Correlation between the superconducting pairing symmetry and magnetic anisotropy in $f$-electron unconventional superconductors

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Abstract. The superconducting pairing symmetry and the magnetic anisotropy of the normal state are found empirically to be strongly correlated in $f$-electron unconventional superconductors having crystallographic symmetry lower than cubic. Effectively, there are three categories: 1) In antiferromagnetic systems, unconventional superconductivity appears with singlet ($d$-wave) pairing or mixed-parity pairing (in non-centrosymmetric compounds) for cases of XY anisotropy. 2) In ferromagnetic systems, unconventional superconductivity with triplet ($e.g.$ $p$-wave) pairing appears for cases of Ising anisotropy. 3) A few exceptional cases: The origin of the observed correlation will be discussed in terms of the orbital $f$-electron states.

1. Introduction

In $f$-electron systems, unconventional superconducting (SC) states with an anisotropic superconducting gap are found to occur owing to strong correlations between the $f$-electrons. In these superconductors, the SC pairing force is considered to be mediated by magnetic fluctuations. In fact, superconductivity in such cases is often near a magnetic instability, so that magnetic ordering is easily induced through variation of a tuning parameter such as pressure or doping [1].

In compounds with strong electron correlations such as heavy fermion superconductors and high $T_c$ cuprates, $d$-wave superconductivity is favored for antiferromagnetic correlations, whereas $p$-wave superconductivity is favored for ferromagnetic correlations. This relation between the SC symmetry and the nature of the magnetic correlations has been interpreted qualitatively with theoretical models [2].

In addition to the type of magnetic correlations, the magnetic anisotropies of the $f$-electron moments themselves, $i.e.$ Ising, XY, and Isotropic symmetries, emerge as a result of strong spin-orbit coupling, in contrast to $d$-electron systems such as high $T_c$ cuprates that show a nearly isotropic magnetism confirmed by neutron scattering [3]. Monthoux and Lonzarich have pointed out that Ising fluctuations qualitatively favor ferromagnetic $p$-wave superconductivity, whereas isotropic fluctuations favor antiferromagnetic, $d$-wave superconductivity. This is because $p$-wave pairing occurs only through the longitudinal magnetic fluctuation channel, whereas $d$-wave pairing occurs equally through the longitudinal as well as the two transverse fluctuation.
channels [4]. However, up to now the relation between magnetic anisotropy and superconducting symmetry has not been addressed, and no concrete theoretical description has been reported.

In our previous work [5, 6], the experimental correlation between $d$-wave superconductivity and XY anisotropy has been discussed for the case of heavy fermion superconductors. Recently, a clear correlation between $T_c$ and the strength of XY anisotropy has also been reported for $d$-wave superconductivity in $f$-electron heavy fermion systems [7, 8].

In the present report, we address this issue in greater detail. We classify a series of unconventional, $f$-electron superconductors in terms of their magnetic anisotropy and the symmetry of their superconducting order parameters, finding results which show a strong correlation. Since magnetic anisotropy is a consequence of the relevant orbital states and the crystal field scheme, the results imply a relation between those properties and the nature of unconventional superconductivity.

2. Empirical correlation

In Table 1, the magnetic anisotropy of the magnetic ions and the superconducting symmetry are presented for a series of unconventional, $f$-electron superconductors. Generally, the unconventional superconductivity appears around the magnetic instability point (i.e. quantum critical point) in $f$-electron systems. In compounds with ordered ground state at $P=0$, systems approach to the magnetic instability point under pressure. The magnetic anisotropy at correlation wave vector $q_{cor}$ can be determined experimentally by means of static magnetic susceptibility data for ferromagnetic cases ($q_{cor} = 0$), and with NMR and neutron scattering data for antiferromagnetic cases ($q_{cor} = Q \neq 0$).

For the hexagonal, trigonal, and tetragonal compounds in Table 1, the magnetic anisotropy has axial symmetry at magnetic ions; thus, there are two magnetic symmetry axes in the paramagnetic state, i.e. the a-axis in the basal plane and the c-axis perpendicular to the basal plane.

