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SPIN FLUCTUATIONS AND SUPERCONDUCTIVITY IN UPt_3

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A survey is presented on the magnetic and superconducting properties of hexagonal single-crystalline UPt_3. Specific-heat experiments between 0.5 K and 20 K reveal a $T^3 \ln T/T^*$ contribution, which indicates spin-fluctuation phenomena. The magnetic susceptibility in this temperature region, measured in fields up to 35 T, is strongly anisotropic and exhibits distinct anomalies for field directions in the basal plane, suggesting some type of magnetic transition. No sign of magnetic order, however, is found in specific-heat and resistivity measurements. In experiments under hydrostatic pressures up to 5 kbar, a reduction of these features is observed. Superconductivity in UPt_3 is observed below 0.5 K. Although the transition temperature depends on the heat treatment of the single-crystalline samples, there is convincing evidence that superconductivity is a bulk property. Large values are derived for the effective electron mass ($\approx$ 180 m_0) of the pairing electrons from the temperature dependence of the upper critical field.

1. INTRODUCTION

The interest in the electronic properties of the intermetallic compound UPt_3 started with the XPS studies by Schneider and Laubschat (1) in 1981. In order to complete their electronic structure data these authors also measured the susceptibility of the whole series of uranium-platinum compounds and observed a maximum in the susceptibility of polycrystalline UPt_3 around 16 K. This temperature was interpreted as the Néel temperature of this apparently antiferromagnetic compound. This maximum in the susceptibility at 16 K was confirmed by Frings et al. (2). These latter authors studied, in addition, the high-field magnetization curves at 4.2 K and the specific heat of the whole series of uranium-platinum compounds. In the low-temperature magnetization curve of UPt_3 some type of magnetic transition was observed at 20 T. This transition, observed in a polycrystalline sample, seemed to support antiferromagnetism in this compound. Specific heat measurements on the same sample, however, did not reveal any sign of magnetic ordering, in particular not around 16 K. Instead, a large upturn was observed in the c/T versus T^2 curve going to low temperatures. This upturn was found to be nearly field independent between 0 and 5 T. In the meantime large single crystals of UPt_3 became available (3) and large anisotropies in the magnetic properties were found to be present. In subsequent papers by Franse (4,5) the possibility of spin-fluctuation phenomena was suggested for this compound and the analogy with the compound $TiBe_2$ was put forward. Resistivity measurements on the whole series of uranium-platinum compounds were carried out by De Visser et al. (6) in the temperature range between 1.5 K and 300 K and, in addition, for UPt_3 under pressures up to 5 kbar. The resistivity of single-crystalline UPt_3 is strongly anisotropic and reveals pronounced spin-fluctuation effects, that are partly suppressed at a pressure of 4 kbar.

The interest in this material strongly increased since the discovery of superconductivity by Stewart et al. (7) in flux-grown single-crystalline samples. Since that time a large effort was put in this material: critical field studies were undertaken by De Visser et al. (8,9) and by Willis et al. (10) in order to evaluate the superconducting parameters; the anisotropy in the critical field was studied by Chen et al. (11) and by Willis et al. (10); Meissner effect studies by Palstra et al. (12) show that superconductivity in this material must be considered as a bulk property; the spin-fluctuation phenomena in UPt_3 were further investigated by Frings and Franse (13) in high-field magnetization studies between 1.4 K and 77 K, by De Visser et al. (14) in high-field magnetoresistance experiments in the same temperature interval and by De Visser et al. in thermal expansion measurements (15).

At present a variety of experimental techniques and computational effort is employed to investigate this intriguing coexistence of spin fluctuations and superconductivity in UPt_3: polarized neutron scattering experiments above and below the superconducting transition tempe-

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rature establish the existence of magnetic fluctuations (16); to shed light on the electronic structure, reflectivity measurements are performed (17); de Haas-van Alphen experiments along the a-axis and c-axis of annealed Czochralski-grown single crystals in fields up to 40 T at 1.4 K did not reveal any oscillation in the magnetization curves so far; specific heat measurements below 100 mK by an adiabatic method turn out to be difficult to perform by self heating of the bulk samples; neutron diffraction and X-ray measurements put some question marks on the crystallographic structure that have not yet been resolved.

In this contribution we present a survey of the experimental data on spin-fluctuation effects and superconductivity that have been reported so far. We start our review by stressing the importance of sample preparation, handling and specification. The superconducting properties turn out to be sensitive to the sample preparation method in particular.

