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On the interplay of small-moment magnetism and superconductivity in UPt_3

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Abstract

The heavy-fermion compound UPt_3 is the prime candidate for unconventional superconductivity. Various Ginzburg–Landau models have been put forward in order to explain the unusual superconducting phase diagram with three vortex phases in the B–T plane. The most perfected Ginzburg–Landau model is the E-representation model, in which the double superconducting transition in zero field, $\Delta T_c \sim 0.05$ K, is explained by the coupling of a vector superconducting order parameter to a symmetry-breaking field (SBF) ε, where $\Delta T_c \propto \varepsilon$. The natural candidate for the SBF is the weak antiferromagnetically ordered moment m detected at $T_N \sim 6$ K, with $\varepsilon \propto m^2$. By studying UPt_3 doped with small amounts of Pd it is possible to test the SBF model. We here report specific-heat experiments on single-crystalline samples, which show that ΔT_c increases upon doping. The results are compared to recent neutron-diffraction data, which reveal that m^2 also increases upon Pd doping. The analysis of the data within the Ginzburg–Landau approach lends further support for the SBF model.

Keywords: Superconductivity; Antiferromagnetism; Heavy-fermion systems; Ginzburg–Landau model

1. Introduction

In the past decade a host of experiments has demonstrated that the superconducting properties of the heavy-electron material UPt_3 deviate drastically from the standard BCS behaviour. In spite of all the research efforts, the key question whether UPt_3 is a genuine unconventional superconductor, i.e. a superconductor that has a superconducting gap function with a lower symmetry than the Fermi surface, is still not completely settled. The experiments conducted to probe the unconventional superconducting ground state in UPt_3 can roughly be divided into two categories. To the first category belong experiments that probe the structure of the superconducting gap by measuring the temperature variation of the electronic excitation spectrum. The observed temperature variations in the form of power laws of, for instance, the specific-heat [1], the velocity of sound [2] and the thermal conductivity [3], strongly suggest the presence of point nodes and/or line nodes in the gap, as predicted for an unconventional superconductor. However, the relevant temperature regime, $T \ll T_c$, has not been probed reliably yet, especially because the contribution from impurity scattering, which is not easily quantified, obscures the intrinsic behaviour. The second category of experiments is directed towards exploring the multicomponent phase diagram with three vortex phases in the B–T plane [1, 4, 5]. The generic phase diagram, as was obtained, for instance, by dilatometry on a high-quality single-crystalline sample [6, 7], is shown in Fig. 1 for a magnetic field directed along and perpendicular to the hexagonal axis. All phase lines represent second-order transitions (except perhaps the B–C phase line for which a weakly first-order transition cannot be excluded) [7] which allows for a study of the phenomenology of the phase diagram by means of the Ginzburg–Landau (GL) theory. In the past years, a number of GL models have been worked out in order to understand the observed field and pressure variation of the three vortex phases [8]. The most promising model is the so-called

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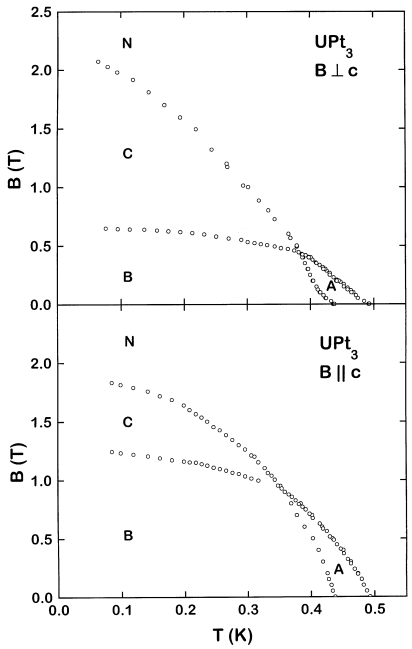

Fig. 1. The superconducting phase diagram of UPt_3 for fields applied perpendicular and parallel to the hexagonal axis as determined by dilatometry [6, 7].

