Physica 127B (1984) 442–447
North-Holland, Amsterdam

SPIN FLUCTUATIONS AND SUPERCONDUCTIVITY IN UPt_3

A. DE VISSER, J.J.M. FRANSE and A. MENOVSKY
Natuurkundig Laboratorium der Universiteit van Amsterdam, Valckenierstraat 65, 1018 XE Amsterdam, The Netherlands

T.T.M. PALSTRA
Kamerlingh Onnes Laboratorium der Rijksuniversiteit Leiden, Nieuwsteeg 18, 2300 RA Leiden, The Netherlands

Electrical resistivity, ac-susceptibility and specific heat measurements on Czochralski-grown single-crystalline UPt_3 reveal a transition into a superconducting state below 0.48 K. For the temperature dependence of the upper critical field, dB_{c2}/dT, a value of -4.4 T/K is found. A numerical analysis of the normal-state specific-heat data below 20 K results in a large $T^3 \ln(T/T^*)$ contribution, indicating pronounced spin-fluctuation effects. The coexistence of superconductivity and spin fluctuations makes UPt_3 a unique material.

1. Introduction

The intermetallic compound UPt_3 (hexagonal $MgCd_3$-type of structure) is attracting a lot of interest because of its unusual low-temperature properties.

Susceptibility measurements on polycrystalline samples, in a temperature range below 200 K [1] and below 1000 K [2], display more or less Curie-Weiss behaviour down to 16 K. Values for the effective moment, μ_{eff}, vary from 2.61 and 2.85 μ_B per mole f.u. [1] up to 3.0 ± 0.1 μ_B per mole f.u. [2]. Around 16 K the susceptibility curves reveal a pronounced maximum.

In single-crystalline samples the magnetic parameters turn out to be strongly anisotropic: the low temperature maximum in the susceptibility is only present for field directions along the a- and b-axis [2]; magnetization curves up to 35 T [2, 3] at 4.2 K and 1.5 K reveal a strongly non-linear behaviour along the a- and b-direction (giving a magnetic moment of 0.9 μ_B per mole f.u. at 35 T), whereas at 20 K and 77 K a nearly linear relationship between magnetic moment and applied field is observed.

Although these data could be indicative of an arrangement of magnetic moments in a complex low-temperature magnetic structure, no sign of magnetic order near 16 K is found in the specific heat [2]. In the specific heat a pronounced $T^3 \ln(T/T^*)$ contribution is observed, nearly field independent below 5 T, together with an unusually large value for the coefficient of the electronic term ($\gamma = 422$ mJ/mole f.u. $\cdot$ K^2) [2, 4].

Similar low-temperature anomalies in specific heat and susceptibility are found in the spin-fluctuation compounds UAl_2 and $TiBe_2$ [5–8]. Therefore it is more likely to interpret the UPt_3 data in terms of spin-fluctuation phenomena. Such a description is supported by resistivity measurements along the different crystallographic directions [9]: a sharp rise at low temperatures is followed by a negative curvature towards the temperature axis and a tendency to saturate at large values in the room-temperature region (ρ reaches 238 $\mu\Omega$ cm and 132 $\mu\Omega$ cm for the basal plane and hexagonal axis, respectively). No sign of magnetic order is observed in the resistivity data either.

Superconductivity has first been observed by Stewart et al. [4]. From specific heat [4] and ac-susceptibility measurements [this work] there is convincing evidence for a transition into a bulk superconducting state near 0.5 K. Such a coexistence of superconductivity and spin fluctuations, in combination with a wealth of anisotropic magnetic properties, makes UPt_3 a unique exotic material. In view of the large electronic term in the specific heat, the superconductor UPt_3 can be

0378-4363/84/$03.00

placed in a new class of materials, which are indicated as 'heavy fermion' superconductors. Other members of this class are $CeCu_2Si_2$ [10], UBe_{13} [11], U_6Fe [12] and U_6Co [13].

In this contribution we report on electrical resistivity, ac-susceptibility and specific heat experiments on the superconducting transition, including the temperature dependence of the upper critical field. In addition the specific heat of a bulk single-crystalline sample up to 20 K is presented. A first attempt is made to give a qualitative analysis of the critical field data.

