Nontrivial interplay between superconductivity and spin-orbit coupling in non-centrosymmetric ferromagnets

Jacob Linder, Andriy H. Nevidomskyy, and Asle Sudbø

Department of Physics, Norwegian University of Science and Technology, N-7491 Trondheim, Norway
Department of Physics and Astronomy, Rutgers University, Piscataway, N. J., 08854-8019

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Motivated by the recent discoveries of ferromagnetic and non-centrosymmetric superconductors, we present a mean-field theory for a superconductor that both lacks inversion symmetry and displays ferromagnetism, a scenario which is believed to be realized in UIr. We study the interplay between the order parameters to clarify how superconductivity is affected by the presence of ferromagnetism and spin-orbit coupling. One of our key findings is that the spin-orbit coupling seems to enhance both ferromagnetism and superconductivity in all spin channels. We discuss our results in the context of the heavy fermion superconductor UIr and analyze possible symmetries of the order parameter by the group theory method.

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In the past decade, a number of superconductors have been discovered that are called `unconventional' as they fall outside the Bardeen-Cooper-Schrieffer (BCS) paradigm of electron-phonon mediated pairing with an isotropic gap. Of those, UPt$_3$, Sr$_2$RuO$_4$, and the heavy fermion compound UIr under pressure, where the presence of an internal FM moment strongly suggests that only the equal-spin triplet pairing survives. In this latter example both the time-reversal and the gauge symmetry due to SC order are spontaneously broken, which made UGe$_2$, as well as its cousins URhGe and UCoGe an exciting avenue for theoretical and experimental research.

For spin-triplet pairing, Anderson noticed that inversion symmetry is required to obtain a pair of degenerate states $c^{\uparrow}_{k}(0)$ and $c^{\downarrow}_{k}(0)$ capable of forming a Cooper pair. It was therefore surprising that superconductivity was discovered in the heavy fermion compound CePt$_3$Si which lacks inversion symmetry. It soon became clear however that in the case of a non-centrosymmetric crystal, the spin-orbit coupling (SOC) mixes different spin states, so that the division into triplet and singlet symmetry of the SC order parameter becomes meaningless. A bulk of theoretical work exists that has provided a symmetry-based phenomenology to explain this in detail. The symmetry of the superconducting (SC) gap in this and other unconventional superconductors is presently a matter of intense investigation.

An intriguing question is what happens if time-reversal symmetry is broken in a crystal that lacks a centre of inversion. Can such a material become a superconductor? This question was answered affirmatively when superconductivity was discovered in the non-centrosymmetric ferromagnetic compound UIr under pressure. The symmetry of the SC order parameter and its connection to FM nevertheless remains unclear, which motivates the present study. Spontaneous symmetry breaking in condensed matter systems is conceptually of immense importance, as it may provide clues for what could be expected in systems belonging to vastly different areas of physics. The study of a condensed-matter system such as UIr with multiple broken symmetries is likely to have impact on a number of disciplines of physics, including such disparate phenomena as mass differences between elementary particles and extremely dilute ultra-cold atomic gases.

In this work, we study a model system of a non-centrosymmetric superconductor with substantial spin-orbit coupling, which at the same time exhibits itinerant ferromagnetism. The origin of the SOC may be either that the crystal structure lacks a center of inversion, such as in UIr, or due to a thin-film geometry where the breakdown of inversion symmetry near the surface induces transverse electrical fields, leading to the well-known Rashba SOC. Our model should therefore be relevant both to the non-centrosymmetric and centrosymmetric heavy fermion compounds, since the SOC is considerable in any case due to the high atomic number. Specifically, materials that exhibit coexistence of SC and FM order and where SOC is large include UGe$_2$, URhGe, UCoGe, and UIr. For this model, we construct a mean-field theory, solve the saddle point equations for the order parameters and study the effect of spin-orbit coupling on the superconducting order parameters. Finally, we discuss application of this model to the case of UIr.