E-representation model, which is based on the coupling of a two-dimensional superconducting order parameter to a symmetry-breaking field (SBF) [8–11]. Here the order parameter can be expressed as the coefficients of the E_1 or E_2 basis functions constituting the gap. The underlying mechanism is that a weak SBF lifts the degeneracy of the order parameter, which results in two superconducting phases in zero field. In an alternative model, the so-called AB-model [12], the double superconducting transition is understood as arising from the coupling of two nearly degenerate one-dimensional basis functions (A_1, A_2, B_1 or B_2 representation). These different GL scenarios are still subject of lively debates.

The key-issue in the E-model is to identify the SBF and to proof that it indeed couples to superconductivity. The most likely candidate for the SBF is the weak antiferromagnetic order which sets in below the Néel temperature T_N of about 6 K [13]. The tiny ordered moment amounts to $m = 0.02 \pm 0.01\ \mu_B$/U-atom (for $T \to 0$ K) and is directed along the a^*-axis. The magnetic ordering consists of a doubling of the nuclear unit cell along the a^*-axis, which results in an orthorhombic distortion. Within the E-model [8–11] the splitting of the superconducting transition $\Delta T_c = T_c^+ - T_c^-$ is proportional to the strength of the SBF or $\Delta T_c \propto \varepsilon$, where $\varepsilon \propto m^2$. Direct evidence for the coupling of ΔT_c and m^2 was deduced from specific-heat [14] and neutron-diffraction [15] experiments under hydrostatic pressure. It was observed that both ΔT_c, determined by specific heat, and $m^2(T_c)$, measured by neutron diffraction, vary linearly with pressure and vanish at a critical pressure $p_c \sim 3$ kbar. However, T_N did not change noticeably under pressure which puts some question marks as to the interpretation of the neutron-scattering data. A second candidate for the symmetry breaking field is the weak incommensurate structural modulation that was detected by transmission electron microscopy [16]. As-cast samples of UPt_3 show structural inhomogeneities on a length scale of the order of the superconducting coherence length $\xi \sim 200$ Å. Specific-heat data taken on these samples reveal a broad superconducting transition of width ΔT_c. In the annealed samples the structural modulation is well developed with a domain size $d \approx 10\,000$ Å $\gg \xi$ and the two superconducting transitions are well resolved in the specific heat. This shows that the structural modulation affects superconductivity, but, so far, it could not be correlated to the size of ΔT_c. The appealing possibility that the weak structural modulation permits the appearance of two nearly-degenerate order parameters as required in the AB-model [12] has not been documented so far.

The purpose of the present paper is to investigate in more detail the E-representation model with the weak magnetic moment acting as the SBF. By performing specific-heat experiments on polycrystalline samples Vorenkamp and collaborators [17] showed that ΔT_c can be increased by substituting small amounts of Pd for Pt in UPt_3. This then directly prompted the question whether the enhancement of ΔT_c is due to an increase of the ordered moment *m*. Since it was known [18] that the $U(Pt_{1-x}Pd_x)_3$ pseudobinaries exhibit long-range antiferromagnetic order in the concentration range $x =$ 0.02–0.07, with a maximum $T_N = 5.8$ K and an ordered moment $m = 0.6 \pm 0.2\ \mu_B$/U-atom at optimum doping ($x = 0.05$), an increase of the tiny ordered moment upon Pd doping seemed probable. In order to follow $m(x)$, a detailed study of the magnetism in $U(Pt_{1-x}Pd_x)_3$ pseudobinaries ($x \leqslant 0.05$), by means of neutron-diffraction and μSR techniques is in progress [19–21]. The idea is to test the relation $\Delta T_c \propto m^2$, where both quantities are obtained on the same single-crystalline samples. The values for ΔT_c were obtained by specific-heat experiments carried out for $x \leqslant 0.002$. We here report the first results.