2. Experimental

Single-crystalline UPt_3 was prepared in a tri-arc melting equipment by the Czochralski method [14]. All single-crystalline samples described in this work were machined from the same melt, a cylinder of approximately 6 mm diameter and 20 mm length. Experiments were performed before and after annealing the samples at 900 °C during one day. Residual resistance ratio's (i.e. $\rho(300\ \mathrm{K})/\rho_0$) were 33, 33 and 37 for the a-, b- and c-direction, before annealing; after annealing the ρ_0-values decreased to 6.2, 3.0 and 1.7 $\mu\Omega$ cm, leading to residual resistance ratio's of 38, 79 and 78, respectively. The differences in ρ_0-values, possibly due to small deviations from stoichiometry, are indicative of remaining inhomogeneities after annealing.

Low-frequency ac-susceptibility ($B_{ac} < 2 \times 10^{-5}$ T, $f = 10.9$ Hz) in external fields up to 0.5 T, and electrical resistivity (standard four-point dc-method, $I < 2$ mA) experiments were performed down to 0.30 K. The specific heat was measured with an adiabatic method down to 0.13 K.

3. Results

Superconductivity in our case was first observed on three annealed samples (see table I: sample #1, #2 and #3) in electrical resistivity measurements along different crystallographic directions. From our previous experiments [9] it was evident that the temperature dependence of the resistivity only follows a T^2-behaviour (characteristic for a spin-fluctuation system [15]) at approaching 1.3 K. In our investigation down to 0.30 K we found a drop to zero resistivity in all three samples between 0.47 K and 0.53 K (see fig. 1).

Table I
Superconducting transition temperature, T_c, and width of the superconducting transition temperature, ΔT_c, for Czochralski-grown UPt_3 (all sample #1 till #8 out of one single-crystalline batch) and for arc-melted polycrystalline UPt_3 (samples #9 and #10). In the ac-susceptibility experiments T_c- and ΔT_c-values are defined at the 50% point and between the 10 and 90% points, respectively.

Sample		T_c (K)	ΔT_c (K)	ρ_0 ($10^{-8} \times \Omega$ m)	Experiment
#1	unannealed	—		7.2	ρ_{dc}(a-axis)
	annealed	0.47–0.53		6.2	ρ_{dc}(a-axis)
#2	unannealed	—		7.2	ρ_{dc}(b-axis)
	annealed	0.47–0.53		3.0	ρ_{dc}(b-axis)
#3	unannealed	—		3.6	ρ_{dc}(c-axis)
	annealed	0.47–0.53		1.7	ρ_{dc}(c-axis)
#4	unannealed	0.40	0.05		χ_{ac}
#5	unannealed	0.43	0.05		χ_{ac}
#6	annealed	0.48	0.05		χ_{ac}(a-axis)
#7	annealed	0.48	0.10		χ_{ac}(c-axis)
#8	unannealed	0.43			specific heat
#9	annealed (arc-melted)	0.50	0.10		χ_{ac}
#10	annealed (powdered)	0.45	>0.10		χ_{ac}

A second indication of superconductivity was obtained by ac-susceptibility measurements on two unannealed samples (#4 and #5): large diamagnetic signals were observed below 0.40 K and 0.43 K, respectively. The annealed samples, #6 and #7, give a superconducting transition temperature, T_c, of 0.48 K, consistent with the values that were derived from the resistivity measurements on samples #1, #2 and #3. Obviously, annealing of the samples increases the T_c-values. The temperature dependence of the upper critical field, $B'_{c2} \equiv -\mathrm{d}B_{c2}/\mathrm{d}T$ was studied in applied magnetic fields up to 0.5 T, along the a- and c-directions (see fig. 2). For sample #6 $B'_{c2} = 4.4$ T/K (field along the a-axis), a value comparable to B'_{c2}-values for other 'heavy fermion' superconductors (see table II).