To label the SOC+FM split bands, it is possible to introduce a pseudospin basis in which the normal-state Hamiltonian is diagonalized. In the original spin basis, the SC matrix order parameter is characterized, in analogy to the $p$-wave state, by a vector $\mathbf{d}_k$ and scalar $\Delta_s$ so that $\Delta_{\alpha\beta}(k) = i\mathbf{d}_k \cdot \hat{\sigma} \delta_{\alpha\beta} + i[(\mathbf{d}_k \cdot \hat{\sigma})\delta_{\alpha\beta}]$. Note that, unlike the usual $p$-wave SC, a singlet component $\Delta_s$ of the gap will also be present since antisymmetric SOC in general mixes the parity of the order parameter. Below, $\hat{\sigma}$ is used for $2 \times 2$ matrices. We now proceed to write down the effective Hamiltonian $H = H_N + H_{SC}$ for our system. In the normal state, the Hamiltonian in momentum-space read is

$$H_N = H_0 + \sum_{k\alpha\beta} [c^\dagger_{k\alpha} (\varepsilon_k \hat{1} - \hbar \hat{\sigma}_z + \mathbf{g}_k \cdot \hat{\sigma}) c_{k\beta}],$$

where $H_0 = INM^2/2$. Above, the dispersion relation $\varepsilon_k$ is measured from chemical potential $\mu$, and the magnetization $M = |\mathbf{M}|$ is taken along the easy-axis, while $\hbar = IM$ is
the exchange splitting of the bands and \( g_k \) is the SOC vector. When superconductivity coexists with FM, the SC pairing is generally believed to be non-unitary, characterized by \( d_k \times d_k^\dagger \neq 0 \). In such a scenario, the SC order parameter couples to the spontaneous magnetization \( M \) through a term \( \gamma M \cdot d_k \times d_k^\dagger \) in the free energy, where the sign of \( \gamma \) is determined by the gradient of the DOS at Fermi level and \( \langle S_k \rangle = i d_k \times d_k^\dagger \) is the spin associated with the Cooper pair. Thus, for \( \gamma < 0 \) it is expected that a SC pairing state obeying \( i d_k \times d_k^\dagger \parallel M \) is energetically favored, implying that \( d_k \) must be complex-valued. Our model captures broken time-reversal symmetry in addition to antisymmetric SOC. As shown by Anderson, the presence of the latter is detrimental to spin-triplet SC pairing state, unless \( d_k \parallel g_k \). In our case, it is obvious that a non-unitary SC pairing state cannot satisfy this condition since \( d_k \) is complex, whereas \( g_k \) must be real for the Hamiltonian to be hermitian.

The SOC vector reads \( g_k = -g_{-k} \), and we introduce \( g_{k,x} = -i g_{k,y} \) for later use. We consider the SOC in the Rashba form, namely \( g_k = \lambda (\varepsilon_{p_x} - e_{p_y}) \). This corresponds to a situation where an asymmetric potential gradient is present along the \( z \)-axis, and is also the scenario realized in non-centrosymmetric CePt\(_3\)Si\(_2\). We have introduced fermion operators \( \{c_{k,\sigma} \} \) in a basis \( \varphi_k = |c_k,\sigma\rangle \rangle^2 \).

Diagonalizing the normal-state Hamiltonian yields the quasiparticle excitations \( E_k = \varepsilon_k - \sigma \sqrt{\hbar^2 + \lambda^2 k^2} \), which due to the SOC are characterized by the pseudospin \( \sigma = \pm 1 \). For later use, we define \( N_k = |1 + \lambda^2 k^2/(\hbar^2 + \lambda^2 k^2)|^{-1/2} \). The superconducting pairing is now assumed to occur between the excitations described by \( \varphi_k \). Due to the presence of antisymmetric spin-orbit coupling, this automatically leads to a mixed-parity SC state in the original spin basis. To see this, we introduce

\[
H_{\text{SC}} = \frac{1}{2N} \sum_{kk'\sigma} V^{kk'}_{\sigma} c_{k\sigma}^\dagger c_{k'\sigma}^\dagger e^{-i\mathbf{k}\cdot\mathbf{k'}} c_{-k'\sigma} c_{k\sigma},
\]

and perform a standard mean-field decoupling, which after an additional diagonalization yields the total Hamiltonian in the superconducting state:

\[
H = H_0 + \sum_{k\sigma} (E_k - E_{k'} - \Delta_{k\sigma} \lambda_{k\sigma}^\dagger 2n_{k\sigma} n_{k\sigma}^\dagger)/2, \quad E_k = (E_{k\sigma} + |\Delta_{k\sigma}|^2)^{1/2}
\]

