Spin susceptibility in superconductors without inversion symmetry

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Abstract. In materials without spatial inversion symmetry, the spin degeneracy of the conduction electrons can be lifted by an antisymmetric spin–orbit coupling. We discuss the influence of this spin–orbit coupling on the spin susceptibility of such superconductors, with a particular emphasis on the recently discovered heavy Fermion superconductor CePt₃Si. We find that, for this compound (with tetragonal crystal symmetry) irrespective of the pairing symmetry, the stable superconducting phases would give a very weak change of the spin susceptibility for fields along the c-axis and an intermediate reduction for fields in the basal plane. We also comment on the consequences for the paramagnetic limiting in this material.

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1. Introduction

Our understanding of conventional superconductors is based on the BCS theory of Cooper pairing induced by electron–phonon interaction. The electrons pair up in the superconducting phase with the most symmetric pair wave function possible, i.e. in the s-wave orbital and spin-singlet channel. According to Anderson (1959) this type of pairing requires time reversal symmetry, because the paired electrons need to be in degenerate states of opposite momentum and opposite spin [1]. This theorem guarantees that the conventional superconducting phase remains stable even if a sample is dirty as long as time reversal symmetry is present, and allows us to construct degenerate electron pairs with vanishing total momentum.

The situation becomes more complex for unconventional superconductivity with Cooper pairs of lower symmetry. Impurities of all kind are detrimental to such pairing due to the momentum averaging effect of random potential scattering. For clean samples, new symmetry criteria have been formulated by Anderson (1984) [2]. While spin-singlet pairing only requires time reversal symmetry, spin-triplet pairing also requires inversion symmetry. These symmetries are present in most materials and no discussion on this issue is usually required.

Motivated by the recent discovery of the heavy fermion superconductor CePt$_3$Si which has no inversion centre, we will discuss the magnetic properties of the superconducting phase [3]. While the absence of an inversion centre does not immediately imply an unusual behaviour of the superconducting phase, special interest in this case has arisen with the observation that the upper critical field dramatically exceeds the paramagnetic limit. For heavy fermion superconductors the coherence length is generally rather short, roughly 100 Å, giving rise to large orbital depairing fields. Thus, if there were a paramagnetic limiting effect, then it would be likely be observed; as for example in CeCoIn$_5$ [4]. CePt$_3$Si has a critical temperature of $T_c = 0.75$ K and a zero-temperature extrapolated upper critical field of 5 T. The estimated paramagnetic limit is a factor 5 smaller than $H_{c2}$ ($H_p \approx \Delta / \sqrt{2} \mu_B \approx 1$ T) [3]. Paramagnetic limiting is expected for spin-singlet pairing, as the Zeeman coupling to the spins would break up the Cooper pairs. Since Cooper pairs in spin-triplet configurations would not be destroyed by spin polarization, it might be concluded that spin-triplet pairing is realized in CePt$_3$Si. However, the absence of an inversion centre appears to be an obstacle for spin-triplet pairing.

It has been shown that broken inversion symmetry is not indiscriminately destructive for the spin-triplet pairing and, additionally, it softens the effect of the paramagnetic depairing for spin-singlet pairs [5]. For many types of crystal lattices, the absence of an inversion centre introduces an antisymmetric spin–orbit coupling (SOC), analogous to the well-known Rashba type of spin–orbit coupling. Naturally, this influences the Cooper pairing through the modification of the band structure as shown by various groups [5]–[7].

We would like to discuss here the problem of the spin susceptibility of the superconducting phase, since this is also directly connected with the issue of paramagnetic limiting. The spin susceptibility measured by means of the NMR-Knight shift in the superconducting phase is often used to distinguish between spin-singlet and spin-triplet pairing. We will show here that this kind of discrimination between the two types of states is no longer generally possible. Rigorously speaking, it is, of course, not possible to separate spin-singlet and triplet pairing in the presence of SOC. We will consider here weak SOC which we can turn on adiabatically to follow the evolution of the originally well-defined singlet and triplet pairing states. Our basic result is that the spin susceptibility of the spin-singlet states gradually approaches the behaviour of the spin-triplet state which survives the presence of the spin–orbit coupling. Moreover, we can predict that the
spin susceptibility for fields along the $z$-axis of the tetragonal crystal lattice of CePt$_3$Si would be less suppressed than for fields along the basal plane.