In compounds that do not order magnetically, the magnetic anisotropy is determined by the anisotropy of magnetic fluctuations, i.e. the magnetic fluctuation energy along the a-axis $\Gamma_a$ is smaller than that along the c-axis $\Gamma_c$ for the XY-type, whereas $\Gamma_a > \Gamma_c$ for the Ising-type. Compounds that order ferromagnetically or antiferromagnetically are designated XY-type if the ordered magnetic moment is in the basal plane and Ising-type if it lies along the c-axis.

Ising-type behavior is assigned to orthorhombic ferromagnetic compounds UGe$_2$, URhGe and UCoGe, all of which have three magnetic a,b,c-axes at the magnetic ions.

In some compounds, especially compounds showing superconductivity under high pressure, the symmetry of the superconducting order parameter has not yet been determined. Even in these cases, the correlation between the magnetic anisotropy and the type of magnetism (antiferromagnetic or ferromagnetic) is clear. It should be noted that such a correlation has not yet been confirmed in CeCu$_2$Si$_2$ or CeIrSi$_3$.

Generally, we find there to be three categories of behavior:

- In antiferromagnetic systems, superconductivity occurs for XY behavior with:
  a) singlet pairing ($d$-wave symmetry);
  b) possibly mixed parity pairing for non-centrosymmetric compounds.
- In ferromagnetic systems, superconductivity occurs for Ising behavior with triplet pairing (e.g. $p$-wave) symmetry.
- Exceptional cases which cannot be categorized in the above pattern.

2.1. Ordinary cases

The observed strong correlation between the symmetry and the magnetic anisotropy suggests that unconventional superconductivity should be related to the magnetic anisotropy. We call the
Table 1. Magnetic Anisotropy and Symmetry of Superconducting State in Heavy Fermion Systems.

| Compound* (Crystal structure) | Ground state ($P=0$)* (SC $T_N$, $T_{HD}$) | Magnetic anisotropy at $q_{cor}$ ($P=0$) | SC symmetry | Anisotropy of static $\chi$ (200K) |
|------------------------------|------------------------------------------|----------------------------------------|-------------|-----------------------------------|
| **Ferromagnetic**            |                                          |                                        |             |                                   |
| UGe$_2$ (O)                  | F (0.7K at 1.2GPa)                       | I ($m_0 [001]$)[28]                   | Tri[29]     | I[28]                             |
| URhGe (O)                    | SC (0.25K)                               | I ($[001]$)[30]                       | Tri[31]     | I[30]                             |
| UCoGe (O)                    | F+SC (0.6K)                              | I ($m_0 [001]$)[32, 33]              |?                         | I[32]                             |
| **Antiferromagnetic**        |                                          |                                        |             |                                   |
| CeCoIn$_5$ (T)               | SC (2.3K)                                | XY [34]                               | d[35]        | I[34]                             |
| CeRhIn$_5$ (T)               | SC (0.4K)                                | XY [36]                               | d[37]        | I[36]                             |
| CeRhIn$_5$ (T)               | AF (2K at 2GPa)                          | XY ($m_Q [110]$)[23]                 | Node[17]    | I[21]                             |
| PuRuGa$_5$ (T)               | SC (9K)                                  | XY [5]                                | d[38]        | I(~Iso)[39]                       |
| PuCoGa$_5$ (T)               | SC (18K)                                 | XY [7]                                | d[40]        | ?                                 |
| NpPd$_2$Al$_2$ (T)           | SC (5K)                                  | XY [41]                               | d[42]        | XY[43]                            |
| CeCu$_2$Si$_2$ (T)           | SC (0.6K: S phase)                       | XY [44]                               | d[45]        | I[46]                             |
| CePd$_2$Si$_2$ (T)           | AF (0.4K at 2.8GPa)                      | XY ($m_Q [110]$)[47]                 | ?                        | XY[47]                            |
| CeNi$_2$Ge$_2$ (T)           | P (0.4K at 2.6GPa)                       | XY ($m_Q [110]$)[48]                 | ?                        | I[49]                             |
| UPd$_2$Al$_3$ (H)            | AF+SC (1.8K)                             | XY($m_Q [1010]$)[50]                | d[51]        | XY[52]                            |
| **Exceptional case**         |                                          |                                        |             |                                   |
| CePt$_3$Si (T NC)            | AF+SC (0.75K)                            | XY ($m_Q [001]$)[53]                 | Mixed[54]    | I[55]                             |
| CeIrSi$_3$ (T NC)            | AF (1.6K at 2.6GPa)                      | ?                                     | ?                        | XY[56]                            |
| CeRhSi$_3$ (T NC)            | AF (1K at 2.5GPa)                        | XY ($m_Q [001]$)[57]                 | ?                        | XY[58]                            |
| **Cubic systems**            |                                          |                                        |             |                                   |
| CeIn$_3$                     | AF (0.2K at 2.4GPa)                      | ?                                     | p[72]        | Iso                               |
| PrOs$_3$Sb$_{12}$            | SC (1.85K)                               | ?                                     | p[73]        | Iso                               |
| UBe$_{13}$                   | SC (0.95K)[31]                           | ?                                     | p[73]        | Iso                               |