2. The E-representation model with SBF

The irreducible representations for the superconducting gap with the appropriate D_{6h} symmetry of UPt_3 have been tabulated by e.g. Yip and Garg [22]. For a two-dimensional (2D) representation with even parity, E_{1g} or E_{2g}, or odd parity, E_{1u} or E_{2u}, the superconducting gap function is given by $\Delta_E(\boldsymbol{k}) = \eta_x \Gamma_{E,x}(\boldsymbol{k}) + \eta_y \Gamma_{E,y}(\boldsymbol{k})$, where $\Gamma_{E,x}$ and $\Gamma_{E,y}$ are the basis functions for the relevant 2D-representation. The complex vector $\boldsymbol{\eta} = (\eta_x, \eta_y) = (|\eta_x| e^{i\phi_x}, |\eta_y| e^{i\phi_y})$ determines the order parameter. Within

the most simplest GL model with SBF, the free energy functional is written as the sum of three terms [8–11]:

$$F = F_S + F_M + F_{SM}. \quad (1)$$

Here F_S is the free energy functional of the superconductor.

$$F_S = \alpha_S(T - T_c)|\eta|^2 + \tfrac{1}{2}\beta_1|\eta^4| + \tfrac{1}{2}\beta_2|\eta^2|^2, \quad (2)$$

where the coefficients α_S, β_1 and β_2 are stability parameters. The contribution from the magnetic order to the free energy is given by

$$F_M = \alpha_M(T - T_N)m^2 + \tfrac{1}{2}\beta_M m^4, \quad (3)$$

where $\boldsymbol{m} = m(1, 0, 0)$ is the small ordered moment oriented along a principal axis in the basis plane ($T \leqslant T_N$) and α_M and β_M are stability parameters. The mixing term of magnetic order and superconductivity can be written as

$$F_{SM} = \gamma|\boldsymbol{m}\cdot\boldsymbol{\eta}|^2 + \alpha m^2|\boldsymbol{\eta}|^2, \quad (4)$$

where $\varepsilon = \gamma m^2$ is the SBF. By minimizing the free energy it follows that the single superconducting transition at T_c splits into two transitions at T_c^+ and T_c^-, where

$$\Delta T_c = T_c^+ - T_c^- = \frac{|\gamma|}{\alpha_S}\frac{\beta_1 + \beta_2}{2\beta_2}m^2. \quad (5)$$

The ratio $(\beta_1 + \beta_2)/2\beta_2$ can be determined from the calculated jumps in the specific heat at T_c^+ and T_c^-:

$$\frac{\Delta c(T_c^-)/T_c^-}{\Delta c(T_c^+)/T_c^+} = 1 + \frac{\beta_2}{\beta_1}. \quad (6)$$

The weak-coupling estimate for β_2/β_1 is 0.5. As it is our purpose to verify Eq. (5) by experiments, one also needs, besides values for ΔT_c and β_2/β_1, which can be deduced from the specific-heat data, and the value of m, which follows from the neutron-diffraction experiments [19], an estimate for the parameter $|\gamma|/\alpha_S$. We shall discuss this in Section 4.

3. Specific heat of U(Pt,Pd)$_3$

The specific-heat experiments have been carried out on annealed single-crystalline samples of $U(Pt_{1-x}Pd_x)_3$ for $x = 0.000$, 0.001 and 0.002. The specific heat was measured employing the relaxation method. The mass of the samples amounted to 80 mg. A full account of the experimental details will be given elsewhere. Annealed single-crystalline samples, cut from the same batches, show residual resistivity values for a current along the a-axis, $\rho_{o,a}$, of 0.5, 1.6 and 2.4 μΩ cm for $x = 0.000$, 0.001 and 0.002, respectively. T_c^+ measured resistively is suppressed linearly with Pd contents at a rate of 0.85(5) K/ at% Pd, and, because $\rho_{o,a}$ increases linearly with Pd contents, at a rate of 0.073(2) K/μΩ cm. The specific-heat data are shown in Fig. 2. For pure UPt_3 $T_c^+ = 0.543(2)$ K and $T_c^- = 0.489(2)$ K and $\Delta T_c = 0.054(4)$ K. The most striking feature of the data in Fig. 2 is the large increase of ΔT_c upon doping for these small amounts of Pd. For 0.2 at% Pd, ΔT_c has doubled compared to pure UPt_3. The resulting values for T_c^+, T_c^-, ΔT_c, $\Delta c(T_c^+)/T_c^+$, $\Delta c(T_c^-)/T_c^-$ and β_2/β_1 are collected in Table 1, where we made use of an equal entropy construction in order to determine the ideal values for the jumps in the specific heat. The values for ΔT_c are in good agreement with the data taken on polycrystalline samples [17]. The ratio β_2/β_1 decreases upon doping, which is attributed to the Kondo-hole effect due to Pd impurities, which results in a strong pair-breaking.

