The Superconducting Phases of UPt_3

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Abstract

The heavy fermion superconducting UPt_3 compound is an example of unconventional superconductivity, in which both the gauge and the point group symmetries are broken in ordered phase. I present the experiments that provide clues about the order parameter and clearly indicate a coupling between antiferromagnetism and superconductivity. I mention the leading theoretical models that aim to explain the multicomponent phase diagram and identify the superconducting phases, although up to now no single theoretical scenario is completely consistent with all of the data.

1 *Introduction*

Uranium-platinum compounds $UPt_n(n - 0, 1, 2, 3, 3, 5)$ were studied in the early 1980s to understand the nature of the U 5f wave functions. Systematic studies of valence-band photoemission on these compounds showed a strong peak (associated with U 5f electrons) below the Fermi energy, a peak that gradually disappears as n is increased. In UPt_3 the peak is only a shoulder, evidence that suggested that this compound might be in an interesting intermediate hybridization range. The first anomalous behavior (Visser, 1987) observed in UPt_3 is the upturn in the curve of $\frac{C_v}{T}$ as a function of T^2, which is associated with spin fluctuations. Also, the strong degree of electron correlations and narrow quasiparticle bands, as reflected in a large γ (440 $\frac{mJ}{K^2 molU}$), which explains why UPt_3 is called a "heavy-fermion" system (HFS).

A lot of interest has been shown to this HFS since convincing evidence has been discovered [7] for bulk superconductivity below $T = 0.5K$ based on specific heat, resistance, and ac susceptibility measurements. Much of the interest has been fueled by the possibility that the superconducting instability in this metal is mostly the result of direct electron-electron interactions. To test this idea, a lot of direct information has been accumulated about the dependence on the temperature of the thermodynamic and transport properties.

There are three types of evidence to indicate that UPt_3 is an uncoventional superconductor: (*a*) anisotropic transport properties in the superconducting state, most notably the ultrasonic attenuation and the thermal conductivity, (*b*) the multiplicity of phases, most notably seen in the specific-heat and ultrasonic measurements, and (*c*) the absence of activated temperature dependence in any of its physical properties. Qualitatively, these very disparate physical properties are naturally explained in the framework of unconventional superconductivity.

Why is this compound such a fascinating challenge for both theoretical and experimental physicists? First of all, UPt_3 is the only compelling instant in nature that exhibits a new physical phenomenon–superconductor with multiple phases. The study of the phenomenon carries us to the deepest issues of correlated electron physics. Then, there are several phenomena that raise a tantalizing situation when attempting to explain the behaviour of UPt_3.

Consider the split transition. The 2D and 3D representations give a natural explanation for the transition; the split is caused by the coupling of superconductivity to magnetism, a hypothesis supported by the pressure data. However, sound velocities data and neutron-scattering experiments do not support these representations. One of the primary ideas in this paper [1] is that if the two troublesome issues arising from these two types of experiments can be solved in the framework of the 2D picture, this representation will give very nice agreement of the theory with the experiment for the entire H-T-P phase diagram. Also, given the fact that the class of theories in which the K parameter (which couples the direction of the order parameter to the field direction) vanishes or is small, has difficulty with the behavior of the normal-superconducting phase boundaries under pressure, the paper concludes that the E_1g representation, in which the K parameter is appreciable, is the best candidate to solve the problem.

Another teasing situation shows up in the phenomena of low-temperature and anisotropic properties. The specific heat suggests that there are gap nodes, but sheds little light on the nodal structure. Ultrasonic attenuation clearly shows that there is a line of nodes in the basal plane, so the presumption now is that the gap is odd under reflection in the x-y plane. The only tool that we have to probe in detail what is happening at the gap nodes is low-temperature measurement of the thermal conductivity. Data in the asymptotic regime shows gap nodes at the poles with quadratic dispersion. In the gapless regime, no theory appears to account for the data too well.

The approach used in the paper of Joynt and Taillefer is to discuss the topics in which meaningful comparison between theory and experiment can be made. In order to make this comparison, the theories and experiments are classified into groups, so that, within a class of theories, the predictions for experiment are rather similar, and respectively, within a class of experiments, the constraints on theories resemble each other. Because of this choice of approaching the problem, theories of vortices, vortex lattices and the pairing mechanism are discussed only briefly. There is no mention at all of surface effects, Andreev scattering and collective modes.