T: Tetragonal, H: Hexagonal, O: Orthorhombic, NC: Non-centrosymmetric, AF: Antiferromagnetic ordering, F: Ferromagnetic ordering, P: Paramagnetic, HD: Hidden ordering, SC: Superconducting, I: Ising, Iso: Isotropic, $m_Q$: Antiferromagnetic ordered moment, $m_0$: Ferromagnetic ordered moment, Node: Existence of SC gap node is suggested, Tri: Triplet pairing, Mixed: Mixed parity paring.

* Please see ref. [27]. 1) Antiferromagnetic ordered state under pressure, 2) There is another maximum of $T_c = 2K$ at 17 GPa, 3) There may be another SC phase with $T_c = 0.75K$. 

Tetragonal systems include tetragonal, orthorhombic and non-centrosymmetric, with $q_{cor} = 0$ (fixed). The other columns contain various SC symmetries and magnetic anisotropies, with $q_{cor}$ denoting the wavevector of magnetic ordering. The last column lists the SC temperatures, $T_c$, with the maximum value for each compound.
first and second categories "ordinary cases", since it appears natural that the antiferromagnetic-XY and ferromagnetic-Ising systems are found to be more favourable for $d$-wave and $p$-wave superconductivity, respectively, in compounds with symmetry lower than cubic. It should be noted, however, that $d$-wave superconductivity occurs in systems with strong XY anisotropy rather than in nearly isotropic systems in the case of f-electron compounds [5, 7], an effect which goes beyond the previous theoretical explanation [4].

The possibility that mixed symmetry occurs in compounds without inversion symmetry is well known by now.

2.2. Exceptional case

Now let us examine exceptional cases which do not show the same pattern as the ordinary cases. In a certain sense, these exceptional cases are more curious compared with ordinary cases, since there may be interesting reasons for the exception.

In URu$_2$Si$_2$, superconductivity appears only in the phase with hidden order, thus the Ising antiferromagnetic fluctuations found near the antiferromagnetic ordered phase may not be relevant to the superconductivity. In fact, an incommensurate magnetic excitation at $q_{cor} = (1 \pm 0.4, 0, 0)$ was found recently in the hidden ordered phase [9], which is different from $q_{cor} = (0, 0, 1)$, which characterizes the antiferromagnetic ordering. The superconductivity could be mediated by fluctuations of the hidden order parameter, which may be multipolar (spin$\otimes$orbital) ordering [10, 11].

In UPt$_3$, the nature of the antiferromagnetic ordering is quite different from an ordinary case, i.e. it appears to be a tiny moment ($\sim 0.02 \mu_B$) which exhibits slow fluctuations [12]. Thus, the relation between superconductivity and antiferromagnetic fluctuations may be different from usual. In fact, it has been proposed that superconductivity is mediated by other fluctuations (e.g. ferromagnetic fluctuations) in this compound [12].