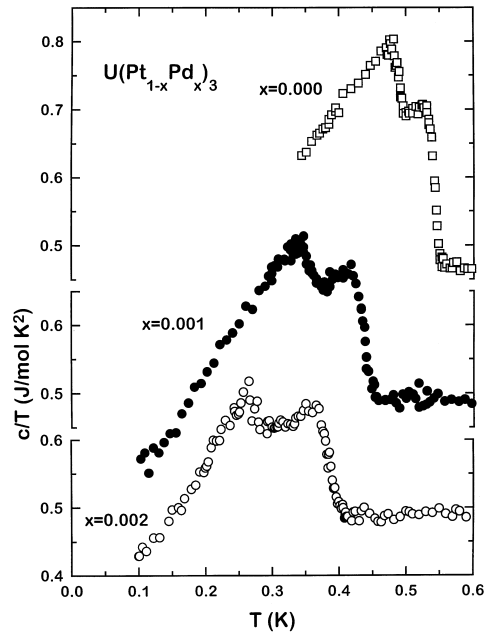

Fig. 2. Specific heat of single-crystalline $U(Pt_{1-x}Pd_x)_3$ for $x = 0.000$, 0.001 and 0.002.

4. Testing the relation $\Delta T_c \propto m^2$

Neutron-diffraction experiments were carried out on single-crystalline samples of the $U(Pt_{1-x}Pd_x)_3$ series with $x \leqslant 0.05$ [17]. In the case of $x = 0.001$ and $x = 0.002$, the experiments were carried out on unannealed samples. The neutron intensity grows quasi-linearly below $T_N \sim 6$ K, just as observed for pure UPt_3. T_N does not change for $x \leqslant 0.01$. The values for $m(T_c^+)$ are 0.018 ± 0.003 and 0.034 ± 0.006 μ_B/U-atom, for $x = 0.001$ and

Table 1
Parameters deduced from the specific heat of single-crystalline $U(Pt_{1-x}Pd_x)_3$ for $x = 0.000$, 0.001 and 0.002. The ratio β_2/β_1 is calculated with the help of Eq. (6)

$U(Pt_{1-x}Pd_x)_3$	T_c^+ (K)	T_c^- (K)	ΔT_c (K)	$\Delta c(T_c^+)/T_c^+$ (J/mol K^2)	$\Delta c(T_c^-)/T_c^-$ (J/mol K^2)	β_2/β_1
$x = 0.000$	0.543(2)	0.489(2)	0.054(4)	0.257(9)	0.353(8)	0.37(2)
$x = 0.001$	0.437(2)	0.355(2)	0.082(4)	0.222(8)	0.280(9)	0.26(2)
$x = 0.002$	0.384(2)	0.276(3)	0.108(5)	0.189(9)	0.213(8)	0.13(2)

Table 2
Residual resistivity $\rho_{o,a}$, the ordered moment $m(T_c^+)$ [19, 20] and the ratio $|\gamma|/\alpha_S$ calculated with the help of Eq. (5) (see text) for single-crystalline $U(Pt_{1-x}Pd_x)_3$ with $x = 0.000$, 0.001, 0.002 and 0.005

$U(Pt_{1-x}Pd_x)_3$	$\rho_{o,a}$ ($\mu\Omega$ cm)	$m(T_c^+)$ (μ_B/U-atom)	$\lvert\gamma\rvert/\alpha_S$ (K/(μ_B/U-atom)^2)
$x = 0.000$	0.5	0.02(1)[a]	73(73)
$x = 0.001$	1.6	0.018(3)	105(24)
$x = 0.002$	2.4	0.034(6)	21(8)
$x = 0.005$	6.2	0.050(5)[b]	–

[a] Value taken from Ref. [11].
[b] $T_c^+ = 0.126$ K.

