MAGNETISM AND SUPERCONDUCTIVITY IN HEAVY FERMION SYSTEMS

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The normal and superconducting properties of heavy fermion compounds are reviewed. The discussion is focus on the three uranium compounds : UBe₃, UPt₃ and URu₂Si₂. Special attention is given : 1) to unusual (H,T) superconducting phase diagram as discovered in UPt₃ where two successive superconducting phases seem to occur in zero magnetic field ; 2) to the role of long range ordering as found in URu₂Si₂ and UPt₃.

1. Introduction

The observation of a large linear temperature term γT in the specific heat C of different intermetallic compounds containing f electrons (mainly Ce, Yb or U ions) suggests that heavy fermions are built at low temperatures. Now the occurrence of such heavy quasiparticles is well established ; in the Haas-van Alphen experiment, a direct proof is given by the detection of well defined electronic orbits with effective masses m* near 100 m₀ (m₀ the free electron mass). The heaviness of the mass is mostly the consequence that the effective characteristic Fermi temperature T_F is weak ; the f electron recovers its full magnetic entropy (Rlog₂ for a spin S=1/2) at rather low temperature (T~ 100K). This heavy quasiparticle is the result of the coupling between initially well localized f electron and light itinerant electron ; by hybridization, the f electrons can be delocalized and appear to move with a low Fermi velocity. A large part of the mass enhancement is obtained by local magnetic fluctuation reminiscent of the single impurity kondo effect with an intersite magnetic coupling J_ij comparable to the local Kondo energy k_BT_K. The consequences of this interplay are that i) heavy fermion compounds are near magnetic instability i.e. near the transition between a long range magnetic ordering (generally antiferromagnetic with a Neél temperature (T_N)) and a Pauli paramagnetic ground state and ii) the weight of the dynamical magnetic susceptibility χ(q,ω) is distributed over a wide range of frequency ω and wavevector q. A large variety of different ground state occurs in heavy fermion compounds ; the first observation of a superconducting transition in CeCu₂Si₂ was a surprise as it was generally considered in metals that magnetism and superconductivity are antagonist.

Today, the idea is that, when the quasiparticles are built below T*≤T_F/10 i.e. when the T² resistivity law characteristic of electron-electron interactions are observed, the large magnetic fluctuating medium detected in the dynamical susceptibility response can furnish an attractive potential for the moving heavy quasiparticles and thus can lead to the superconducting pairing of
heavy electrons. If the ferromagnetic fluctuation dominates, the p pairing with odd parity will occur (analogy with the superfluid phases of $^3$He) ; for a large enough amplitude of the antiferromagnetic fluctuations, the d pairing with even parity will emerge. As for liquid $^3$He, the new features are the possible occurrence of a line of zero or points of zero of the energy gap at the Fermi surface and of a multicomponent order parameter which may be not a simple scalar as in the usual BCS superconductors.

2. Evidences for the unconventional nature of the superconductivity

We will concentrate here only on the three well known uranium heavy fermion superconductors UBe$_3$, UPt$_3$ and URu$_2$Si$_2$ which become superconducting respectively at $T_c \sim 0.9K$, 0.5K, 1.2K. Figure 1 represents the ratio $C/T$ of the specific heat by the temperature versus the reduced unit $T/T_c$. As the specific heat jump at $T_c$ is comparable to the value $\gamma_n T_c$ of the normal phase just above $T_c$, it is clear that heavy fermions participate to the Cooper pairing. For UBe$_3$, a positive curvature of $C/T$ on approaching $T_c$ appears; for UBe$_3$ and URu$_2$Si$_2$ a unique transition is observed whereas for UPt$_3$ a splitting in the transition seems to exist.

At low temperature, for UBe$_3$ the observation of a nearly $T^3$ dependence of $C$ was taken as the first indication of an axial superconducting state with points of zero in the energy gap ; the deviation from $T^2$ law at lower temperature ($Tc$=150mK) was interpreted as due to impurities in a strong coupling with the pure lattice. It must be stressed here that an important source of defects is due to weak fluctuations in the atomic distances (near dislocation, stacking fault, inversion among sites). The weak value of the characteristic temperature $T_F$ leads to a unusual high relative sensitivity to any local volume (V) change ; the corresponding Grüneisen parameter $\Omega = -\partial \log T_F / \partial \log V \sim 100$ scales roughly the huge mass enhancement. Thus impurities can be easily induced by imperfections. They have drastic effects and may spoil the intrinsic behavior of the pure lattice.

