a constant, the interaction vertex (9) inherits the form factor of the parity-breaking order parameter \(\hat{Q}\), which is strongly pseudospin and momentum dependent. It consists of two types of terms: \(V = V^e + V^o\), where \(V^e\) contains the product of components with the same momentum:

\[
V^e_{a'b'd'}(k, k') = -\frac{V_0}{8} \sum_{i,j} (d_k^i d_{k'}^j + d_k^j d_{k'}^i) (\sigma^i_{a'b'} \sigma^j_{\gamma\delta} - \sigma^i_{a'\gamma'} \sigma^j_{\beta\delta'}),
\]

and \(V^o\) contains the cross terms:

\[
V^o_{a'b'd'}(k, k') = -\frac{V_0}{8} \sum_{i,j} (d_k^i d_{k'}^j + d_k^j d_{k'}^i) (\sigma^i_{a'b'} \sigma^j_{\gamma\delta} + \sigma^i_{a'\gamma'} \sigma^j_{\beta\delta'}).
\]

Note that \(V^e (V^o)\) is an even (odd) function of \(k, k'\), and antisymmetric (symmetric) under exchanging the pseudospin indices either \(a\beta\) or \(\gamma\delta\). Therefore, \(V^e\) and \(V^o\) correspond to the even-parity pseudospin-singlet and odd-parity pseudospin-triplet pairing channels, respectively.

The above pairing interaction can be decomposed into different superconducting channels that belong to different representations of the crystal symmetry group. Before proceeding, we describe the general classification of time-reversal-invariant superconducting order parameters, taking the form

\[
\hat{F}^\dagger = \frac{1}{2} \sum_{k, a'b'} c_{a'b'} F_{a'b'}(k) c_{k_{a'}}^{\dagger} c_{-k_{b'}}^{\dagger},
\]

where the form factor \(F(k)\) satisfies the symmetry condition.
$F^\dagger(k) = F(k) = -eF^\dagger(-k)e$.  \hfill (11)

Moreover, it follows from (3) that $e_{\beta\gamma}c^\dagger_{k\alpha}c^{-1}_{-k\beta}$ has the same
transformation law under crystal symmetry operations as $c^\dagger_{k\alpha}c_{-k\beta}$. This implies every time-reversal-invariant superconducting order parameter has a counterpart in the particle-hole channel, with the same symmetry. In particular, odd-parity superconducting order parameters, which have $F(k) = -F(-k)$, admit the same classification as particle-hole orders $P$ described earlier.

To proceed with the classification, it is instructive to first consider the most symmetric group $O(3)$, the group of all joint 3D rotations and reflections of spin and momentum, from which all point groups descend. In this case, all possible odd-parity orders defined by the form factor (5) are classified by the total angular momentum $J$ and the orbital angular momentum $L$ (which must be odd) \cite{18}. At the lowest order $L = 1$, there are three types of particle-hole orders: gyrotropic, ferroelectric, and multipolar. The corresponding form factors are listed in Table I. Classification for 2D systems with $O(2)$ symmetry is also presented.

As expected from the one-to-one correspondence between particle-hole and particle-particle orders, these form factors also classify odd-parity pairing symmetries of spin-orbit-coupled superconductors. For example, the pairing order parameter with the isotropic form factor $\Gamma_1 = \mathbf{k} \cdot \sigma$ coincides with a particular choice of order parameters for the Balian-Werthamer phase of He-3 \cite{26}. On the other hand, the pairing order parameters with anisotropic form factors $\Gamma_2$ and $\Gamma_3$ are time-reversal invariant and spontaneously break the rotational symmetry, resulting in an odd-parity superconductor with nematic order \cite{6}. To our knowledge, such anisotropic phases have not been found in He-3; their existence requires spin-orbit coupling.

We now use the effective interaction $H_{\text{eff}}$ given by Eq. (6) to study superconductivity in the vicinity of each type of parity-breaking order in Table I; for multicomponent operator $\hat{Q}^\mu$, summation over $\mu$ is taken. In all cases, we restrict $H_{\text{eff}}$ into a Cooper pairing channel with zero momentum, and decompose the pairing interaction $H_p$ into various superconducting channels:

$$H_p = V_0 \left( a_0 \hat{S} + \sum_n a_n \sum_\mu \hat{F}_n^\dagger \hat{F}_n^\mu \right),$$  \hfill (12)

where $\hat{S} = (1/2)\sum_{k,\alpha\beta} e_{\alpha\beta} c^\dagger_{k\alpha} c_{-k\beta}$ is the s-wave superconducting order parameter, and $\hat{F}_n^\dagger$ denotes various odd-parity superconducting order parameters defined in Eq. (10) and classified in Table I. Here $n$ numerates different odd-parity pairing channels and, again, summation over $\mu$ implies summation over different components in the case of multicomponent pairing orders. Coefficients $a_n$ take different values for different types of interactions, and all are gathered in the Table II. Details of our calculation can be found in the Supplemental Material [20]. Since $V_0 < 0$, $a_n > 0$ means attractive interaction in the corresponding pairing channel.