In CeRh$_2$Si$_2$, CeCoGe$_3$ and CeCu$_2$Ge$_2$, superconductivity occurs in Ising or Isotropic-like antiferromagnets. Remarkably, these compounds have a complex antiferromagnetic ordered state in common. In CeRh$_2$Si$_2$, there are two antiferromagnetically ordered phases with different multi-$K$ structures, i.e. $4K$-structure below 36 K and $2K$-structure below 25 K [13], although the magnetic structure around the critical pressure 1.1GPa for magnetic instability has not yet been determined. In CeCoGe$_3$ there are three different antiferromagnetically ordered phases at $P = 0$, occurring at $T_{N1} = 20$ K, $T_{N2} = 11.5$ K, and $T_{N3} = 7.5$ K [14], in addition, the transitions at $T_{N2}$ and $T_{N3}$ may be 1st order phase transitions with a change of ordered structure [15]. In CeCu$_2$Ge$_2$, two different antiferromagnetically ordered states appear upon substitution of a few percent of Ni in the place of Cu [16]. In this compound, a complex incommensurate $[q_{cor} = (0.284, 0.284, 0.543)]$ ordered structure with a sinusoidally modulated amplitude is found with $m_Q$ inclined 10 degree from $q_{cor}$.

Perhaps the exceptional behavior in these compounds is related to these complex ordered phases, indicating a frustrated exchange interaction. In any case, in order to confirm these exceptional properties, a determination of magnetic anisotropy and superconducting symmetry under pressure is certainly necessary for these compounds. In fact, $q_{cor}$ may be found to depend on pressure [17].

It is somewhat surprising that UNi$_2$Al$_3$ and UPd$_2$Al$_3$ have a different superconducting symmetry, since these compounds are quite similar. As it was difficult to synthesize a high quality sample of UNi$_2$Al$_3$, it may be necessary to check the superconducting symmetry of UNi$_2$Al$_3$ using a better sample.

2.3. Cubic case

Finally, we address the case of cubic systems. In the cubic compound CeIn$_3$ with cubic local symmetry for Ce, magnetic fluctuations should be isotropic in the paramagnetic state. However,
a recent study implied that the AF ordered moment is along the [110] direction \([18]\), indicating that XY anisotropy develops near the ordered state. It may then be possible to categorize CeIn\(_3\) as an ordinary case for future work.

Concerning the systems UBe\(_{13}\) and PrOs\(_4\)Sb\(_{12}\), it is difficult to discuss the pairing symmetry and magnetism here, since many physical properties remain controversial at the present time.

3. Orbital state and superconductivity

Several interesting issues emerge from the categorizations we propose here.

- Is XY more favourable than Ising-type fluctuation for unconventional superconductivity in \(f\)-electron antiferromagnets?
- Do anisotropic fluctuations develop around the superconducting transition even in cubic systems?
- What is the origin of the exceptional cases?

It is clear that the magnetic anisotropy is due to the spin-orbit coupling, crystal field and anisotropic orbital states. Thus, the relation between the orbital states and the superconductivity needs to be considered. For example, a particular orbital state with XY-anisotropy can be more favourable than an isotropic one for antiferromagnetic cases. Concerning fluctuations relevant to superconductivity, orbital and multipolar fluctuations should be addressed in addition to spin fluctuations. Although the orbital states are difficult to treat in itinerant systems, the following theoretical approaches have recently been proposed.