2. The basic model and normal state Green's function

For the following discussion we will use the model introduced by Frigeri et al [5] for CePt$_3$Si, which has the following single-particle Hamiltonian:

$$H_0 = \sum_{k,s,s'} [\xi_k \sigma_0 + \alpha \mathbf{g}_k \cdot \sigma] c_{k,s}^\dagger c_{k,s'},$$

where $c_{k,s}^\dagger$ ($c_{k,s}$) creates (annihilates) an electron with momentum $k$ and spin $s$. The band energy $\xi_k = \epsilon_k - \mu$ is measured relative to the chemical potential $\mu$ and $\alpha \mathbf{g}_k \cdot \sigma$ introduces the antisymmetric spin–orbit coupling with $\alpha$ as a coupling constant (we set $\langle g_k^2 \rangle_k = 1$ where $\langle \rangle$ denotes an average over the Fermi surface).

We give here a brief discussion on the origin and some basic properties of the antisymmetric SOC. In a crystal lattice, the electrons move in a periodic potential $U(r)$. In the absence of inversion symmetry there is no symmetry point in the unit cell relative to which $U(r) = U(-r)$ is satisfied. This also implies that the Bloch function does not have the property that $u_{k}(r) = u_{-k}(-r)$. Ignoring for the moment relativistic effects, the potential yields the following contribution to the single-particle Hamiltonian:

$$H_p = \sum_{k,s} \int_{u.c.} d^3r \ u_k^*(r) U(r) u_k(r) c_{k,s}^\dagger c_{k,s} = \sum_{k,s} \tilde{U}(k) c_{k,s}^\dagger c_{k,s}. $$

The integral runs over the unit cell of the lattice. The resulting potential $\tilde{U}(k)$ is even in $k$, i.e. $\tilde{U}(k) = \tilde{U}(-k)$. On this (non-relativistic) level, the lack of inversion symmetry does not affect the band structure which remains symmetric under the operation $k \rightarrow -k$ due to time reversal symmetry, i.e. the fact that $u_{k}^*(r) = u_{-k}(r)$.

Now we include the SOC. The SOC of each ion couples different atomic orbitals and requires a multi-band description. For a lattice without inversion symmetry, SOC already appears on the level of a single-band model yielding the second term in equation (1) [8]. The vector function $\alpha \mathbf{g}_k$ is derived from the relativistic correction ($e/2mc^2)[\mathbf{v} \times \nabla_r U(r)] \cdot \mathbf{S}$, which yields

$$\alpha \mathbf{g}_k = -\frac{e}{2mc^2}\int_{u.c.} d^3r \{ J_k(r) \times \nabla_r U(r) \}$$

with

$$J_k(r) = \frac{\hbar}{2m_1} [u_k^*(r) (i\mathbf{k} + \nabla_r) u_k(r) + u_k(r) (i\mathbf{k} - \nabla_r) u_k^*(r)].$$

It is easy to verify that $\alpha g_k = 0$, if $U(r) = U(-r)$ and $u_k(-r) = u_{-k}(r)$. In the absence of inversion symmetry, however, $g_k$ is finite and satisfies $g_k = -g_{-k}$, since $J_{-k}(r) = -J_k(r)$ (note if $g_k = g_{-k}$
The interaction satisfies the relations

\[ \mathcal{I}[\alpha g_k \cdot \hat{\sigma}] \mathcal{I}^{-1} = -\alpha g_k \cdot \hat{\sigma} \quad \text{and} \quad \mathcal{T}[\alpha g_k \cdot \hat{\sigma}] \mathcal{T}^{-1} = \alpha g_k \cdot \hat{\sigma}. \]

SOC yields a modified band structure. We parametrize the Green’s function by

\[ \hat{G}_0 (k, i\omega_n) = G^0_x(k, i\omega_n) \hat{\sigma}_0 + (\hat{g}_k \cdot \hat{\sigma}) G^0_{-}(k, i\omega_n), \]

where

\[ G_{\pm}(k, i\omega_n) = \frac{1}{2} \left[ \frac{1}{i\omega_n - \xi_k - \alpha|g_k|} \pm \frac{1}{i\omega_n - \xi_k + \alpha|g_k|} \right] \]

and \( \hat{g}_k = g_k / |g_k| \) (\(|g| = \sqrt{g^2} \)). The band splits into two spin-dependent parts with energies \( E_{k,\pm} = \xi_k \pm \alpha|g_k| \). The spinor is twisted on the two bands in a way that is described by the antisymmetric part of the Green’s function, \((\hat{g}_k \cdot \hat{\sigma}) G^0_{\pm}(k, i\omega_n)\).