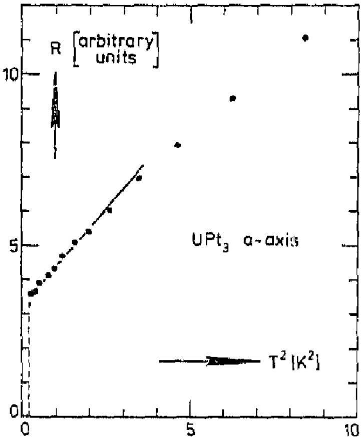

Fig. 1. Electrical resistivity (in arbitrary units) versus T^2 (sample #1), superconductivity is observed below 0.47 K.

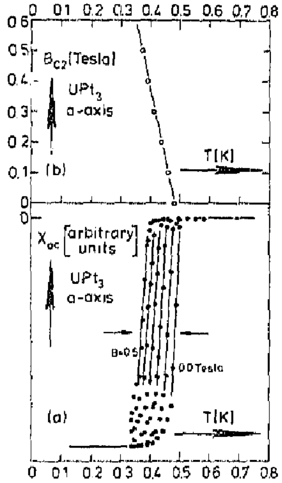

Fig. 2. (a) Ac-susceptibility (in arbitrary units) in zero magnetic field and in fields of 0.1, 0.2, 0.3, 0.4 and 0.5 T (field direction along the a-axis) for the sample #6; and (b) the upper critical field as function of temperature: T_c is defined at the 50% point (indicated by arrows in (a)).

Table II
Some important parameters which characterize the 'heavy fermion' superconductors UPt_3 [this work], $CeCu_2Si_2$ ([17] and references therein, sample No. 7), UBe_{13} [11], U_6Fe [12] and U_6Co [13]. Fermi wave numbers calculated for the normal state and superconducting state are denoted by k_F^n and k_F^s, respectively. For UPt_3 and $CeCu_2Si_2$ the Fermi velocity, v_F, and the effective mass, m^*, are calculated from k_F^s, whereas for UBe_{13} calculations are made with k_F^n. The meaning of the other symbols is explained in the text.

Parameter	Units	UPt_3	$CeCu_2Si_2$	UBe_{13}	U_6Fe	U_6Co
γ	(mJ/mole f.u. · K^2)	422	1006	1100	155	126
T_c	(K)	0.48	0.64	0.85	3.8	2.3
B'_{c2}	(T/K)	4.4	5.8	25.7	3.4	3.7
ρ_0	(10^{-7} Ωm)	3.0	3.5		50	
Z	(per atom)	2.4	2.5	0.81		
k_F^n	(10^{10} m^{-1})	1.6	1.6	1.36		
k_F^s	(10^{10} m^{-1})	1.1	1.7			
v_F	(10^3 m/s)	6.8	8.7	8.2		
m^*	(m_0)	180	220	192		
ξ_0	(10^{-8} m)	2.0	1.9			
l	(10^{-8} m)	3.6	1.2			
λ_L	(10^{-7} m)	3.6	2			
κ_{GL}		23	22			

Evidence for bulk superconductivity was derived from specific heat experiments on an unannealed single-crystalline sample (#8, see fig. 3). Here, the onset of the superconductivity is at 0.43 K, and the maximum in the specific heat

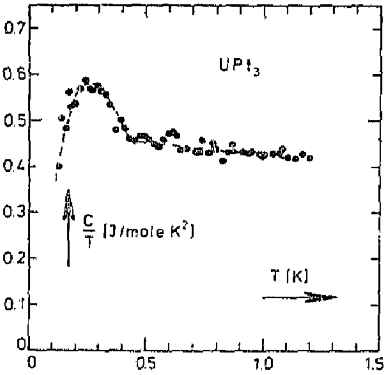

Fig. 3. C/T versus T (sample #8). The broken line is a guide to the eye.