From Table II, we obtain the superconducting instability driven by each type of parity fluctuations. In all cases, there is an instability in the s-wave channel, similar to phonon-mediated pairing in conventional superconductors. More importantly, in all cases except the multipolar orders $\Gamma_3$ and $\Gamma_4$, there is also an instability in the odd-parity channel with the same symmetry as the incipient particle-hole order that drives superconductivity. Remarkably, for Ising type orders described by a single-component $\hat{Q}$, the pairing attraction in the odd-parity channel is weaker than, but still of the same order of magnitude as, the one in the s-wave channel.

### Table I. Classification of odd-parity order parameters for spin-orbit coupled systems, and their transformation properties under joint spin and momentum rotations in three and two dimensions. Rank 2 tensor $\Gamma_3(\Gamma_4)$ is symmetric and traceless, and hence has 5 (2) independent components. All parity orders $F(k)$ admit the same classification as $\Gamma(k)$ and have exactly the same functional form.

| 3D system with $O(3)$ symmetry | Transformation property |
|---------------------------------|------------------------|
| $\Gamma_1(k) = (\mathbf{k} \cdot \sigma)$ | pseudoscalar |
| $\Gamma_2(k) = [\mathbf{k} \times \sigma]^\dagger$ | vector |
| $\Gamma_3(k) = \hat{k}^\dagger \sigma^\dagger + \hat{k}^\dagger \sigma^\dagger - \frac{3}{2} (\hat{k} \cdot \sigma) \delta^{ij}$ | rank 2 tensor |

| 2D system with $O(2)$ symmetry | Transformation property |
|---------------------------------|------------------------|
| $\Gamma_1(k) = \hat{k}^\dagger \sigma^\dagger + \hat{k}^\dagger \sigma^\dagger$ | pseudoscalar |
| $\Gamma_2(k) = \hat{k}^\dagger \sigma^\dagger - \hat{k}^\dagger \sigma^\dagger$ | vector |
| $\Gamma_3(k) = \hat{k}^\dagger \sigma^\dagger$ | |
| $\Gamma_4(k) = \hat{k}^\dagger \sigma^\dagger - (\hat{k} \cdot \sigma) \delta^{ij}$ | rank 2 tensor |

### Table II. Decomposition of the different types of interaction into different pairing channels; see Eq. (12). $n = 0$ denotes the s-wave channel; $n = 1, \ldots, 4$ denotes the odd-parity channels classified in Table I. $a_n > 0$ corresponds to attractive pairing interaction.

| Type of interaction | $a_0$ | $a_1$ | $a_2$ | $a_3$ |
|---------------------|-------|-------|-------|-------|
| $Q_1(q)Q_1(-q)$    | 1     | 1     | -1    | 0     |
| $Q_2(q)Q_2(-q)$    | 2     | -4/3  | 1/2   | -1/4  |
| $Q_3(q)Q_3(-q)$    | 20/3  | 0     | -5/3  | -1/2  |

| Type of interaction | $a_0$ | $a_1$ | $a_2$ | $a_3$ | $a_4$ |
|---------------------|-------|-------|-------|-------|-------|
| $Q_1(q)Q_1(-q)$    | 1     | 1     | -1    | -1    | 0     |
| $Q_2(q)Q_2(-q)$    | 1     | -1    | 1     | -1    | 0     |
| $Q_3(q)Q_3(-q)$    | 1     | -1/2  | -1/2  | 1     | -1/4  |
| $Q_4(q)Q_4(-q)$    | 4     | 0     | 0     | -4    | 0     |
Although fluctuations of multipolar orders in rotationally invariant systems do not lead to pairing in any odd-parity channel, the situation becomes different in real materials where the crystal symmetry is taken into account. In any crystals, the five components of rank 2 tensor \( \Gamma_3 \) invariably split into more than one representations of the point group. For example, for \( O_h \) point group, the diagonal and off-diagonal components of \( \Gamma_3 \) split to form \( e_g \) and \( t_{2g} \) representations, which have 2 and 3 independent components respectively. For many point groups such as \( D_{4h} \), \( \Gamma_4 \) also splits into one-dimensional representations, with form factors \( k_x \sigma_x - k_y \sigma_y \) and \( k_x \sigma_y + k_y \sigma_x \) respectively. We find fluctuations of such multipolar orders of reduced symmetry generate attractive pairing interaction in the odd-parity channel of the same symmetry. The interaction strength is weaker than the \( s \)-wave channel in the case of \( e_g \) and \( t_{2g} \) orders, and is equal to the latter in the case of Ising type \( \Gamma_4 \) orders [20].

The above finding of odd-parity pairing mediated by parity fluctuations in spin-orbit-coupled systems is the main result of this work. It is interesting to make a comparison with the mechanism of triplet pairing mediated by spin fluctuations. In that case, the effective interaction is given by

\[
\sum_{\mathbf{q}} V(\mathbf{q}) s(\mathbf{q}) \cdot s(-\mathbf{q}),
\]

where \( s \) is the spin operator and \( V(\mathbf{q}) = I/|1 + i \chi^*(\mathbf{q})| \) is determined by the spin susceptibility \( \chi^*(\mathbf{q}) \). Importantly, to obtain the pairing interaction in the triplet channel requires \( \chi^*(\mathbf{q}) \) to have a nontrivial \( \mathbf{q} \) dependence. Approximating \( \chi^*(\mathbf{q}) \) by its zeroth spherical harmonic, which is a constant, does not generate triplet pairing, simply because two electrons at the same spatial location cannot form a triplet. In contrast, we obtained odd-parity pairing in this leading-order approximation, without relying on any special features of the susceptibility of parity-breaking order.