2 *Theoretical Models*

2.1 Description of the superconducting states and classification

The unusual phase diagram and thermodynamics of UPt_3 shows unambiguously that the superconductivity of UPt_3 has unconventional symmetry–there are not one, but several superconducting phases in UPt_3. It becomes superconducting at about T_{c+}=0.5K, entering the A phase, and has a second transition at about T_{c-}=0.44K entering the B phase. Experiments done on two polycrystals [5] and on a single crystal [6] have shown "shoulders" in the shape of the specific heat. The samples showed two distinct field dependent transition temperatures maxima $T_{c1}(H)$ and $T_{c2}(H)$ observed in the specific heat, thermal expansion, ultrasound and mechanical measurements. The precise nature of its superconducting order parameter remains elusive, however.

The s-wave is unique, so at least two of the three phases have novel symmetry. The analysis of symmetry defines the possible superconducting states. Their classificaton is done by looking at the way the Cooper-pair wave function changes when rotated or reflected by the operations that leave the crystal unchanged.

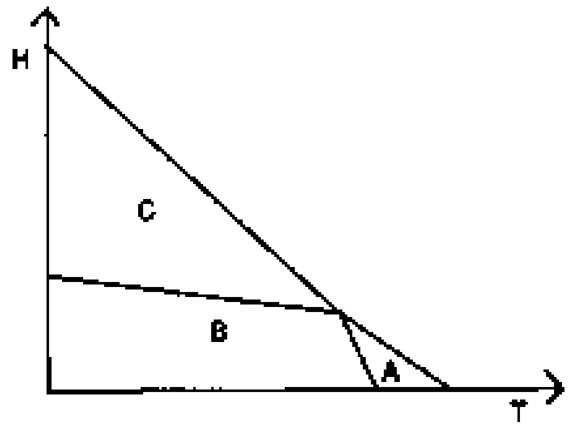

Figure 1: Superconducting phase diagram of UPt_3 in the magnetic field-temperature plane.

The formal analysis in the paper begins by writing down the Hamiltonian for superconductivity, to which is then applied a mean field treatment, defining the gap function and neglecting its fluctuations. This leads to the equation for critical temperature:

$$v\Delta_{s,s'}(n\mathbf{k}) = -\sum_{n'k's_3s_4} V_{s_2s_1s_3s_4}(n\mathbf{k},\mathbf{n}'\mathbf{k}') * \Delta_{s_3s_4}(n'\mathbf{k}')\delta(\epsilon_{n'\mathbf{k}'}) \quad (1)$$

This equation is taken as the basis for discussion of the symmetry of the gap function. From it the authors draw the following four conclusions, which are very difficult to test experimentally and even difficult to go on with a phenomenological analysis:

- the eigenfunctions of the equation transform according to a definite representation of the symmetry group of V;
- if the representation is multidimensional, then the order parameter has more than one component;
- the critical temperature is a monotonically increasing function of the eigenvalue, hence the eigenfunction belonging to the highest eigenvalue is realized in the system;
- generally, there are no symmetries in the set of band indices n, so the eigenvectors will generally have nonzero components in all "directions" in this space, and superconductivity occurs simulaneously in all bands, even in bands where the interaction is repulsive. The relative magnitude of the coefficients of V belonging to different bands may be very different - the gap may be much smaller in some bands than others, and can even change sign;

For the zero-momentum case, the only relevant symmetry group of the Hamiltonian is D_{6h}, with operations that act only on the orbital degrees of freedom ($\mathbf{k}$). There is a finite number of representations, but since the number of eigenfunctions of the eq. (1) is infinite, then there must be many eigenfunctions associated with each representation. The gap function chosen by the system is the function (or pair of functions) with the biggest eigenvalue. The order parameter should be drawn from a single representation of the symmetry group, but since this has proven difficult to explain some of the experiments, several interesting possibilities arose outside this framework.

The authors briefly describe the proposal of a D_6 x $SU(2)$ group, for the case when the spin-orbit coupling is negligible, which will involve the spin degrees of freedom. In this case there are representations that are one or two dimensional in the space group, but three dimensional in the spin rotation group. The paper points out that the spin-orbit coupling term in the uranium wavefunctions cannot be neglected (being on the order of 1 eV), but it is unlikely that the symmetry of $V_{s'ss_3s_4}(n\mathbf{k}, \mathbf{n'k'})$ would be higher than that of the crystal, although this issue has not been investigated.

Another possibility to account for the order parameter is that two different represenations are nearly degenerate, so that even a small difference in the eigenvalues leads to a large difference in the critical temperatures (assuming that the weak coupling theory is valid). The mixed representation picture, presented in the next section, will deal with the issues that might rule out this possibility.