In UPt$_3$, as well as in URu$_2$Si$_2$, the $T^2$ dependence of $C$ was taken as the proof of a polar superconducting state with a line of zero although no unique $T^2$ law even in a restricted range of temperature can represent the specific heat data (for example in UPt$_3$ coexistence of $T$ and $T^2$ contribution). The observation of power law instead of the usual exponential decrease on cooling of other different physical quantities like ultrasonic attenuation, nuclear relaxation time $T_1$ and thermal conductivity $\kappa$ seems to confirm the idea of the unconventional nature of the superconductivity pairing. However, the exponents of power laws measured for different observables may be not consistent in a unique frame of axial or polar state ; for example in UBe$_3$, $T^3$ law of $C$ predicts points of zero while the $T^3$ behavior of $T_1$ suggests line of zero. One experimental test of the gap topology is to realize thermal conductivity experiments on single crystals in order to detect not only the temperature dependence of $\kappa$ but also its crystalline anisotropy which must be characteristic of the gap structure.

In Fig. 2, $\kappa (T)$ is plotted as a function of $T$ for two different hexagonal single crystals of UPt$_3$ with the heat current $J$ along the c-axis and along the
Fig. 2 Thermal conductivity $\kappa (T)$ of $\text{UPt}_3$ as $\kappa (T)/T$ versus $T$ for sample A with heat current $J$ parallel to $c$-axis and for sample B with $J$//b.

To reveal the anisotropy of $\kappa (T)$ which goes beyond that of the normal phase of $\text{UPt}_3$, we must consider the normalized conductivity $\kappa (T)/\kappa (T_C)$ versus the reduced temperature $T/T_C$. As these normalized conductivities are virtually the same, the possibility of highly anisotropic gaps such as only polar or axial seems to be rejected. No definitive statement can be drawn since we cannot reject that differences in impurity effects among the crystals may give an accidental quasi-isotropy in the normalized thermal conductivity. In agreement with recent muon experiments, the superconducting state of $\text{UPt}_3$ may be at least an hybrid state with line of zero in the basal plane and point of zero along the $c$-axis. A further elegant way to probe the gap is to analyze near $H_{c2}$ on a given crystal the influence of the relative orientation of the magnetic field $H$ and of the thermal current. Near $T_C$, an hybrid gap is in relative agreement with the experiments.

Concerning parity, one major argument for claiming even parity is that, in the case of a strong spin orbit coupling, for odd parity, lines of zero are excluded for hexagonal or tetragonal lattices. We have already emphasized the uncertainty in the determination of the gap structure. A strong experimental support for even parity will be to observe a change in the Pauli susceptibility at $T_C$ which are generally detected through Knight shift or magnetic form factor measurements. Up to now, only $\text{CeCu}_2\text{Si}_2$ show a clear decrease of the Knight shift below $T_C$. Muon experiments give in $\text{UBE}_{1.3}$ contradictory conclusions. More surprising is the absence of any detectable variation in the Knight shift of $\text{UPt}_3$ despite its large value, thus the possibility to realize sensitive measurements. The rather good quality of the material (residual resistivity near $1 \mu \Omega \cdot \text{cm}$) is not favourable to invoke strong spin orbit impurity scattering for a Knight shift invariance through $T_C$. Today, it is still open that the parity of $\text{UPt}_3$ may be odd. This possibility is supported by a recent analysis of the crossing of the upper critical fields parallel and perpendicular to the $c$-axis and by a study of the pairing mechanism in a lattice of interacting kondo ions.

One important support for a superconducting pairing through spin fluctuations is the link between the volume variation of their Fermi temperature and of their superconducting temperature. As shown in Table 1, their respective Gruneisen parameters $\Omega_{T_C}, \Omega_{T_F}$ have comparable magnitude but opposite sign. The simple scheme is that the pairing interaction $V$ is directly related to the strength of the electron-electron interaction as measured by the coefficient $A V^2$ of the $T^2$ coefficient of the resistivity. For heavy fermion compounds, $A$ scales roughly $y^2$ thus $T_F^{-2}$. Under pressure $T_F$ increases, $A$ decreases and thus $T_C$. Another interesting possibility is also to change the pairing potential by a magnetic field; this seems to occur in $\text{UBE}_{1.3}$ where the unusual shape of the upper critical field may be related to the field variation of $\lambda (H)$.1
3. Long range magnetic coupling