and \( \{n_{k\sigma}, \lambda_{k\sigma} \} \) are fermion operators. The merit of this procedure is that we can now obtain simple self-consistency equations for the gaps \( \Delta_{k\sigma} \), which may then be transformed back to the gaps in the original spin-basis \( \varphi_k \) by means of the unitary transformation \( R_k \). We assume a chiral p-wave symmetry for the gaps with a corresponding pairing potential \( V^{kk'\sigma} = -g_{\sigma \omega} e^{i\sigma\phi} \delta(k'k) \), where \( \omega = k_x/k_y \). The motivation for this is that this choice is consistent with the condition \( d_k \parallel g_k \) is satisfied exactly for \( h \to 0 \), and corresponds to a fully gapped Fermi surface which favors the condensation energy. The gaps obtain the form \( \Delta_{k\sigma} = -\sigma \Delta_{k\sigma} e^{i\sigma\phi} \) and we find a self-consistency equation of the standard BCS form with a cutoff \( \omega \) on the pairing-fluctuation spectrum which we do not specify further. Moreover, \( N^\sigma(\varepsilon) \) is the pseudospin-resolved density of states (DOS) for the \( E_k \) \((\sigma = \pm)\) bands of the quasiparticle excitations. Introducing the total DOS at the Fermi level for a normal metal \( N_0 = n V/\sqrt{2m\mu} \), we define the energy gaps as

\[
\Delta_k = \sqrt{\langle N^\sigma(\varepsilon) \rangle (\varepsilon^2 + \Delta_{k\sigma}^2)^{1/2}}.
\]

and defining \( c = g N_0/2 \), the analytical solution for the gaps \( \Delta_{k,0} = 2 e\exp[-1/\langle c R_0(\varepsilon) \rangle] \), \( R_0(\varepsilon) = 2 N^\sigma(\varepsilon)/N_0 \). With the analytical solution for \( \Delta_{k,0} \) in hand, we may exploit the unitary transformation \( R_k \) to express the superconducting gaps in the original spin basis as follows:

\[
\Delta_k = -e^{i\sigma\phi} [\Delta_{k,0}(\lambda_{k\sigma}^\dagger \lambda_{k\sigma})^2 + \Delta_{k,0}(\lambda_{k\sigma}^\dagger \lambda_{k\sigma})^2 \lambda^{i\sigma\phi}],
\]

where we have defined \( \lambda_{k\sigma} = N_{k\sigma} = N_{k\sigma} \sigma(0) \) and \( \Delta_{k\sigma} = |h + \sqrt{\hbar^2 + \lambda^2 k_x^2(0)}|^{-1} \). Note that in the original spin basis, the superconducting order parameter is in general a mixture of triplet \( \langle \Delta_{k\sigma} \rangle \) and singlet \( \langle \Delta_{k1} \rangle \) components. The self-consistency equation for the magnetization is:

\[
h + \frac{i}{\tilde{I}} \sum_{\sigma} \int \frac{\sigma dR(\varepsilon) e\varepsilon}{\sqrt{[\hbar^2 + \lambda^2 k_x^2(\varepsilon)](\varepsilon^2 + \Delta_{k,0}^2)}} = 0,
\]

where the integration is over the bandwidth and \( \tilde{I} = I N_0 \). Eqs. (5) are the main analytical results of this work.

Let us briefly investigate some important limiting cases of Eq. (3). In the absence of spin-orbit coupling \((\lambda \to 0)\), one finds \( N_{k\sigma} \to 1 \) and \( \Delta_{k\sigma} = \Delta_{k\sigma} \), while \( \Delta_{k11} \to 0 \), such that we reproduce the results of Refs.\(^{2,23}\). In the absence of exchange energy \((h \to 0)\), one finds that \( \Delta_{k\sigma} \to 1/\sqrt{2} \) and \( \Delta_{k11} = -e^{i\sigma\phi} (\Delta_{1,0} + \Delta_{1,0})/2 \), \( \Delta_{k1} = e^{-i\sigma\phi} (\Delta_{1,0} + \Delta_{1,0})/2 \), and \( \Delta_{k11} = (\Delta_{1,0} - \Delta_{1,0})/2 \). As demanded by consistency, the triplet gaps are equal in magnitude since there is no exchange field and the singlet component is nonzero since \( \Delta_{1,0} \neq \Delta_{1,0} \) in general. Finally, Eq. (4) reproduces the well-known Stoner criterion \( \tilde{I} \geq 1 \) for the onset of FM in the absence of SOC and SC \((\lambda \to 0, g \to 0)\).

We now focus on the general case in which \( h \neq 0 \) and \( \lambda \neq 0 \). First of all, we must specify the range of the pa-
rameters in the problem that corresponds to a physically realis-
cenario. We allow \( h \) to range, in principle, from 0 to \( \mu \), the latter denoting a fully polarized ferromagnet. As a
convenient measure of the strength of SOC, we introduce
the dimensionless quantity \( \alpha_{soc} \equiv \sqrt{2\lambda^2m/\mu} \) which has a
direct physical interpretation: namely, it is the ratio of the
SOC (at \( E_F \)) to the Fermi energy \( \mu \). The parameter \( \alpha_{soc} \) is
allowed to vary from 0 to \( \delta \), where \( \delta \) denotes a fraction of
the Fermi energy. We take \( \delta = 0.5 \) as a sensible upper limit.
Note that generically, the SOC strength at the Fermi level is
different for the two quasiparticle bands, and moreover de-
pends on \( h \). For a given value of \( h \), one may derive that
\( \lambda \leq \delta \mu/[2\mu m + \sqrt{2m^2(h^2 + \delta^2\mu^2)}]^{1/2} \) ensures that the spin-
orbit energy is less than \( \delta \times \mu \) for both quasiparticle bands.