One of the strengths of this review is the discussion of gap functions belonging to different representations. Most of the papers treating this subject give examples of basis functions without explaining how these functions are actually realized. There are several tables of polynomial functions and tight-binding-type functions given in the literature that, as the authors point out, present two flaws. First of all, these tables do not give a complete list of functions, which is needed for a good understanding of all possibilities and whose absence can lead to serious errors. Secondly, those functions are not periodic in reciprocal space, as it is required by translational symmetry (the gap function must be periodic in the crystal momentum space, in the extended zone scheme, and it is imperious to be careful about this for the case of UPt_3, where the sheets of the Fermi surface intersect the brillouin zone). Both aspects are rectified by giving full tables of polynomial functions and functions that form a complete periodic basis for the singlet and triplet representations of D_{6h}, and by explaining how they were constructed and how to use them. The actual gap function is an infinite linear combination of all the functions that transform according to a specific representation. Because of completeness (i.e., all possible functions in a given representation can be written as a combination of the functions in the table) it is sufficient to examine only the functions in the tables in order to find the properties of the gap function.

In the two dimensional singlet case, the gap function is

$$\psi(\mathbf{k}) = a_1 \sum_{i}^{2} c_{i1} F_i^{(1)}(\mathbf{k}) + a_2 \sum_{i}^{2} c_{i2} F_i^{(2)}(\mathbf{k}) \tag{2}$$

The complex coefficients are determined by nonlinear effects. The symmetry of the low-temperature phase is known when we know the values of the a_1 and a_2. The authors give examples of nodal structure of the gap for different coefficients and talk about the symmetry of the states. The paper also deals with the most complicated case of gap structure, that of a two-dimensional triplet representation, also shown in Figure (b).

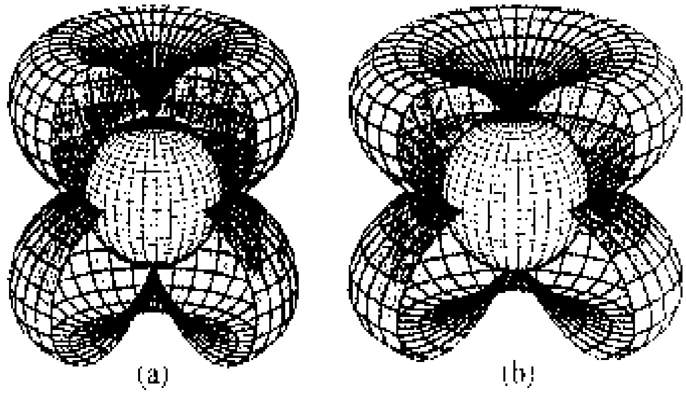

Figure 2: The angular dependence of the gap function for a state with $a_1 = 1$ and $a_2 = i$ is shown on a single spherical Fermi surface. This gap structure is characterized by a combination of an equatorial line node (at $k_z = 0$) and point nodes at the poles ($k_x = k_y = 0$).

2.2 The theories of the superconducting state

The leading theoretical models that aim to explain the multicomponent phase diagram and identify the superconducting phases of UPt_3 are divided into three classes:

- *two-dimensional representations (mostly E_{1g} and E_{2u})*
- *spin-triplet theories (three-dimensional representations)*
- *mixed-representation theories*

The two-dimensional representation and spin-triplet scenarios have traditionally dominated the field, because they give a natural explanation for the split transition. Within this picture, the splitting is caused by the the coupling of superconductivity to magnetism. This was very strongly supported by the pressure data, in which the superconducting transitions arise just when the magnetism disappears. On the other hand, the sound velocities data suggests that the coalescence is illusory and may actually be a crossing, evidence that supports the mixed-representation theory.

Another issue is the in-plane near isotropy of the critical fields. To accomodate this result, which showed up in the neutron-scattering experiments, the two-component theories must invoke the hypothesis that the magnetization is rotated by the field.

The most important features of any theory are the number of components and the nodal structure in the A and B phases, so the authors focus on this piece of information when they review the candidate theories, after presenting the original motivations of each theory. The order parameter is postulated in order to explain experiments, because we are not able to derive it from a microscopic Hamiltonian. Each picture must have the following features: the order parameter should have the same symmetry as the Hamiltonian; it should not involve tuning of the parameters; and it should be consistent will absolutely all experimental data.

3 *UPt_3 experimental results*

From the point of view of comparison to experiment, the most stringent constraints on theory come from detailed measurements of the thermal conductivity κ [10] and the ultrasonic attenuation α [9]. Because of the directional information inherent in these probes, they offer the best chance to distinguish between order parameters that may have only relatively subtle differences [8].