The other new superimposed effect is the occurrence of long range magnetic ordering with very weak sublattice magnetization $m_0$. For URu$_2$Si$_2$, well above $T_C$, at $T_N \sim 17$K, a $\lambda$ type anomaly appears. It was rapidly recognized that it coincides with an antiferromagnetic ordering by neutron diffraction; the magnetic phase preserves the tetragonal crystal symmetry and corresponds to a propagation vector $Q = (0, 0, 1)$ with a very weak $m_0 \sim 0.03 \mu_B$ pointed along the c-axis. This low amplitude, the linear temperature dependence of the neutron or X-ray intensity $I \sim m_0^2$, a resistivity anomaly at $T_N$ reminiscent of the Cr case and the variation of $C/T$ suggest the formation of a spin density wave which coexists below $T_C$ with the superconducting phase without any detectable change in the neutron intensity.

For UPt$_3$, the understanding of the magnetism is not so obvious. The occurrence of a long range ordering is detected by neutron diffraction or muon precession but macroscopic experiments like specific heat, magnetization or transport fail to observe any track of long range ordering. First dynamical experiments show the combination of a broadened local signal of energy near 10meV and of an antiferromagnetic coupling around the wavevector $Q_0 = (0, 0, 1)$ with typical fluctuating energy 5meV. The discovery of long range magnetic ordering in substituted lattices of UPd$_{1-x}$Pt$_x$ or U$_{1-x}$Th$_x$Pt$_3$ with a quite different wavevector $Q_N(1/2, 0, 1)$ than $Q_0$ pushes to explore this novel domain in the pure lattice. A new energy fluctuation spectrum emerges around 0.3meV for $Q_N$. Furthermore below $T_N \sim 5K$, a static diffraction pattern is detected at $Q_N$ with a temperature dependence and strength comparable to those observed in URu$_2$Si$_2$. The weak sublattice magnetization $m_0 \sim 0.02 \mu_B$ is directed along the b-axis in the basal plane; the hexagonal structure of the lattice is broken and an orthorhombic symmetry is established below $T_N$. For $x \sim 5\%$ of Pd or Th, the Neel temperature is comparable to the pure lattice one although $m_0$ has increased by a factor 30; that emphasizes the duality between localized and itinerant magnetism. One may still argue on the intrinsic origin of long range ordering in UPt$_3$ since up to now no experiment shows a correlation length exceeding 300A. However yet, the magnetic ordering has always been detected by neutron diffraction in different crystals measured with a low energy window in order to minimize the background.

In heavy fermion compounds, the strong magnetic fluctuating medium may lead to superconductivity. Superimposed to this medium, a weak long range magnetic ordering may exist. For URu$_2$Si$_2$ and UPt$_3$, the sequence $T_N > T^* > T_C$ is obeyed.

For UBe$_{13}$, at zero pressure, no evidence of antiferromagnetism has seriously been reported. No clear Fermi liquid regime is established when the superconductivity appears i.e. $T^* < T_C$. It has been pointed out that UBe$_{13}$ may show a large fluctuation regime near $T_C$ up to $(T_C - T)/T_C \sim 0.3$ which will explain the specific heat anomaly of Fig.1. Under pressure, $T_C$ vanishes near 40kbar when a magnetic ordering seems to appear in thermoelectric experiments. Systematic pressure measurements must be realized in order to estimate the relative influence of $A$ (or $T^*$), $T_C$, $T_N$ and $m_0$.

4. Successive transitions - Multicomponent superconducting order parameter

U$_{1-x}$Th$_x$Be$_{13}$ : The substitution of U ions by Th ions in U$_{1-x}$Th$_x$Be$_{13}$ produces
a non monotonic depression of the superconducting transition temperature (Tc₁) accompanied by a second transition at Tc₂<Tc₁ for 0.019<x<0.043. Despite the existence of two successive transitions, the new feature not observed for x<0.019 is the appearance at Tc₂ of an increase of the zerofield relaxation rate σkt by μSR on cooling which corresponds to an electronic moment ~ 10⁻³ μg/U atom and to a second order phase transition. Recent magnetization measurements and μSR experiments give the T-x phase diagram of Fig.3.13 Below Tc₂, magnetism is detected by μSR. It is still not clear if it is associated with an antiferromagnetic transition on the uranium sites combined with a new superconducting phase or with a magnetic superconducting phase (time reversal violating state) or with an antiferromagnetic spin density wave transition. The first two proposals are only possible for a multicomponent order parameter. Different theoretical cases have been discussed. A new proposal is the role played by the inelastic scattering of conduction electrons on impurities since it can stimulate s pairing and suppress any non s pairing offering then a non-monotonic behavior of Tc. The recent demonstration (see Fig.3) that the transition line Tc₂ ends up on the Tc₁ line implies that the order parameters of the two phases are strongly coupled. There is still a need for a better understanding of the origin of magnetism below Tc₂ and also for a more quantitative relation between normal and superconducting phases.