3. Superconducting phase

Now we turn to the superconducting phase and introduce the general pairing interaction

\[ \mathcal{H}_{\text{pair}} = \frac{1}{2} \sum_{k,k',s_i} V_{ss_1s_2s_3s_4}(k, k') c^\dagger_{k,s_1} c_{-k',s_2} c_{-k,s_3} c_{k',s_4}. \]

The interaction satisfies the relations \( V_{\alpha\beta\gamma\delta}(k, k') = -V_{\beta\alpha\gamma\delta}(-k, k') = -V_{\alpha\beta\delta\gamma}(k, -k') = V_{\delta\gamma\alpha\beta}(k', k) \) to assure that the Fermion sign and the time-reversal symmetry (\( \mathcal{T} \)) are preserved.

For the following calculations it is advantageous to discuss the superconducting phase by means of Green’s functions

\[ G_{\lambda\mu}(k, \tau) = -\langle T_\tau [c_{k,\lambda}(\tau) c_{k',\mu}^\dagger(0)] \rangle, \]

\[ F_{\lambda\mu}(k, \tau) = \langle T_\tau [c_{k,\lambda}(\tau) c_{-k',\mu}(0)] \rangle, \]

\[ F_{\lambda\mu}^\dagger(k, \tau) = \langle T_\tau [c_{-k',\lambda}(\tau) c_{k',\mu}^\dagger(0)] \rangle, \]

where the operators \( c_{k,\lambda}(\tau) \) and \( c_{k,\lambda}^\dagger(\tau) \) are expressed in the Heisenberg representation. These Green’s functions have to satisfy Gor’kov equations of the following form:

\[ (\hat{G}_0(k, i\omega_n) + \hat{\Delta}(k) \hat{G}_0^\dagger(-k, -i\omega_n)) \hat{G}(k, i\omega_n) = \hat{\sigma}_0, \]

\[ \hat{F}(k, i\omega_n) = \hat{G}_0(k, i\omega_n) \hat{\Delta}(k) \hat{G}^\dagger(-k, -i\omega_n), \]

\[ \hat{F}^\dagger(k, i\omega_n) = \hat{G}_0^\dagger(-k, -i\omega_n) \hat{\Delta}^\dagger(k) \hat{G}(k, i\omega_n), \]

where \( \hat{\Delta}^\dagger(k) \) is the gap function defining the order-parameter of the superconducting state and is a \((2 \times 2)\)-matrix in spin space.

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In general, the gap function has a singlet \( \psi(k) \) and a triplet component \( d(k) \), i.e. \( \Delta^\uparrow(k) = i(\psi(k) + d(k) \cdot \hat{\sigma})\hat{\sigma}_y \), where \( \psi(k) = \psi(-k) \) is even and \( d(k) = -d(-k) \) is odd. For a finite \( \alpha \), the gap equations for the spin-singlet and triplet channel are coupled [5, 6] and thus the gap function is a mixture of both channels. However, this coupling is of the order \( \alpha/\epsilon_F \) which we take to be small (\( \epsilon_F \) is the Fermi energy, or analogue to the band width). Consequently, we ignore this coupling and consider the singlet and triplet channels separately. The coupling of the singlet and triplet channels manifests itself in two ways. It gives rise to different magnitude gaps on the two SOC split Fermi surface sheets. It should also be observable through a non-vanishing spin-current contribution in the excess current associated with Andreev scattering.