In Fig. 1b-d, we present the self-consistent solutions for the
order parameters in Eqs. (3) and (4) as a function of the
FM exchange parameter \( I \) for several values of \( \alpha_{soc} \). We have
defined \( \Delta_1 = |\Delta_k\sigma| \) and \( \Delta_{11} = |\Delta_k| \), and fixed \( \omega/\mu =
0.01 \) and \( m/\mu = 5 \times 10^4 \) with \( \epsilon = 0.2 \), which are standard
choices. For \( \alpha_{soc} = 0 \), the onset of FM occurs at \( I = 1.0 \)
which lifts the degeneracy of \( \Delta \) and \( \Delta_1 \), while \( \Delta_{11} \) is always
zero. Upon increasing \( \alpha_{soc} \), it is interesting to note that the
PM-FM transition occurs at lower values of \( \tilde{I} \), indicating that
spin-orbit coupling favors ferromagnetic ordering. For \( \alpha_{soc} \neq
0 \), it is seen that \( \Delta_{11} \) is also non-zero, although it becomes
suppressed at the onset of ferromagnetism. A common feature
for all gaps is that they increase with \( \alpha_{soc} \) in the absence of
ferromagnetism and deep inside the ferromagnetic phase \( \tilde{I} \geq
1.02 \). In the intermediate regime, there are crossovers between
the gaps for different values of \( \alpha_{soc} \) due to the different onsets
of ferromagnetic order. By comparing the behaviour between
the gaps for increasing \( I \) with \( \alpha_{soc} \neq 0 \), one infers that \( \Delta_1 \) and
\( \Delta_{11} \) eventually saturate at a constant non-zero value, while
\( \Delta_1 \) continues to increase steadily. This is quite different from
the case when \( \alpha_{soc} = 0 \), where the minority spin-gap goes
to zero rapidly with increasing \( \tilde{I} \). This seems to suggest that
the presence of spin-orbit coupling in the system ensures the
survival of the minority-spin gap \( \Delta_1 \) and the singlet gap \( \Delta_{11} \)
even though the FM exchange energy becomes strong.

In Fig. 1a and 1f, we plot the ratio of the singlet and triplet
gaps, defined as \( R_\Delta = \Delta_{11}/(\Delta_1 + \Delta_1) \), and the maximal
critical temperature \( T_{\text{c, max}} \) for the onset of superconductivity.
It is seen from the left panel that \( R_\Delta \) increases with \( \alpha_{soc} \)
in the PM regime, suggesting that the singlet component be-
comes more prominent in the system as compared to the triplet
gaps. However, at the onset of FM order, \( R_\Delta \) decreases since
the singlet component becomes suppressed by the Zeeman-
splitting. In the right panel, one observes that \( T_{\text{c, max}} \) in-
creases both with \( \alpha_{soc} \) and \( \tilde{I} \). Our findings suggest that the
presence of antisymmetric SOC, originating from e.g. non-
centrosymmetricity of the crystal structure, enhances both the
tendency towards ferromagnetism and the magnitude of the
SC gaps in all spin channels. In the absence of spin-orbit cou-
lpling, it was shown in Ref. 22 that the simultaneous coexistence
of FM and non-unitary triplet superconductivity is the ther-
modynamically favored state as compared to the pure normal,
FM, or SC state. Since the presence of spin-orbit coupling is
seen to enhance both the FM and SC order parameters, it is
reasonable to expect that the coexistent state is still thermody-
namically the most favorable one even when \( \alpha_{soc} \neq 0 \).

Out of the known non-centrosymmetric superconductors,
UIr is the only compound that is also a ferromagnet. This ma-
terial, which is ferromagnetic at ambient pressure, develops
superconductivity in a narrow pressure region around \( P \sim
2.6 \text{ GPa} \) right next to the FM–PM quantum phase transition,
with a maximum SC transition temperature \( T_{\text{SC}} \sim 0.14 \text{ K} \).
At this pressure, the saturated magnetic moment was mea-
sured to be \( 0.07\mu_B \) per U atom, and such a small value clearly
indicates the itinerant character of the ferromagnetism, pre-
sumably due to \( 5f \) electrons of uranium. UIr crystallizes in
the monoclinic structure (space group \( F2_1 \)) which lacks in-
version symmetry, and the FM moment is Ising-like, oriented
along the [101] direction in the (ac)-plane.