Investigations into the superconducting behavior of UPt_3 are typically divided into three categories, aiming to elucidate the origin of phase multiplicity and to identify the superconducting order parameter of the various phases:

- *phase diagram*
- *nodal structure of the gap*
- *Cooper-pair spin structure and spectroscopy*

One lengthy section in the review article is dedicated to discussing in detail the superconducting phase diagram as a function of magnetic field, temperature, and pressure, in the context of multiple scenarios that explain its various features. Another section is devoted to reviewing the main physical properties of UPt_3, focusing on the B phase - the low-temperature, low-field, zero-pressure phase. The other two phases are not easily accesible because of experimental and theoretical constraints: phase A (high-temperature) because it exists over a very limited temperature range, phase C (high-field) because of the complicated presence of vortices, and the possible high-pressure phase for the impossibility of realizing that high-pressure regime.

A magnetic field has a profound effect in UPt_3, creating five phases: phase C (a vortex phase), and phases A and B, each divided into a Meissner phase and a vortex phase (below and above $H_{c1}(T)$).

The superconducting transition at T_c^+ is immediately visible in many types of experiments, but this does not happen for the lower transition. Apart from specific heat data, the transition at T_c^- between phase A and phase B is seen in thermal expansion and ultrasound measurements. All measurements agree on the value of T_c^- relative to that of T_c^+. Sound velocity has been a key tool in mapping the phase diagram as a function of field and pressure, as shown in the figures. Its relevance at zero field and pressure is that the anomalies observed confirm the specific heat results.

The specific heat of UPt_3 is shown in Figure 5. The data of Brison et al, on a high-quality single crystal annealed for three days at 1200^{o}C, exhibits the following main features: onset of superconductivity at ~ 0.5 K, the appearance of a second transition at a slightly lower temperature, the roughly linear decrease in C/T with temperature, and the large upturn below *0.1* K.

All characteristics of the two transitions are found to converge on the same set of values for all crystals. The difference ΔT_c between the two T_cs commonly referred to as the "splitting" is remarkably invariant. Transitions are very sharp, with no single transition being observed

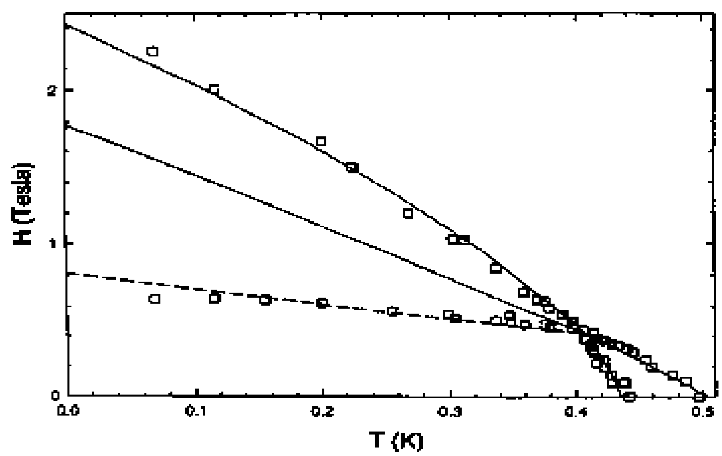

Figure 3: Phase diagram for H in the a-b plane. The data has been obtained in sound velocity measurements [11], and the line is a theoretical fit by Park and Joynt using the E_{1g} gap structure.

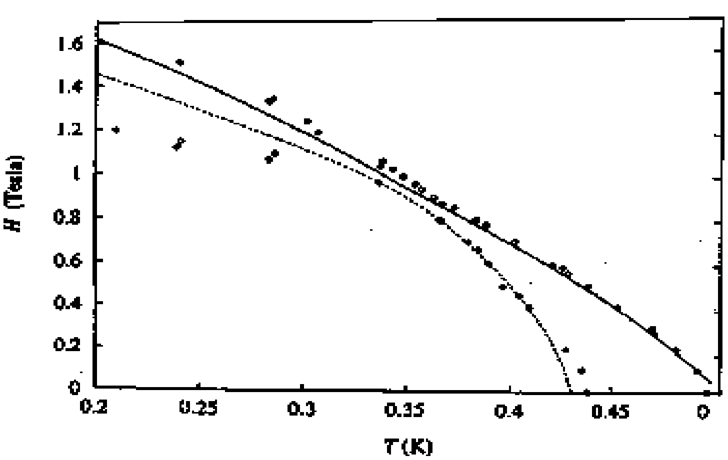

Figure 4: Phase diagram for H along the c-axis. The data has been obtained in sound velocity measurements [11], and the line is a theoretical fit by Park and Joynt using the E_{1g} gap structure.