0.002, respectively, which should be compared to the value $m(T_c^+) = 0.02 \pm 0.01$ μ_B/U-atom, reported in the literature for pure UPt_3 (so far, we did not perform neutron-diffraction experiments on the pure UPt_3 sample used in the specific-heat experiment). Values for $m(T_c^+)$ are collected in Table 2. We conclude that m grows as function of Pd contents. Because we have only few data points and because of the large error bar in the literature value of m for pure UPt_3, the data do not allow for a reliable estimate of the functional relation between m and x.

In the case of the pressure experiments [14, 15], it was found that $\Delta T_c(p)$ is directly proportional to $m^2(p)$. The proportionality factor $(|\gamma|/\alpha_S)(\beta_1 + \beta_2)/2\beta_2$ was taken pressure independent, which is supported, as far as the ratio β_2/β_1 is concerned, by the specific-heat experiments [14]. This is consistent with pair-weakening being the dominant mechanism for the suppression of T_c by hydrostatic pressure. In the case of Pd doping, however, $\Delta T_c(x)$ is not directly proportional to $m^2(x)$ and the effect of pair breaking on the proportionality factor in Eq. (5) should be taken into account. Using the experimental values for ΔT_c, $\Delta c(T_c^+)/T_c^+$, $\Delta c(T_c^-)/T_c^-$, β_2/β_1 (Table 1) and $m(T_c^+)$ (Table 2) we have calculated $|\gamma|/\alpha_S$ using Eq. (5). The ratio $|\gamma|/\alpha_S$ shows a tendency to decrease with increasing Pd content, albeit with a considerable error bar, due to the uncertainty in the value of m for pure UPt_3. Assuming that the coupling parameter $|\gamma|$ is a constant, the decrease of $|\gamma|/\alpha_S$ is attributed to an increase of α_S. In the standard GL theory for superconductors $\alpha_S \propto 1/\xi^2$ [23], where ξ is the superconducting coherence length. An estimate of the variation of ξ with impurity contents can be obtained from the Pippard relation $1/\xi(x) = 1/\xi_0 + 1/l(x)$ [23], where ξ_0 is the coherence length of the pure material and l is the electronic mean free path. For our pure UPt_3 sample we estimate $l = 2200$ Å and $\xi_0 = 200$ Å [18]. Upon doping, l and ξ will decrease, but, in order to give a reliable estimate of $\xi(x)$, measurements of the upper critical field B_{c2} are needed. The tendency of $|\gamma|/\alpha_S$ to decrease with Pd content is naturally explained, since $|\gamma|/\alpha_S \propto |\gamma|\xi^2$. Therefore, we conclude, albeit with caution, that the data in Tables 1 and 2 are consistent with the E-representation model with magnetic order as the symmetry-breaking field. We have to bear in mind, however, that the specific-heat experiments have been performed on annealed samples, while the samples used for neutron diffraction were unannealed.

5. Concluding remarks

We have studied the double superconducting transition in $U(Pt_{1-x}Pd_x)_3$ for $x \leqslant 0.002$ by specific-heat experiments on single-crystalline samples. Upon doping pure UPt_3 with Pd, a pronounced increase of the splitting $\Delta T_c(x)$ is observed. We have correlated $\Delta T_c(x)$ with $m^2(x)$, where m is the weak antiferromagnetically ordered moment ($T_N \sim 6$ K) determined by neutron diffraction. Our findings lend further support for the E-representation (even or odd parity) Ginzburg–Landau model for the double superconducting transition with the weak antiferromagnetic moment acting as the symmetry-breaking field. The analysis would benefit from a more reliable estimate of the ordered moment in pure UPt_3 and from knowledge of the superconducting coherence length in the doped samples. Measurements of the upper critical field to access ξ are underway.

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