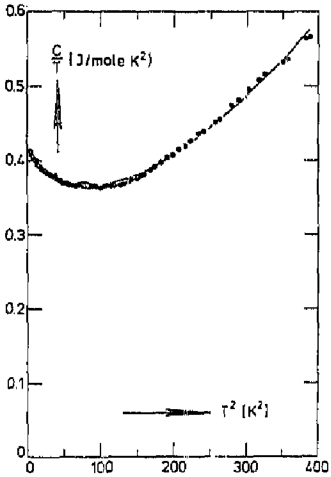

Fig. 4. C/T versus T^2 for UPt$_3$ (sample #8). The full line represents a fit to all data points in the temperature interval 1.2–20 K, see table III.

discontinuity is at 0.26 K. The value of $\Delta C/\gamma T$ at 0.26 K is 0.35, i.e. one quarter of the value predicted from BCS theory: 1.43. This could be illustrative of the still not perfect sample quality of the unannealed samples. For a flux-grown sample a value for $\Delta C/\gamma T$ at 0.40 K of 0.48 has been obtained [4].

Specific heat data on our sample in a temperature range up to 20 K are shown in fig. 4.

4. Analysis and discussion

The above-presented evidence of bulk superconductivity in our Czochralski-grown single-crystalline samples is consistent with the strong evidence presented by Stewart et al. [4] on flux-grown samples. From their electrical resistivity, ac-susceptibility and specific heat data a T_c^{onset} of 0.54 K, 0.53 K and 0.54 K, respectively, is obtained. Two other observations support the evidence for bulk superconductivity: (i) annealed polycrystalline samples of the neighbouring phases of UPt$_3$ (UPt$_2$ and UPt$_5$) that could be present in spurious amounts, showed no traces of superconductivity down to 0.30 K, and (ii) a single crystalline sample (#6), an annealed polycrystalline sample (#9) and an annealed powdered sample (#10) all gave a diamagnetic signal of the same magnitude as a full Meissner effect, after the necessary corrections for demagnetizing factors. A hard proof experiment, i.e. the observation of the bulk Meissner effect, is in preparation.

In an attempt to give a qualitative analysis of the superconducting properties we follow the approach that successfully has been applied to A15 superconductors [16] and to CeCu$_2$Si$_2$ [17]. Under two crude assumptions: (i) a spherical Fermi surface, ignoring possible anisotropy effects, and (ii) the possibility that UPt$_3$ is a strong-coupling superconductor is discarded, the Fermi wave number, k_F, can be derived from the temperature dependence of the upper critical field, according to the relation:

$$B'_{c2} \approx 7.95\times10^{32}(\gamma^2 T_c/k_F^4) + 4780\gamma\rho_0 \qquad (1)$$

Inserting our experimental results (sample #6) for B'_{c2} (= 4.4 T/K), T_c (= 0.48 K), the residual resistivity ρ_0 (although not measured on sample #6, we may estimate it at $\approx 3\times10^{-8}\ \Omega m$ [9]) and the electronic term in the specific heat γ (= 9.94×10^3 J/(m^{3}K^2) with $V_{\text{mole}} = 4.243\times10^{-5}$ m^3), we obtain a k_F-value of $\approx 1.1\times10^{10}$ m^{-1}. Within the Fermi-liquid model we then derive a Fermi velocity $v_F = (k_B^2 k_F^2)/(3\hbar\gamma) \approx 6.8\times10^3$ m/s and an effective mass $m^* = \hbar k_F/v_F \approx 180\ m_0$, values that are of the same order of magnitude as those derived for other 'heavy fermion' superconductors (see table II). The clean limit value for B'_{c2}, calculated with the above given values for γ, T_c and k_F, amounts to 3.3 T/K.

In a Fermi-liquid description the electrons in the normal state and the quasiparticles that take part in the superconducting state are described with one and the same Fermi wave number. In the normal state a value for k_F can be derived from the resistivity data, as has been done for CeCu$_2$Si$_2$ [18] and UBe$_{13}$ [11]. According to Friedel [19] the maximum in the resistivity is thought to be due to incoherent scattering of the conduction electrons at the uranium ions:

$$\rho_{\max} = (2l+1)2hx/(e^2 k_F Z). \qquad (2)$$

Here $l = 3$ for f-electrons, Z is the number of conduction electrons per atom, x ($=\frac{1}{4}$) is the concentration of scattering centers and $k_F = (3\pi^2 Z/\Omega)^{1/3}$ with Ω ($= 17.62$ Å^3) as the mean volume per atom. With the experimental value of $\rho_{\max}$ in the order of $240 \times 10^{-8}\,\Omega$m [9], we calculate $Z \approx 2.4$ and derive a k_F-value of $\approx 1.6 \times 10^{10}\,\mathrm{m}^{-1}$, which differs not too much from the above given value.

