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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 : UBe_{13}, UPt_3 and URu_2Si_2. Special attention is given : 1) to unusual (H,T) superconducting phase diagram as discovered in UPt_3 where two successive superconducting phases seem to occur in zero magnetic field ; 2) to the role of long range ordering as found in URu_2Si_2 and UPt_3.

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 de Haas-van Alphen experiment, a direct proof is given by the detection of well defined electronic orbits with effective masses m^* near 100 m_0 (m_0 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 Néel temperature (T_N)) and a Pauli paramagnetic ground state and ii) the weight of the dynamical magnetic susceptibility $\chi(q,\omega)$ 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 $CeCu_2Si_2$ 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 $T^* \lesssim T_F/10$ i.e. when the T^2 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[2] which may be not a simple scaler as in the usual BCS superconductors.

2. Evidences for the unconventional nature of the superconductivity

We will concentrate here only on the three wellknown uranium heavy fermion superconductors UBe_{13}, UPt_3 and URu_2Si_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.[3] 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_{13}, a positive curvature of C/T on approaching T_c appears[4] ; for UBe_{13} and URu_2Si_2[3] a unique transition is observed whereas for UPt_3 a splitting in the transition seems to exist.[5]

At low temperature, for UBe_{13} 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^3 law at lower temperature (T<150mK) was interpreted as due to impurities in a strong coupling with the pure lattice.[6] 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 ;their corresponding Grüneisen parameter $\Omega_F = -\partial \log T_F / \partial \log V \sim 100$ scales roughly the huge mass enhancement.[7] Thus impurities can be easily induced by imperfections. They have drastic effects and may spoil the intrinsic behavior of the pure lattice.

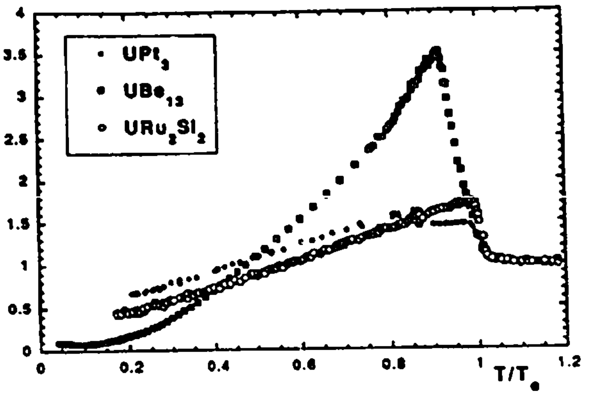

Fig.1. Specific heat of UBe_{13}, UPt_3 and URu_2Si_2 as $(C/T)/\gamma_n$ versus T/T_c.[3]

In UPt_3, as well as in URu_2Si_2, the T^2 dependence of C was taken as the proof of a polar superconducting state with a line of zero[8] 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[9], nuclear relaxation time T_1[10] and thermal conductivity κ[8] 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_{13}, T^3 law of C predicts points of zero while the T^3 behavior of T_1^{-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 κ but also its crystalline anisotropy which must be characteristic of the gap structure.[11]

In Fig.2, κ (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

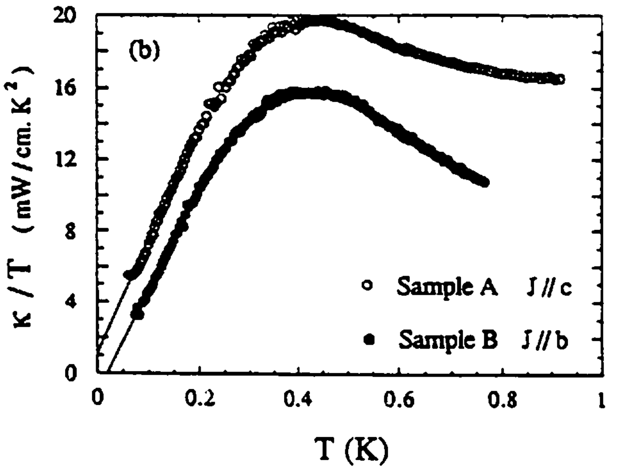

Fig.2 Thermal conductivity κ (T) of UPt_3 as κ (T)/T versus T for sample A with heat current J parallel to c-axis and for sample B with J//b.