UPT₃ : The specific heat of polycrystals of UPT₃ shows two different structures at Tc. It is not obvious to claim that such a splitting cannot reflect the sample inhomogeneity. However, the observations of two well resolved transitions at Tc⁺ and Tc⁻ in different single crystals give a confidence in the intrinsic nature of the phenomena and thus drive a new generation of experiments. A strong support of the intrinsic origin of the splitting is that, under magnetic field, the two transitions collapses at H* equal respectively to 5kOe and 9kOe for H//c and //c. Furthermore, a similar convergence is also obtained under pressure for p*~ 3.7kbar. At H=0, the two successive transitions are observed also in thermal expansion and sound velocity experiments. As it has been verified that i) the same antiferromagnetic ordering persists through Tc⁺ and ii) the superconductivity persists also at Tc⁻, the succession A–B of two different superconducting transitions is admitted. Another line separates a low field B phase from a high field C phase. This quasi-horizontal boundary first detected by acoustic attenuation and confirmed by thermal conductivity is now observed in a sound velocity experiments; its thermodynamic validity is then not ambiguous. The three different states A, B, C seem to end up at a tetracritical point. The Fig.4 represents the phase diagram for H//c. The phase diagram is rather isotropic i.e. similar for H//c and H⊥c. The tetracritical point H* corresponds to a clear isotropic kink in
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the $H_{c2}$ curve measured in the basal plane. The fast pressure disappearance of the kink correlated with the rapid pressure drop of the sublattice magnetization ($m_0 \rightarrow 0$ for $P \sim 3$ kbar) supports the idea that the splitting may be originated through a coupling between the magnetic order parameter and the superconducting multicomponent order parameter. A crucial point is that the magnetic ordering play the role of a symmetry breaking field (see ref.31). A scenario is that the multicomponent order parameter belongs to the two-dimensional representation $E_{1g}$ which is compatible with the hybrid gap previously mentioned. One difficulty is that a kink in $H_{c2}$ will appear in the basal plane only for $H \perp m_0$ while the experiments show its isotropicity. To overcome this paradox, either superconducting glass state or arguments assuming that $m_0$ is always perpendicular to $H$ in the basal plane due to a weak magnetic anisotropy have been presented. The physical basis of a superconducting glass state is that the coherence superconducting length ($\psi_0 \sim 120\AA$) and the magnetic coherence length ($\psi_m \sim 300\AA$) are comparable. Recently, a new one-dimensional representation with spin degeneracy of odd parity has been proposed.

$\text{UPt}_3$ seems to have a multicomponent superconducting order parameter; the magnetic ordering play the role of a symmetry breaking field. This statement is reinforced as $\text{U Ru}_2 \text{Si}_2$ presents no superconducting splitting$^3,3^3$ (at least for good quality samples) in agreement with the preservation of the lattice symmetry at $T_N$. Let us pointed out that recent neutron experiments show that under pressure in $\text{UPt}_3$, $m_0$ drops but $T_N$ is mostly pressure independent. That reminds the results of the series $\text{U(Pt}_{1-x}\text{Pd}_x)_3$ where $T_N$ can have the same value for a quite different amplitude of $m_0$. Different mechanisms are needed to obtain this particular situation.

5. Conclusion

Magnetism and superconductivity interact by different ways in heavy fermion compounds. Strong evidences are now given that superconductivity may have a multicomponent parameter. However a large experimental effort must be pursued to precise the gap topology and the parity, to have a complete knowledge of the Fermi surface as reached in $\text{UPt}_3$, to relate the normal and superconducting properties notably by pressure and uniaxial studies. The first necessary step is to obtain high quality materials in order to dominate the interplay of intrinsic and extrinsic effects. Then experiments at very low temperature ($T \sim 20$ mK) can be revisited; for example the possibility of a third superconducting transition in $\text{UPt}_3$ can be verified. Recently, the family of the heavy fermion uranium superconductors have been extended with the discovery that $\text{UNi}_2\text{Al}_3$ and $\text{UPd}_2\text{Al}_3$ transit in a superconducting phase after crossing an antiferromagnetic transition.

References
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