It has been shown that once \( \alpha \gg k_BT_c \) for the spin-triplet channel, then the only spin-triplet state that is permitted satisfies \( d(k)||g(k) \) [5]. We assume that \( k_BT_c < \alpha < \epsilon_F \) and solve in sequence the equations (8)–(10). The solutions are formally the same for both singlet \( \Delta(k) = i(\psi(k))\hat{\sigma}_y \) pairing and triplet \( \Delta(k) = i[d(k) \cdot \hat{\sigma}]\hat{\sigma}_y \) with \( d(k)||g(k) \) pairing. We find

\[
\tilde{G}(k, i\omega_n) = G_+(k, \omega_n)\hat{\sigma}_0 + (\tilde{g}_k \cdot \hat{\sigma})G_-(k, i\omega_n),
\]

\[
G_{\pm}(i\omega_n) = -\frac{1}{2} \left[ \frac{i\omega + E_+}{(\omega_n^2 + |\Delta|^2 + E_+^2)} \pm \frac{i\omega + E_-}{(\omega_n^2 + |\Delta|^2 + E_-^2)} \right],
\]  

(11)

where \( E_{\pm} = \xi \pm \alpha|g| \), and

\[
\tilde{F}(k, i\omega_n) = [F_+(k, i\omega_n)\hat{\sigma}_0 + (\tilde{g}_k \cdot \hat{\sigma})F_-(k, i\omega_n)]\Delta(k),
\]

\[
\tilde{F}^\dagger(k, i\omega_n) = \Delta^\dagger(k)[F_+(k, i\omega_n)\hat{\sigma}_0 + (\tilde{g}_k \cdot \hat{\sigma})F_-(k, i\omega_n)],
\]

(12)

\[
F_{\pm}(k, i\omega_n) = \frac{1}{2} \left[ \frac{1}{(\omega_n^2 + |\Delta|^2 + E_+^2)} \pm \frac{1}{(\omega_n^2 + |\Delta|^2 + E_-^2)} \right].
\]

Note that the anomalous Green’s function has less symmetry than the gap function. In particular, \( \Delta(k) \) is either symmetric (singlet) or antisymmetric (triplet) with respect to \( k \rightarrow -k \), while the resulting anomalous Green’s function has both a symmetric and an antisymmetric component. This leads to the mixing of the spin-singlet and spin-triplet Cooper pairs and has been discussed by Gor’kov and Rashba for the case of 2D metals [6]. However, \( F_- \) is an odd function in \( \xi \) and does not contribute to the gap equation in its weak coupling formulation. In this way, the magnitude of the gap \( \psi = \psi(k) \) for the singlet s-wave order parameter can be approximated by the standard and universal BCS gap equation,

\[
\ln(\psi) = -\int_{-\infty}^{\infty} dx \frac{1}{\sqrt{\psi^2 + x^2}} \frac{1}{\exp[(\pi/\gamma) (\sqrt{\psi^2 + x^2}/k_BT)] + 1},
\]

(13)

where \( T \) is expressed in units of \( T_c \), \( \psi \) in units of \( \psi(T = 0) = \psi(T = 0)/k_BT_c = \pi/\gamma \) and \( C = \ln(\gamma) = 0.577 \) corresponds to the Euler constant. The deviation will be of the order \( (\alpha/\epsilon_F)^2 \), with \( \epsilon_F \) the cut-off energy of the attractive interaction, which is small in our case. For the protected spin-triplet state \( d(k) = \Delta g(k) \) we have \( \Delta_g(T = 0)/k_BT_c \sim \psi(T = 0)/k_BT_c \) and to avoid numerical complication superfluous for the following discussion, we have set \( \Delta_0(T/T_c) = \psi(T/T_c) \).
4. The static uniform spin susceptibility

In materials with a spatial inversion centre, the measurement of the Knight shift in the resonance frequency ($\delta \omega$) by nuclear magnetic resonance (NMR) is an important experimental tool in determining the nature of the superconducting state. In particular, it allows the determination of the spin structure of the Cooper pairs. The measurement of the temperature dependence of the ratio $\frac{\delta \omega_s}{\delta \omega_n} = \frac{\chi_s}{\chi_n}$ is a direct measure of the behaviour of the spin susceptibility of the superconducting state ($\chi_s$) ($\chi_n$ is the spin-susceptibility of the normal state). For spin-singlet superconductors it is known that the paramagnetic susceptibility is proportional to the density of normal electrons, which vanishes at zero temperature. In the spin-triplet case, the spin of the Cooper pairs can contribute to the susceptibility. In particular, if the external field ($H$) is parallel to the spin of the Cooper pair ($H \perp d$), then the susceptibility coincides with that of the Fermi-liquid normal state. This property was used to experimentally confirm spin-triplet superconductivity, for example, in Sr$_2$RuO$_4$ [9]