Given that the pairing interaction we found has comparable or even identical strengths in the \( s \)-wave and odd-parity channels, small residual interactions or external perturbations become important in lifting the degeneracy and eventually determine which one of the two competing pairing symmetries is realized. An in-depth study of the effects of residual interactions necessarily involve material-specific details, which is beyond the scope of this work. Nonetheless, it should be noted that the Coulomb repulsion is maximal and has the strongest pair-breaking effect in the \( s \)-wave channel: this fact can make the odd-parity pairing energetically favorable. This role of Coulomb interaction in the competition between \( s \)-wave and odd-parity pairings has been recognized [3] and emphasized [27] in recent model studies.

In addition to Coulomb interaction, the \( s \)-wave pairing is suppressed by a magnetic field \( B \) that splits the spin degeneracy, which is pair-breaking and sets the Pauli limit for the upper critical field. However, Zeeman spin splitting has variable effects on odd-parity superconducting states in spin-orbit-coupled systems, as we show now. First, let us consider how the doubly degenerate bands at every \( \mathbf{k} \), or the pseudospin, split under a Zeeman field. The coupling of pseudospin to Zeeman field takes the general form

\[
H_Z = \sum_{\mathbf{k}} c^\dagger_{\mathbf{k}} g_{ij}(\mathbf{k}) B_i \sigma_j(\mathbf{k}) c_{\mathbf{k}}.
\]

The \( g \)-factor \( g_{ij}(\mathbf{k}) \) is a function of \( \mathbf{k} \), and can be expanded into different spherical harmonics over the Fermi surface. Importantly, since the pseudospin operator \( \sigma_i \) is defined in the manifestly covariant Bloch basis and has the same symmetry as an electron’s spin, \( g_{ij}(\mathbf{k}) \) generally has a dominant zeroth spherical harmonic component \( g^0_{ij} \).

Assuming \( g_{ij}(\mathbf{k}) = g^0_{ij} \), we obtain a uniform spin splitting over the Fermi surface, with a spin quantization axis in the direction of \( h \), for all \( \mathbf{k} \) on the Fermi surface. For example, in 2D systems with rotational symmetry, an in-plane field \( B \) induces a spin splitting in the direction parallel to the field. The odd-parity pairing with \( \Gamma_3(\mathbf{k}) = (k_x \sigma_z, k_y \sigma_z) \), whose \( d \)-vector is out of plane, is not Pauli limited. Therefore, Zeeman field is an effective way of tuning the competition between different pairing symmetries and promoting certain types of odd-parity superconductivity for which the Pauli limit is absent or largely enhanced.

Finally, we propose candidate materials for odd-parity superconductivity in the vicinity of parity-breaking order. First, the pyrochlore oxide \( \text{Cd}_2\text{Re}_2\text{O}_7 \) undergoes a continuous parity-breaking phase transition at \( T_p = 200 \text{ K} \) [28,29] with a large mass enhancement of conduction electrons [30], and becomes superconducting at \( T_c = 1.1 \text{ K} \) [31]. The application of high pressure has significant effects on these phases, and generates a variety of new phases identified from resistivity anomalies. Remarkably, around a critical pressure of \( P_c = 4.2 \text{ GPa} \) where the parity-breaking order is suppressed, an anomalously large upper critical field of 7.8 T is observed, which is 27 times larger than at ambient pressure and significantly higher than the Pauli limit 4.2 T evaluated as \( H_p = 1.84 T_c \) [32]. These phenomena seem to fit into the theoretical picture presented in this work. Therefore, we propose that the superconducting state of \( \text{Cd}_2\text{Re}_2\text{O}_7 \) around \( P_c \) is driven by parity fluctuations, and may have an odd-parity pairing symmetry.

Another candidate system is inversion-symmetric heterostructure of doped \( \text{SrTiO}_3 \) with intrinsic spin-orbit coupling [33]. Bulk \( \text{SrTiO}_3 \) is close to the ferroelectric instability and becomes superconducting upon electron doping [34,35]. In doped \( \text{SrTiO}_3 \) heterostructures with the superconducting dopant layer of a few nanometers thickness, the in-plane upper critical field exceeds the conventional Pauli limit [33]. It is worthwhile to examine the possibility of an odd-parity superconducting state under...
a large in-plane field, as we discussed earlier. A model study for possible superconducting phases in SrTiO$_3$ heterostructure will be presented elsewhere.

Throughout this work, we have stayed away from the immediate neighborhood of the quantum phase transition point, where long-wavelength and low-frequency fluctuations of the parity-breaking order pile up and the RPA type effective interaction used in this work is inapplicable. The physics in the quantum critical regime is an interesting topic which is left to future study.

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