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LETTER TO THE EDITOR

Spin fluctuations and superconductivity in UPt_3 †

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Abstract. Measurements of electrical resistivity, AC susceptibility and specific heat on Czochralski-grown single-crystalline UPt_3 reveal a transition into a superconducting state below 0.48 K. From the experimental value of the upper critical field, $dB_{c2}/dT = -4.4\ \mathrm{T\,K^{-1}}$, an effective mass of $180m_0$ is deduced. 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 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 (Schneider and Laubschat 1981) and below 1000 K (Frings *et al* 1983), 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 FU (Schneider and Laubschat 1981) up to $3.0 \pm 0.1\ \mu_B$ per mole FU (Frings *et al* 1983). 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 present only for field directions along the *a* and *b* axes (Frings *et al* 1983); magnetisation curves up to 35 T (Frings *et al* 1983, Franse 1983a, b) at 4.2 and 1.5 K reveal a strongly non-linear behaviour along the *a* and *b* directions (giving a magnetic moment of 0.9 μ_B per mole FU at 35 T), whereas at 20 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, the specific heat shows no sign of magnetic order near 16 K. In the specific heat (Frings *et al* 1983, Stewart *et al* 1984) 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(mol FU)$^{-1}$ K^{-2}). Similar low-temperature anomalies in specific heat and susceptibility are found in the spin-fluctuation compounds UAl_2 and $TiBe_2$ (Arko *et al* 1973, Acker *et al* 1981, Stewart *et al* 1982, 1983). Therefore it is more likely that the UPt_3 data can be interpreted in terms of spin-fluctuation phenomena. Such a description is supported by resistivity measurements along the different crystallographic directions (de Visser *et al* 1984a): a sharp rise at low

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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 was first observed by Stewart *et al* (1984). From measurements of specific heat (Stewart *et al* 1984) and AC susceptibility (this work) there is convincing evidence for a transition into a bulk superconducting state near 0.5 K. In view of the large electronic term in the specific heat, the superconductor UPt_3 can be placed in a new class of materials, which are termed 'heavy fermion' superconductors. Other members of this class are $CeCu_2Si_2$ (Steglich *et al* 1979), UBe_{13} (Ott *et al* 1983), U_6Fe (DeLong *et al* 1983) and U_6Co (Menovsky *et al* 1984).

In this paper we report 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 data on the critical field.

Single-crystalline UPt_3 was prepared in a tri-arc melting apparatus by the Czochralski method (Menovsky and Franse 1983). 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 ratios (i.e. $\rho(300\ \mathrm{K})/\rho_0$) were 33, 33 and 37 for the a, b and c directions before annealing; after annealing the ρ_0 values decreased to 6.2, 3.0 and 1.7 $\mu\Omega$ cm, leading to residual resistance ratios of 38, 79 and 78, respectively. The differences in ρ_0 values, possibly due to small deviations from stoichiometry, are indicative of inhomogeneities remaining after annealing.

Superconductivity in our case was first observed on three annealed samples (samples #1, #2 and #3) in measurements of the electrical resistivity (using a standard four-point DC method with $I < 2$ mA) along different crystallographic directions. It was evident from our previous experiments (de Visser *et al* 1984a) that the temperature dependence of the resistivity follows only a T^2 behaviour, characteristic of a spin-fluctuation system (Kaiser and Doniach 1970), on 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 and 0.53 K. A second indication of superconductivity was obtained by AC susceptibility measurements ($B_{AC} < 2 \times 10^{-5}$ T, $f = 10.9$ Hz) on two unannealed samples (#4 and #5): large diamagnetic signals were observed below 0.40 and 0.43 K, respectively. The annealed samples, #6 and #7, gave a superconducting transition temperature, T_c, of 0.48 K, consistent with the values 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 figure 1). For sample #6, $B'_{c2} = 4.4\ \mathrm{T\ K^{-1}}$ (field along the a axis), which is comparable with B'_{c2} values for other 'heavy fermion' superconductors. Evidence for bulk superconductivity was derived from specific heat experiments on an unannealed single-crystalline sample (#8). Here the onset of superconductivity is at 0.43 K and the maximum in the specific heat 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 imperfect quality of the unannealed samples. A value for $\Delta C/\gamma T$ at 0.40 K of 0.48 has been obtained for a flux-grown sample (Stewart *et al* 1984).