basal plane (b-axis).[19] To reveal the anisotropy of κ (T) which goes beyond that of the normal phase of UPt_3, we must consider the normalized conductivity κ (T)/κ (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[12], the superconducting state of 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.[11]

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.[3] 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[10] or magnetic form factor measurements. Up to now, only $CeCu_2Si_2$[10] show a clear decrease of the Knight shift below T_c. Muon experiments give in UBe_{13} contradictory conclusions.[13,14] More surprising is the absence of any detectable variation in the Knight shift of UPt_3[10] despite its large value, thus the possibility to realize sensitive measurements. The rather good quality of the material (residual resistivity near 1 $\mu\Omega$ cm) is not favourable to invoke strong spin orbit impurity scattering for a Knight shift invariance throught T_c. Today, it is still open that the parity of 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[15] and by a study of the pairing mechanism in a lattice of interacting Kondo ions.[16]

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 (Ω_{T_c}, Ω_{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 $A \sim V^2$ of the T^2 coefficient of the resistivity. For heavy fermion compounds, A scales roughly γ^2 thus T_F^{-2}. Under pressure T_F increases, A decreases and thus T_c.[4] Another interesting possibility is also to change the pairing potential by a magnetic field ; this seems to occur in UBe_{13} where the unusual shape of the upper critical field may be related to the field variation of A(H).[4]

Table 1 : Grüneisen parameters Ω_{T_c} and Ω_{T_F}

	UPt_3	URu_2Si_2	UBe_{13}
Ω_{T_c}	-65	-59	-21
Ω_{T_F}	50	40	52

3. Long range magnetic coupling

The other new superimposed effect is the occurrence of long range magnetic ordering with very weak sublattice magnetization m_o. For URu_2Si_2, well above T_c, at $T_N \sim 17K$, a λ type anomaly appears. It was rapidly recognized that it coincides with an antiferromagnetic ordering by neutron diffraction[17] ; the magnetic phase preserves the tetragonal crystal symmetry and corresponds to a propagation vector $Q=(0,0,1)$ with a very weak $m_o \sim 0.03\mu_B$ pointed along the c-axis. This low amplitude, the linear temperature dependence of the neutron or X-ray intensity $I \sim m_o^2$ [18], a resistivity anomaly at T_N reminiscent of the Cr case[19] 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.[20] 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 $Q_o=(0,0,1)$ with typical fluctuating energy 5meV. The discovery of long range magnetic ordering in substituted lattices of $U(Pd_xPt_{1-x})_3$ or $U_{1-x}Th_xPt_3$ with a quite different wavevector $Q_N(1/2,0,1)$ than Q_o pushes to explorate 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_2Si_2. The weak sublattice magnetization $m_o \sim 0.02\mu_B$ 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_o 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 300Å.[20,22] However yet, the magnetic ordering has been always 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_2Si_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.[4] Under pressure, T_c vanishes near 40kbar when a magnetic ordering seems to appear in thermoelectric experiments.[23] Systematic pressure measurements must be realized in order to estimate the relative influence of A (or T^*), T_c, T_N and m_o.

4. Successive transitions - Multicomponent superconducting order parameter

$U_{1-x}Th_xBe_{13}$: The substitution of U ions by Th ions in $U_{1-x}Th_xBe_{13}$ produces

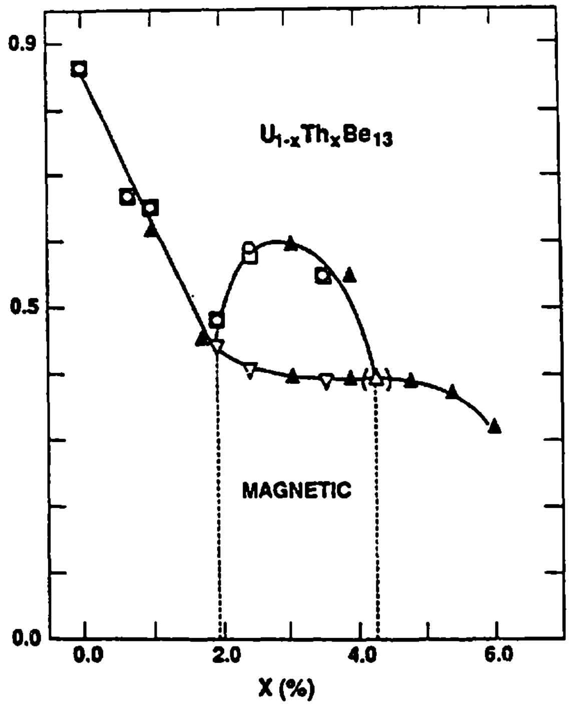

Fig.3. Phase diagram for $U_{1-x}Th_xBe_{13}$.[13]