Accepting this type of analysis we are able to estimate some important parameters which characterize the superconducting state of UPt$_3$. Inserting the above values for k_F ($\approx 1.1 \times 10^{10}\,\mathrm{m}^{-1}$), T_c, ρ_0 and γ in the relations given in ref. 16, we derive the BCS-coherence length $\xi_0 \approx 2.0 \times 10^{-8}$ m, the mean free path $l \approx 3.6 \times 10^{-8}$ m and the London penetration depth ($T \to 0$) $\lambda_L \approx 3.6 \times 10^{-7}$ m; $\xi_0 \approx l$ means that our samples are nor in the dirty limit ($l \ll \xi_0$) nor in the clean limit ($l \gg \xi_0$), as could be expected from the observed ρ_0-values [9]. The Ginzburg–Landau parameter, κ_{GL}, is found to be about 23, a value characteristic for type II-superconductivity. Corresponding parameters for CeCu$_2$Si$_2$ are listed in table II.

At this point we want to stress once more that the above presented calculations could only be made under crude assumptions. Especially, ignoring anistropy effects must be far from appropriate for UPt$_3$, in view of its anisotropic magnetic properties. In particular our observation that one of the samples (#7), although exhibiting a rather broad transition, seems to have an increasing slope of the upper critical field with decreasing temperature ($B'_{c2} = 3.6$ T/K at T_c and $B'_{c2} = 6.0$ T/K near 0.40 K, with the magnetic field applied along the c-axis) asks for further detailed investigations.

Two normal-state properties of our Czochralski-grown single-crystalline samples are indicative for spin-fluctuation phenomena: a $T^3 \ln(T/T^*)$-contribution to the specific heat [20] and a T^2-term in the resistivity [15].

To start with the first: it was shown [6, 8] that the specific heat data of the spin-fluctuation compounds TiBe$_2$ and UAl$_2$ could well be fit to the equation

$$C/T = \gamma + \beta^* T^2 + \delta T^2 \ln T, \qquad (3)$$

where $\beta^* = \beta - \delta \ln T^*$, with β the usual phonon coefficient. A similar type of specific heat curve was obtained by Frings et al. [2, 21] on polycrystalline UPt$_3$. Recently, Stewart et al. [4] published results for the specific heat data on their flux-grown single-crystalline UPt$_3$. We performed a least-square fit to 85 data points that we obtained in the temperature range 1.2–20 K (see fig. 4), using eq. (3), in different temperature intervals. The results of these fits are listed in table III. The values for the coefficients in eq. (3) do not change dramatically for the different temperature intervals, indicating that eq. (3) gives a proper description of the low-temperature specific heat of UPt$_3$. The results agree fairly well with a similar fit on polycrystalline UPt$_3$ [21].

A second evidence for spin-fluctuation phenomena follows from the resistivity data down to T_c. It was shown that the overall resistivity could be interpreted in terms of a spin-fluctuation model although a T^2-term was observed only at approaching 1.3 K [9]. Our new low-temperature data, plotted in fig. 1 versus T^2, reveal a similar observation: a straight line, denoting a T^2-dependence, can be drawn through the data, but in view of the scatter this

Table III
Coefficients derived from a fit of the specific heat data of UPt$_3$ (sample #8) to the relation $C/T = \gamma + \beta^* T^2 + \delta T^2 \ln T$ in several temperature intervals.

Interval	γ (mJ/Mole f.u. · K^2)	β^* (mJ/mole f.u. · K^4)	δ (mJ/mole f.u. · K^4)
1.2–20 K	422	−3.72	1.38
1.2–15 K	423	−3.80	1.41
1.2–10 K	421	−3.53	1.30
6–10 K	424	−3.78	1.40
8–20 K	413	−3.45	1.30
1.2–17 K	422	−4.18	1.54

* From ref. [21].

can not be called 'hard proof'. It must be mentioned that the resistivity of UAl_2 [22] follows a T^2-dependence only in a very limited temperature range up to 2 K.