The evidence presented above for bulk superconductivity in our Czochralski-grown single-crystalline samples is consistent with the strong evidence presented by Stewart *et al*

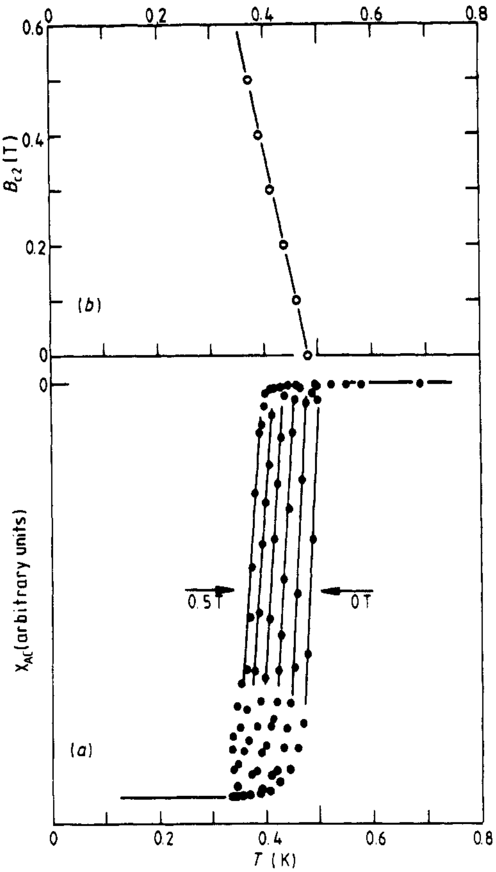

Figure 1. (*a*) AC susceptibility (in arbitrary units) in zero magnetic field and in fields of 0.1, 0.2, 0.3, 0.4 and 0.5 T (directed along the *a* axis) for sample #6. (*b*) Upper critical field as a function of temperature; T_c is defined at the 50% point (indicated by arrows in (*a*)).

(1984) on flux-grown samples. From their data on electrical resistivity, AC susceptibility and specific heat they obtain values of T_c^{onset} of 0.54, 0.53 and 0.54 K, respectively. 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), which 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 demagnetising factors. An experiment to provide hard proof, i.e. the observation of the Meissner effect, is in preparation.

In an attempt to give a qualitative analysis of the superconducting properties we follow the approach that has been applied successfully to A15 superconductors by Orlando *et al* (1979) and to $CeCu_2Si_2$ by Rauchschwalbe *et al* (1982). If we make two crude assumptions—(i) that there is a spherical Fermi surface, ignoring possible anisotropy effects, and (ii) that we discard the possibility of UPt_3 being a strong-coupling superconductor—then the Fermi wavenumber, k_F, can be derived from the temperature dependence of the upper critical field according to the relation

$$B'_{c2} \simeq 7.95 \times 10^{32}(\gamma^2 T_c / k_F^4) + 4780\gamma\rho_0. \quad (1)$$

We obtain a value of $k_F \simeq 1.1 \times 10^{10}$ m^{-1} if we insert our experimental results (sample #6) for B'_{c2} (=4.4 T K^{-1}), T_c (=0.48 K), the residual resistivity ρ_0 (although not measured on sample #6 we may estimate it at $\simeq 3 \times 10^{-8}$ Ω m (de Visser *et al* 1984a)) and the electronic term in the specific heat γ (=9.94 × 10^3 J m^{-3} K^{-2} with $V_{mole} = 4.243 \times 10^{-5}$ m^3). This value of k_F does not differ much from a value of $k_F \simeq 1.6 \times 10^{10}$ m^{-1} that can be deduced (de Visser *et al* 1984b) from the maximum in the resistivity ($\simeq$240 μΩ cm near room temperature). Using the value of k_F derived here we now obtain a Fermi velocity $v_F = k_B^2 k_F^2 / 3\hbar\gamma \simeq 6.8 \times 10^3$ m s^{-1} within the Fermi-liquid model and an effective mass $m^* = \hbar k_F / v_F \simeq 180 m_0$, values that are of the same order of magnitude as those derived for other 'heavy fermion' superconductors. The value for B'_{c2}, in the clean limit calculated with the above values for γ, T_c and k_F, is 3.3 T K^{-1}.