Given the proximity of the SC state in UIr to the PM tran-
sition, one may probably consider the magnetization \( h \) as a
perturbation on top of the SOC-split bands. Neglecting the
effect of the former, it is known\textsuperscript{12} that even in the case of
non-centrosymmetric superconductors (and \( h = 0 \), the band
energies still satisfy the relation \( \varepsilon_{\sigma}(\mathbf{k}) = \varepsilon_{\sigma}(-\mathbf{k}) \) due to the
time reversal symmetry of the single-electron Hamiltonian.
As a consequence, the SC order parameter on the \( \beta \)-th sheet
of the Fermi surface transforms according to one of the irre-
ducible representations of the normal state point group. In
the case of UIr, the point group \( C_2 \) has two one-dimensional irre-
ducible representations, denoted A and B. Then the SC order
parameter is an odd function \( \Delta(\mathbf{k}) = -\Delta(\mathbf{k}) \) given by\textsuperscript{22}
\[ \Delta_{A,B}(\mathbf{k}) \propto \tau(\mathbf{k}) \phi_{A,B}^{\mathbf{k}}(\mathbf{k}), \]
where \( \tau(\mathbf{k}) \) is an odd phase fac-
tor\textsuperscript{28} and the basis functions \( \phi_{A,B} \) are even in \( \mathbf{k} \). Denoting
the rotation axis of the \( C_2 \) group as \( z \) (this actually corresponds to
\( b \)-axis in case of UIr), the even functions \( \phi_{A} \) and \( \phi_{B} \) can then be
cast in the following form: \( \phi_A(\mathbf{k}) = (k_x^2 + C) u_1(\mathbf{k}) \), and
\( \phi_B(\mathbf{k}) = k_z(k_z u_2(\mathbf{k}) + k_y u_3(\mathbf{k})) \), where \( C \) is some con-
stant and \( \{u_i(\mathbf{k})\} \) are arbitrary even functions of \( k_x, k_y, k_z \).
Function \( \phi_A \) generically has no nodes, whereas \( \phi_B \) has two
point nodes at the poles \( (k_x = k_y = 0) \) and a line of nodes at the
equator. The symmetry argument does not allow one to de-
termine which pairing channel is realized, however the experi-
mental observation of the strong pair-breaking effect due to
disorder\textsuperscript{12} indicates that the gap must be anisotropic, possi-
bly favouring the gap with the nodes such as \( \Delta_B(\mathbf{k}) \).

One way of experimentally probing the symmetry of the
superconducting order parameter in UIr would be by means of
transport properties such as Josephson tunneling or point-
contact spectroscopy. In particular, it has recently been shown
that the presence of multiple gaps in superconductors with
broken inversion symmetry should manifest itself through
clear signatures at bias voltages corresponding to the sum and
difference of the singlet and triplet components\textsuperscript{22,25,26}. We ex-
pect similar behavior in the present case, at least when the
ferromagnetism is weak, and point-contact spectroscopy data
could then be compared with the predictions for \( R_\Delta \) in Fig.
[1][4]. Alternatively, it should be possible to directly probe the
spin-texture of the superconducting order parameter by study-
ing the effect of an externally applied magnetic field when the
paramagnetic limitation dominates, e.g. in a thin-film struc-
true, where the orbital mechanism of destroying superconduc-
tivity is suppressed.29

In summary, we have developed a mean-field model for a superconductor lacking inversion symmetry and displaying itinerant ferromagnetism. Specifically, we have investigated the interplay between ferromagnetism and asymmetric spin-orbit coupling and how these affect superconducting order, which in general is a mixture of a singlet and triplet components. Our main results are the analytical expression Eqs. (3) and (4) and the belonging discussion. We find that spin-orbit coupling may enhance superconductivity in both the singlet and triplet channels in addition to favoring the Stoner criterion for the ferromagnetic instability. We have applied these considerations to the heavy fermion superconductor UIr, together with group-theoretical analysis of the symmetry of the SC order parameter.

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28 The nontrivial phase factor \( t(k) = -t(-k) \) is defined such that \( T[k] = t(k) - k \), where \( T \) is the time-reversal operator.
29 Experimentally, the measurements on a single crystal of UIr so far indicate that the orbital effects of the magnetic field dominate.