In principle, for materials with strong spin–orbit coupling, the total magnetic susceptibility cannot be split into separate orbital and spin parts. However, if $\alpha \ll \epsilon_F$ it is possible to isolate the two components [10]. As shown in [11], the spin susceptibility tensor $\chi_{ij}^s$ in the superconducting state can be expressed as

$$\chi_{ij}^s = -\mu_B^2 k_B T \sum_k \sum_{\omega_n} \text{tr} [\hat{\sigma}_i \hat{G}(k, \omega_n) \hat{\sigma}_j \hat{G}(k, \omega_n) - \hat{\sigma}_i \hat{\tilde{F}}(k, \omega_n) \hat{\sigma}_j ^\top \hat{\tilde{F}}^\dagger(k, \omega_n)].$$  (14)

If we assume a spherical Fermi surface and a constant density of states $N(\xi)$ close to the Fermi surface, then we can replace the sum over $k$ by $\sum_k \rightarrow N(0) \int \frac{d\Omega}{4\pi} \int d\xi$. In the normal state, the integral over $d\xi$ cannot be carried out before the sum over $\omega_n$ because the regular Green’s function will be formally divergent [12]. Doing the sum first, we find that the normal state spin susceptibility corresponds to the Pauli susceptibility $\chi_n = 2\mu_B^2 N(0)$. This would not be the case if electron–hole asymmetry is taken into account (i.e. if $dN(\xi)/d\xi|_{\xi=0} \neq 0$). For our purposes this is an unnecessary complication which we will neglect here.

To avoid carrying out the summation over $\omega_n$ in the superconducting state, we follow Abrikosov et al and sum and subtract the expression corresponding to the normal state [12]. The integral of the difference between the integrands rapidly converge in this case. Consequently, the order of summation and integration can be interchanged. For a singlet gap function we find a generalization of the result obtained by Gork’ov and Rashba [6] and Bulaevskii et al [13]:

$$\chi_{ii}^s = \chi_n \left\{ 1 - k_B T \pi \sum_{\omega_n} \left[ \frac{1 - \hat{g}_{k,i}^2}{\omega_n^2 + |\psi(k)|^2 + \alpha^2 |g_k|^2} \frac{|\psi(k)|^2}{\sqrt{\omega_n^2 + |\psi(k)|^2}} + \hat{g}_{k,i}^2 \frac{|\psi(k)|^2}{\omega_n^2 + |\psi(k)|^2} \right] \right\}.$$  (15)

For the triplet gap function (with $d(k)\parallel g_k$) the susceptibility is independent of $\alpha$

$$\chi_{ii}^s = \chi_n \left\{ 1 - k_B T \pi \sum_{\omega_n} \left[ \hat{g}_{k,i}^2 \frac{|d(k)|^2}{\omega_n^2 + |d(k)|^2} \right] \right\}.$$  (16)

More precisely, the contribution due to $\alpha$ from the regular Green’s function is cancelled out by the contribution of the anomalous Green’s function.

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Figure 1. The spin susceptibility in the case of singlet s-wave gap function for $g_k \propto (-k_y, k_x, 0)$ (CePt$_3$Si). The spin susceptibility in the $ab$-plane $\chi_\perp$ and along the $c$-axis $\chi_\parallel$ as a function of $T$ for three different values of the spin–orbit coupling $\alpha$. The susceptibility in the superconducting state ($T/T_c < 1$) increases with the spin–orbit coupling strength. The susceptibility is more strongly suppressed in the $ab$-plane than along the $c$-axis. At $T = 0$ we have $\chi_\perp = \chi_\parallel / 2$.