2. SAMPLE PREPARATION AND SPECIFICATION

The intermetallic compound UPt_3 crystallizes in the hexagonal $MgCd_3$-type of structure (also indicated as the Ni_3Sn-type of structure) with the spacegroup $P6_3/mmc$. The unit cell contains two formula units, the U atom being positioned at site 2c (1/3, 2/3, 1/4 etc) and the Pt atoms at site 6h (x, 2x, 1/4 etc, with x = 5/6 in the ideal case) according to the X-ray powder diffraction results by Heal and Williams (18). These structural data have been confirmed by X-ray measurements on a single-crystalline UPt_3 whisker at room temperature. Powder diffractograms performed with neutrons at 300 K, however, revealed a large contamination of the 220 reflection for which no explanation could be offered so far (19). Subsequent neutron diffraction studies on a single-crystalline sphere (diameter 3 mm) at 300 K did not show the expected extinctions for 001 or hhl reflection ($l = 2n+1$) as prescribed for the space group $P6_3/mmc$. Additional diffraction experiments are planned to solve these structural problems.

The compound UPt_3 melts congruently with a melting temperature of 1700°C approximately. A single-crystalline rod (diameter 6 mm, length 20 mm) could be prepared by the Czochralski method in a tri-arc melt installation with a continuously gettered argon atmosphere. Starting materials were: U (2N8) and Pt (4N). No efforts were made so far to optimize the growth conditions and to vary the stoichiometric ratio of the starting materials. A tendency towards (or away from) stoichiometry is inherent to the Czochralski method of single crystal growth and should be considered. There is, however, no evidence from the existing phase diagram for a homogeneity range for the UPt_3 phase. The lattice parameters of the as-grown UPt_3 single crystal are: $a = 5.752(12)$Å, $c = 4.897(3)$Å, in good agreement with literature data on polycrystalline material (18). The calculated density of $(19.49\pm0.04)\,g/cm^3$ is within the experimental error equal to the experimentally derived value of 19.45 g/cm^3. Residual resistance ratios (i.e. $\rho(300\ K)/\rho_0$) of samples cut out of this rod by spark erosion were typically of the order of 35. By an additional annealing at 900°C during one day this number increases up to 80. We must realize that at growing single crystals by the Czochralski method a large temperature gradient exists over the solidified material. Apart from stresses, defects in the atomic structure may be introduced in a less clear way. At the additional annealing the value for ρ_0 decreased to about 2 μΩcm. The superconducting transition temperatures of annealed samples is 0.5 K, whereas for unannealed samples values between 0.40 and 0.45 have been found.

Small-mass (1 mg) needle-like crystals, grown from a bismuth flux, with resistance ratios up to 145, have been used by Stewart et al. in their discovery of superconductivity in UPt_3. The temperature of 0.53 K for the onset of superconductivity reported by Stewart et al. is almost equal to the values observed in annealed Czochralski-grown bulk single crystals, in arc-melted polycrystalline samples and in powdered material after annealing. Menovsky et al. (20) reported the spontaneous growth out of the melt of regularly shaped UPt_3 whiskers. The lattice parameters of these whiskers agree within the experimental error with the above given values. The density of the whiskers, calculated from the lattice parameters equals 19.46 g/cm^3 and is almost identical to the experimentally derived value for a bulk sample. Experiments on these whiskers are in progress.

3. SPIN-FLUCTUATIONS

Investigations into the electronic and magnetic properties of single-crystalline UPt_3 reveal distinct anomalies in the temperature range between 1.5 K and 20 K. The magnetic susceptibility shows a maximum at 16 K for field directions in the basal plane only. High-field magnetization and magnetoresistance measurements at 4.2 K and 1.5 K point to some type of magnetic transition at 20 T for field directions in the basal plane; for fields parallel to the hexagonal axis these transitions do not occur. After the transition the magnetic moment per cell reaches a value of about 1 μ_B. Above 20 K the transitions are no longer visible. Specific heat measurements reveal the presence of a $T^3 \ln T/T^*$-term which is thought to be a characteristic sign for the presence of spin fluctuations. The $T^3 \ln T/T^*$-term in the specific heat manifests itself by a low-temperature upturn in a plot of c/T versus T^2. This upturn is found to be only slightly field dependent in the magnetic field range between 0 and 5 T for field directions in the basal plane and along the hexagonal axis. The value for the coefficient of the $T^3 \ln T/T^*$-term re-