Takimoto \textit{et al} \([19]\) have pointed out that an orbital splitting energy \(\Delta\) between the \(\Gamma(1)\) and \(\Gamma(2)\) levels can be relevant to \(T_c\) in tetragonal Ce-based superconductors. In the crystal field scheme for the Ce\(^{3+}\) ion in tetragonal symmetry, two \(\Gamma(1,2)\) and one \(\Gamma_6\) doublets are formed. In CeTIn\(_5\) superconductors for example, the following \(\Gamma(1,2)\) level is lower than the \(\Gamma_6\).

\[
\begin{align*}
|\Gamma_{6\pm}\rangle &= |\pm \frac{1}{2}\rangle \\
|\Gamma_{7\pm}^{(1)}\rangle &= \sqrt{\frac{5}{6}} |\pm \frac{5}{2}\rangle + \sqrt{\frac{1}{6}} |\pm \frac{3}{2}\rangle \\
|\Gamma_{7\pm}^{(2)}\rangle &= \sqrt{\frac{1}{6}} |\pm \frac{5}{2}\rangle - \sqrt{\frac{5}{6}} |\pm \frac{3}{2}\rangle
\end{align*}
\]

In this model, the \(T_c\) for \(d\)-wave superconductivity increases with increasing \(\Delta\) up to a finite value, after which an antiferromagnetically ordered state appears. The enhancement of \(T_c\) is ascribed to enhancement of spin fluctuations and suppression of orbital fluctuations which occur with increasing \(\Delta\). If the antiferromagnetically XY anisotropy is correlated with \(\Delta\), this model may explain the correlations between \(d\)-wave superconductivity and antiferromagnetic XY anisotropy through the spin and orbital fluctuations.

Flint \textit{et al} \([20]\) have proposed a composite superconducting pairing mechanism for actinide superconductors. In this model, Cooper pairs are formed using three different orbital states. This model could potentially identify the relation between the superconductivity and the magnetic anisotropy.

In \(f\)-electron systems, there may be either a localized or an itinerant origin for the observed magnetic anisotropy: 1) Local magnetic anisotropy determined by the crystal field and the \(f\)-electron state; 2) the Ruderman, Kittel, Kasuya and Yosida (RKKY) interaction due to hybridization between \(f\)-electrons and conduction electrons.

If the localized character is strong, the anisotropy of the static susceptibility can be qualitatively explained in terms of a crystal field scheme based on the \(LS\)-coupling description.
This is the case for many Ce compounds [21]. However, as shown in Table 1, the anisotropy of the static susceptibility \((q = 0)\) does not always coincide with the magnetic anisotropy at the antiferromagnetic wave vector \((q = Q)\). This deviation indicates that the RKKY exchange interaction has a \(q\)-dependence, an effect which is clearly related to the itinerant nature of the system. In order to treat the actual magnetic anisotropy over a wide range of \(q\)-values, it is necessary to treat correctly the intermediate properties of \(f\)-electrons between localised and itinerant. The \(jj\)-coupling description may be suitable for this purpose [22].

It should be noted that the dimensionality and the magnetic anisotropy should be distinguished. Strong XY anisotropy does not mean strong two dimensional magnetic character for systems. The measure of two dimensionality is the anisotropy of the magnetic correlation length \(\xi_a/\xi_c\) (in units of the lattice parameter ratio \(a/c\)), which is typically 0.5 ~ 5 in the tetragonal and hexagonal unconventional superconductors in Table 1 \((\xi_a/\xi_c = \infty\) in the two dimensional systems) [23, 24]. This fact indicates that these compounds are anisotropic, but still three dimensional.

Recently, a possible valence (monopolar) fluctuation-mediated superconductivity has been discussed [25]. The orbital character can be modified at a valence transition. Clearly, valence fluctuations are an important aspect of \(f\)-electron systems; however, the relation between this scenario and the present findings is an open question at the present stage of things. The valence and magnetic fluctuations can also be mixed, a situation which may be realized in CeCu\(_2\)Ge\(_2\) and CeCu\(_2\)Si\(_2\) [26].

4. Conclusion

The symmetry character of unconventional superconductivity in \(f\)-electron systems is correlated with the magnetic anisotropy in the normal state. This indicates strongly that the spin-orbit coupling and orbital character must be incorporated in the mechanism of unconventional superconductivity in \(f\)-electron systems. If spin, orbital, and multipolar degrees of freedom are correctly treated in the mechanism of superconductivity, the existence of these categories is expected to be elucidated in a unified picture.

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