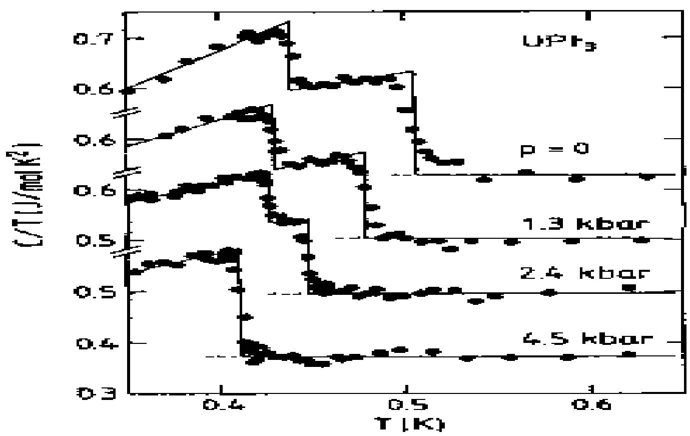

FIG. 13. Specific heat for different hydrostatic pressures plotted as C/T vs T. From Trappmann, Löhneysen, and Taillefer, 1991

Figure 5: Specific heat for different hydrostatic pressures plotted as $\frac{C}{T}$ vs. T.

with a width less than the splitting (50 mK). The published data on single crystals is collected in the table below.

TABLE V. Characteristics of the specific heat of UPt_3 in the vicinity of the superconducting transition in zero applied magnetic field and pressure, for all published data on single crystals. The annealing temperature is in °C, T_c in mK, and C/T in J K^{-2} mol^{-1}.

Reference	Annealing	T_c^-	T_c^+	ΔT_c	γ_N	$\Delta C^-/T_c^-$	$\Delta C^+/T_c^+$	ratio
Hasselbach *et al.* (1989)		434	490	56	0.45	0.10	0.21	0.48
Jin, Carter, *et al.* (1992)	950	464	515	51	0.41	0.12	0.24	0.50
Bogenberger *et al.* (1993)	1200	467	523	56	0.44	0.09	0.22	0.42
Brison *et al.* (1994b)	1200	480	530	50	0.44	0.12	0.23	0.52
Isaacs *et al.* (1995)	1230	460	510	50	0.44	0.11	0.23	0.48
Kimura *et al.* (1995)	1200	530	580	50	0.42	0.09	0.20	0.45
Taillefer, Ellman, *et al.* (1997)		431	492	61	0.43	0.09	0.22	0.41
Kycia (1997)	900	495	545	50	(0.44)	0.13	0.25	0.52

Figure 6: Characteristics of the specific heat of UPt_3

4 *CONCLUSIONS*

UPt_3 is now probably the best studied of any superconducting binary compound, since its physical properties are so exciting and it is easy to prepare in very pure single-crystalline form.

A full understanding of this compound depends on the resolution of the coupling of superconductivity and magnetism. Presently, the weakest link in the picture of UPt_3 is the coupling of superconductivity and magnetism, which is considered to be the origin of the split transition. Is the nature of the magnetic ordering understood? Why is it so poor, and does it affect the coupling to superconductivity? Does the small moment mean that it is a secondary order parameter?

If the questions revolving around this issue have no answer, then the alternative is that the split transition does not rise from this coupling, but from an accidental degeneracy, as considered in different scenarios, (e.g.,the mixed-representation picture). The trouble is that most such scenarios have difficulties producing gap nodal structure consistent with the experiment.

UPt_3 is an exotic superconductor exhibiting multiple superconducting phases. The order parameter of UPt_3 is a problem that was not sorted out. The authors mention the necessity of new probes of the gap structure and the vortex structure, as well as a better understanding of the theory of the low-temperature behavior, including the behavior of the residual normal fluid in the presence of impurities and interactions.

More work still has to be done on solving the issues raised by the phase diagram, especially regarding the coupling of superconductivity with magnetism, and the questions about what is going on at high pressure are still waiting for their answers.

Further work can also be done on orbital degeneracy split by spin-orbit coupling and crystal fields, which was treated crudely or not at all so far. Norman [12] proposes a model of on-U-site pairing which looks promising. The magnetic susceptibilities for all frequencies and wave vectors is still needed as input, and experimental work could push this approach further.

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