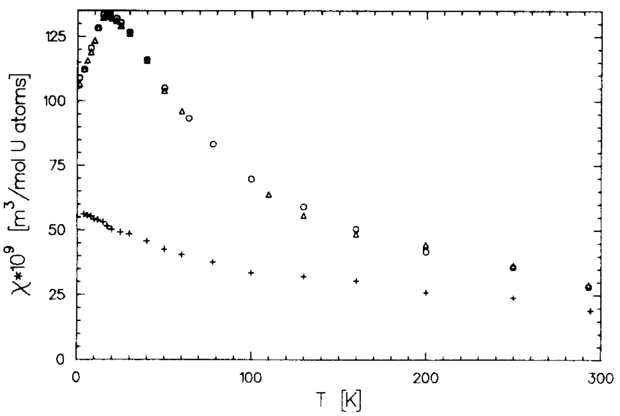

FIGURE 1
$\chi(T)$ of UPt_3, measured in fields up to 1.3 T along a-axis (o), b-axis (Δ) and c-axis (+) |data from ref.13 .

produces very well for different temperature intervals in the temperature range between 1.5 K and 20 K. In experiments on polycrystalline and single-crystalline samples nearly identical results for the specific heat have been obtained. The resistivity of single crystalline UPt_3 is strongly anisotropic and points to pronounced spin-fluctuation effects that are partly suppressed under pressure. Residual resistivity values of 1.7 μΩcm and 3.0 μΩcm have been reported along the hexagonal axis and a basal plane direction, respectively; room-temperature values amount to 132 μΩcm and 238 μΩcm respectively. A T^2-dependence of the resistivity, providing another justification for applying the concept of spin fluctuations to this intermetallic compound, is only observed in the lower limit of the explored temperature interval.

In thermal expansion measurements on polycrystalline and single-crystalline UPt_3 low-temperature anomalies are observed which are very similar to the $T^3 \ln T/T^*$-contribution to the specific heat but even more pronounced pointing to large values for the appropriate Grüneisen parameters. Large volume effects on the spin-fluctuation phenomena are also evident from high-pressure studies on the resistivity and the magnetization: the low temperature anomalies in these quantities are largely suppressed under pressures of 5 kbar; the maxima in the $\partial\rho/\partial T$- and $\partial\sigma/\partial H$-T curves shift towards higher temperatures under pressure.

In order to illustrate the above presented summary of low-temperature anomalies in the electronic and magnetic properties of UPt_3, we present in fig.1 the temperature dependence of the magnetic susceptibility for different crystallographic directions, in figs. 2 and 3 the results of high magnetic field studies on the magnetizations and the susceptibility, in fig.4 magnetoresistance data, in fig.5 the specific heat in zero field and in an applied field of 5 T, in fig.6 the resistivity versus temperature curve for different crystallographic directions and finally in Table 1 the results of a numerical analysis of the specific heat data for different samples and in different temperature intervals.

From fig.1, a characteristic temperature of 16 K and from figs. 3 and 4 a characteristic field of 20.5 T are deduced. Whether these maxima in the $\chi(T,H)$ and $\rho(H)$ curves should be considered as a proof for the existence of spin fluctuations is still a point of discussion. Other interpretations of the peaks in $\chi(H)$ and $\rho(H)$ in figs.3 and 4 in terms of an order to order transition or in terms of a special structure of the density of states curve near the Fermi level, however, are found to be less appropriate. Most convincing evidence for the spin-fluctuation phenomena comes from the presence of the $T^3 \ln T/T^*$-term in the specific heat. These specific heat data enable us through the relation $\beta^* = \beta - \delta \ln T^*$ to derive a value for the characteristic temperature T^*, provided that the coefficient β of the phonon term is known. This coefficient together with the unenhanced coefficient γ of the electronic term in the specific heat have been estimated by Frings (21) from analyzing the specific heat data at temperatures above 16 K:
γ = 225 mJK^{-2}/mole f.u. and