Accepting this type of analysis, we are able to estimate some important parameters which characterise the superconducting state of UPt_3. If we insert the above values for k_F ($\simeq 1.1 \times 10^{10}$ m^{-1}), T_c, ρ_0 and γ in the relations given by Orlando *et al* (1979), we derive the BCS coherence length $\xi_0 \simeq 2.0 \times 10^{-8}$ m, the mean free path $l \simeq 3.6 \times 10^{-8}$ m and the London penetration depth (as $T \to 0$) $\lambda_L \simeq 3.6 \times 10^{-7}$ m; $\xi_0 \simeq l$ means that our samples are in neither the dirty limit ($l \ll \xi_0$) nor the clean limit ($l \gg \xi_0$), as could be expected from the observed ρ_0 values (de Visser *et al* 1984a). The Ginzburg–Landau parameter, κ_{GL}, is found to be about 23, a value characteristic of type-II superconductivity.

At this point we want to stress once more that the calculations presented above could only be made under crude assumptions. That of ignoring anisotropy effects must be far from appropriate for UPt_3, in view of its anisotropic magnetic properties. In particular, our

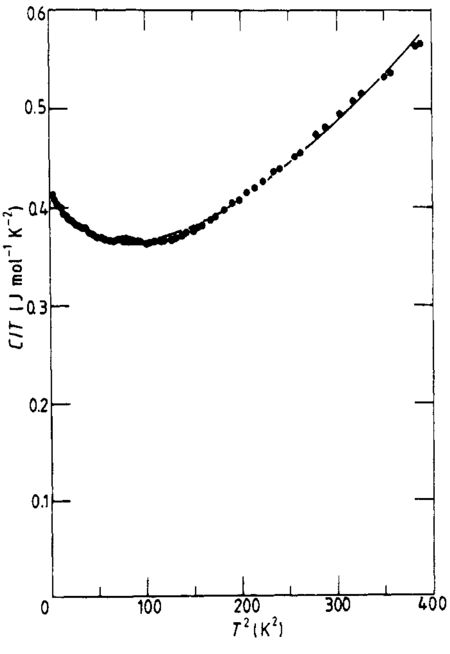

Figure 2. C/T against T^2 for UPt_3 (sample #8). The full curve represents a fit to all data points in the temperature interval 1.2–20 K; see table 1.

Table 1. 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 (K)	γ (mJ(mol FU)$^{-1}$ K^{-2})	β^* (mJ(mol FU)$^{-1}$ K^{-4})	δ (mJ(mol FU)$^{-1}$ K^{-4})
1.2–20	422	−3.72	1.38
1.2–15	423	−3.80	1.41
1.2–10	421	−3.53	1.30
6–10	424	−3.78	1.40
8–20	413	−3.45	1.30
1.2–17†	422	−4.18	1.54

† Frings and Franse (1984).

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^{-1} at T_c and $B'_{c2}=6.0$ T K^{-1} near 0.40 K, with the magnetic field applied along the c axis) asks for further detailed investigations.

The normal-state specific heat of a Czochralski-grown single-crystalline sample, shown in figure 2, reveals a pronounced spin-fluctuation phenomenon—a $T^3 \ln(T/T^*)$ contribution to the specific heat (Doniach and Engelsberg 1966). It was shown previously (Stewart *et al* 1982, 1983) that the specific heat data of the spin-fluctuation compounds $TiBe_2$ and UAl_2 could well be fit to the equation

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

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* (1983) on polycrystalline UPt_3. Recently, Stewart *et al* (1984) published results for the specific heat data on their flux-grown single-crystalline UPt_3. We performed a least-squares fit to 85 data points that we obtained in the temperature range 1.2–20 K (see figure 2), using equation (2), in different temperature intervals. The results of these fits are listed in table 1. The values for the coefficients in equation (2) do not change dramatically for the different temperature intervals, indicating that equation (2) 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 (Frings and Franse 1984).

In summary, we conclude from our experiments on resistivity, AC susceptibility and specific heat 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 data on the critical field points to a characterisation 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.

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