a non monotonic depression of the superconducting transition temperature (T_{c1}) accompanied by a second transition at $T_{c2}<T_{c1}$ for $0.019<x<0.043$.[13] Despite the existence of two successive transitions, the new feature not observed for $x<0.019$ is the appearance at T_{c2} of an increase of the zerofield relaxation rate σ_{KT} by μ SR on cooling which corresponds to an electronic moment $\sim 10^{-3}$ μ_B/U atom and to a second order phase transition. Recent magnetization measurements and μ SR experiments give the T-x phase diagram of Fig.3.[13] Below T_{c2}, magnetism is detected by μ SR. 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[24] 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 T_c. The recent demonstration (see Fig.3) that the transition line T_{c2} ends up on the T_{c1} 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 T_{c2} and also for a more quantitative relation between normal and superconducting phases.

UPt_3 : The specific heat of polycrystals of UPt_3 shows two different structures at T_c.[8] It is not obvious to claim that such a splitting cannot reflect the sample inhomogeneity. However, the observations of two well resolved transitions at T_c^+ and T_c^- in different single crystals[5,25] 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.[12,22] Furthermore, a similar convergence is also obtained under pressure for P* ~ 3.7kbar.[26]

At H=0, the two successive transitions are observed also in thermal expansion and sound velocity experiments.[27,28] As it has been verified that i) the same antiferromagnetic ordering persists through $T_c^{\pm}$ [29] and ii) the superconductivity persists also at T_c^-[5], 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[30] and confirmed by thermal conductivity[11] is now observed in a sound velocity experiments[27,28] ; 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.[5] The phase diagram is rather isotropic i.e. similar for H//c and H ⊥ c. The tetracritical point H*, T* corresponds to a clear isotropic kink in the H_{c2} curve measured in the basal plane.[22] The fast pressure disappearance of the kink correlated with the rapid pressure drop of the sublattice magnetization ($m_0 \rightarrow 0$ for P~ 3kbar) 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.[22] 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 ($\psi_0 \sim 120$Å) and the magnetic coherence length ($\psi_m \sim 300$Å) are comparable.[31] Recently, a new one-dimensional representation with spin degeneracy of odd parity has benn proposed.[32]

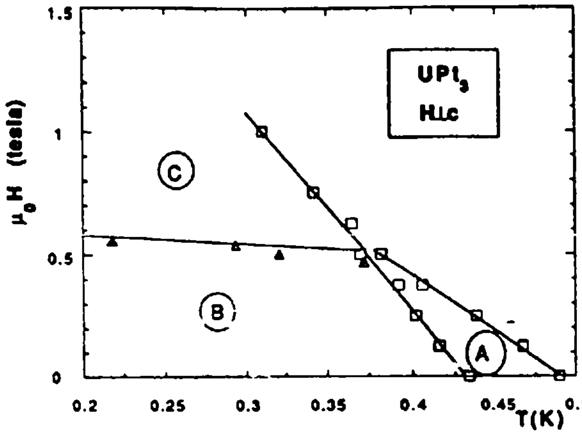

Fig.4. The phase diagram of UPt_3 for $H \perp c$.

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 URu_2Si_2 presents no superconducting splitting[3,33] (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 UPt_3, m_0 drops but T_N is mostly pressure independent.[34] That reminds the results of the serie $U(Pt_{1-x}Pd_x)_3$ 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 complet knowledge of the Fermi surface as reached in 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~ 20mK) can be revisited ; for example the possibility of a third superconducting transition in UPt_3[35] can be verified. Recently, the family of the heavy fermion uranium superconductors have been extended with the discovery that UNi_2Al_3 and UPd_2Al_3 transit in a superconducting phase after crossing an antiferromagnetic transition.[36]

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