Figure 2. The zero-temperature value of $\chi_\parallel / \chi_n$ of the spin-singlet state as a function of $\alpha$. Note that $\chi_\perp = \chi_\parallel / 2$.

We now apply the results to the recently discovered heavy fermion superconductor CePt$_3$Si [3]. In this case, the generating point group symmetry is $C_{4v}$ for which the simplest form of $g_k$ is $g_k \propto k \times \hat{z} = (-k_y, k_x, 0)$ [5]. This has the same form as the well-known Rashba spin–orbit coupling [14]. The spin susceptibility for the singlet s-wave gap function is shown in figure 1. The left plot shows the corresponding behaviour of the susceptibility for the field along the $c$-axis ($\chi_\parallel = \chi_{c,c}$). The right plot shows the spin susceptibility for the field in the $ab$-plane ($\chi_\perp = \chi_{a,a} = \chi_{b,b}$) as a function of the temperature for three different values of the spin–orbit coupling ($\alpha$). In figure 2 we show how the zero-temperature value of the susceptibility rises as a function of $\alpha$. For $\alpha \gg k_B T_c$ the $\chi_\parallel$ approaches $\chi_n$ and $\chi_\perp = \chi_n / 2$. 

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Figure 3. The spin susceptibility for a spin-triplet p-wave gap function \( d(k) \| g_k \propto (-k_y, k_x, 0) \) (CePt_3Si). The spin susceptibility in the \( ab \)-plane \( \chi_\perp \) and along the \( c \)-axis \( \chi_\parallel \) as a function of \( T \). The susceptibility is in this case independent of the spin–orbit coupling \( \alpha \). In the superconducting state, the susceptibility in the \( ab \)-plane coincides with that of the normal state.

We remark that the susceptibility increases with the spin–orbit coupling strength. For \( \alpha \) very large, the resulting susceptibility looks very similar to that obtained for the triplet p-wave gap function shown in figure 3. For the spin-triplet phase we chose in accordance with [5] the pairing state \( d = \hat{x}k_y - \hat{y}k_x \). The similar properties of the spin susceptibilities make it difficult to distinguish between a spin-triplet and spin-singlet order parameter through NMR measurements in the strong SOC limit.

Due the complicated band structure of CePt_3Si [7] and the coexistence of superconductivity with antiferromagnetism [15], our theory does not provide a quantitative description for the spin susceptibility for CePt_3Si. However, our approach illustrates the behaviour expected at a qualitative level. More precisely, the susceptibility, independent of the kind of pairing and of the strength of the SOC, is more strongly suppressed in the \( ab \)-plane than along the \( c \)-axis. This angle dependence of the static uniform susceptibility should be confirmed by NMR-Knight-shift measurements. Moreover, our discussion supports the conclusion of Frigeri et al that the spin ‘singlet’ pairing state acquires a certain robustness against pair breaking due to spin polarization [5]. A rough estimate of the zero-temperature limiting field is obtained by comparison of superconducting condensation and spin polarization energy, leading to

\[
H_p \approx \frac{k_B T_c}{\mu_B \sqrt{1 - \chi'(T = 0)/\chi_n}}.
\] (17)

In principle, this can become very big for fields along the \( c \)-axis and roughly 1–2 T for fields in the basal plane.

5. Conclusions

We have determined the spin susceptibility in superconductors without inversion symmetry. While the spin-triplet and spin-singlet order parameters are mixed in general, we can discuss
predominantly spin-triplet or spin-singlet gaps when the spin–orbit coupling is much smaller than the band width. We have found that for the surviving predominantly spin-triplet gap, the lack of inversion symmetry does not change the spin susceptibility. For a predominantly spin-singlet gap, the lack of inversion symmetry leads to an increase in the spin-susceptibility. For large spin–orbit coupling (relative to $T_c$), the spin susceptibilities for both the spin-singlet and spin-triplet gaps become similar. For the heavy fermion superconductor CePt$_3$Si, we have predicted that, independent of the pairing symmetry, the susceptibility is more strongly suppressed in the $ab$-plane than along the $c$-axis. More generally, we may state that the spin susceptibility in a superconductor without inversion centre is approximately described by the behaviour of the spin-triplet superconductor with $d(k) = \Delta_0 g_k$ and is obtained from (16).

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