In summary we conclude from our resistivity, ac-susceptibility and specific heat experiments that we have strong evidence for the coexistence of spin fluctuations and bulk superconductivity near 0.48 K in Czochralski-grown single-crystalline UPt_3. A first qualitative analysis of the critical field data points to a characterization of UPt_3 as a 'heavy fermion' superconductor. The coexistence of superconductivity and spin fluctuations, in combination with its wealth of anisotropic magnetic properties and the availability of qualitatively good single-crystalline samples, makes UPt_3 a unique exotic material.

Acknowledgements

We would like to thank J.A. Mydosh and co-workers for putting their experimental equipment to our disposal and J.C.P. Klaasse, P.R. Nugteren and E. Louis for their experimental assistance. This work was part of the research program of the Stichting FOM (Foundation for Fundamental Research of Matter).

References

[1] W.D. Schneider and C. Laubschat, Phys. Rev. B23 (1981) 997.
[2] P.H. Frings, J.J.M. Franse, F.R. de Boer and A. Menovsky, J. Magn. Magn. Mat. 31–34 (1983) 240.
[3] J.J.M. Franse, Helv. Phys. Acta 56 (1983) 131; J. Magn. Magn. Mat. 31–34 (1983) 819.
[4] G.R. Stewart, Z. Fisk, J.O. Willis and J.L. Smith, Phys. Rev. Lett. 52 (1984) 679.
[5] A.J. Arko, F.Y. Fradin and M.B. Brodsky, Phys. Rev. B8 (1973) 4104.
[6] G.R. Stewart, A.L. Giorgi, B.L. Brandt, S. Foner and A.J. Arko, Phys. Rev. B28 (1983) 1524.
[7] F. Acker, R. Huguenin, M. Pelizzone and J.L. Smith, Phys. Rev. B24 (1981) 5404.
[8] G.R. Stewart, J.L. Smith and B.L. Brandt, Phys. Rev. B26 (1982) 3783.
[9] A. de Visser, J.J.M. Franse and A. Menovsky, J. Magn. Magn. Mat. 43 (1984) 43.
[10] F. Steglich, J. Aarts, C.D. Bredl, W. Lieke, D. Meschede, W. Franz and H. Schäfer, Phys. Rev. Lett. 43 (1979) 1892.
[11] H.R. Ott, H. Rudigier, Z. Fisk and J.L. Smith, Phys. Rev. Lett. 50 (1983) 1595.
[12] L.E. DeLong, J.G. Huber, K.N. Yang and M.B. Maple, Phys. Rev. Lett. 51 (1983) 312.
[13] A. Menovsky, J.C.P. Klaasse, R. Verhoef and J.J.M. Franse, Proc. 14èmes Journées des Actinides, J. Schoenes, ed. (Davos, 1984) p. 131.
[14] A. Menovsky and J.J.M. Franse, J. Cryst. Growth 65 (1983) 286.
[15] A.B. Kaiser and S. Doniach, Int. J. Magn. 1 (1970) 11.
[16] T.P. Orlando, E.J. McNiff Jr., S. Foner and M.R. Beasley, Phys. Rev. B19 (1979) 4545.
[17] U. Rauchschwalbe, W. Lieke, C.D. Bredl, F. Steglich, J Aarts, K.M. Martini and A.C. Mota, Phys. Rev. Lett. 49 (1982) 1448.
[18] W. Franz, A. Griessel, F. Steglich and D. Wohlleben, Z. Phys. B35 (1978) 7.
[19] J. Friedel, Nuovo Cimento Suppl. 7 (1958) 287.
[20] S. Doniach and S. Engelsberg, Phys. Rev. Lett. 17 (1966) 750.
[21] P.H. Frings and J.J.M. Franse, to be published.
[22] A.J. Arko, M.B. Brodsky and W.J. Nellis, Phys. Rev. B5 (1972) 4564.