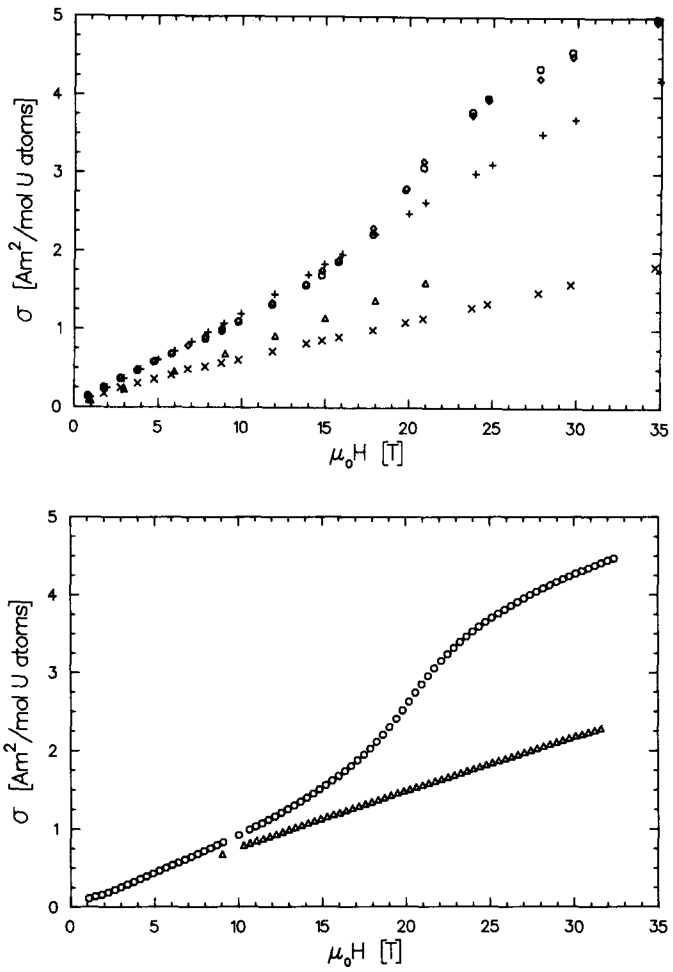

FIGURE 2

Magnetization versus field of UPt$_3$ at different temperatures and along different crystallographic directions |data from ref.13|.
4.2 K: a-axis (◇), b-axis (o), c-axis (x)
20 K: a-axis (+)
77 K: a-axis (Δ).

β = 0.85 mJK^{-4}/mole f.u., resulting in a value for the Debye temperature of 210 K and in a value for T* of 26 K. This value for T* is rather low compared to the temperature interval in which the specific heat can satisfactorily be described with the additional logarithmic term. By writing $\gamma^* = \gamma m^*/m$ we estimate a value for the mass enhancement due to spin fluctuations, m*/m, of 1.9.

The Stoner enhancement factor can be obtained by combining the specific heat and susceptibility data. Neglecting the orbital and diamagnetic contributions to the susceptibility Frings (21) derived a value for S of 2.7 from a susceptibility value for a polycrystalline sample of 103 x 10^{-9} m^3/mole f.u. and an unenhanced γ-value of 225 mJ/K^2 mole f.u. A ten percent larger value for S is derived at using the susceptibility value in the basal plane. This unenhanced γ-value is still enlarged by electron-phonon contributions. Taking these contributions into account a larger value for S must be expected. The uncertainties at deriving a value for β and subsequently for m*/m and T*, however, prevent us from giving a too elaborate analysis of these enhancement effects. Besides that, the anisotropy in the susceptibility puts another restriction on this way of

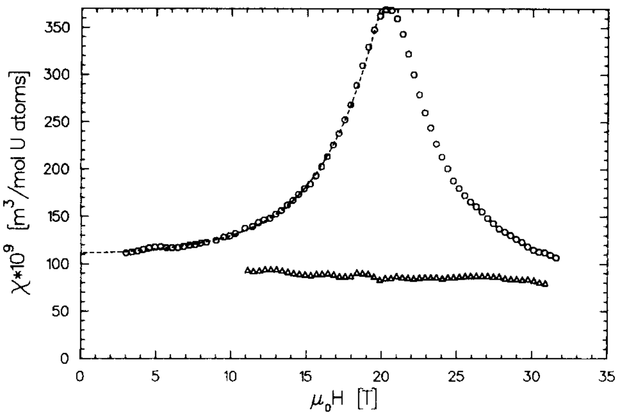

FIGURE 3
The differential susceptibility for UPt_3 at 4.2 K along the b-axis (o) and at 77 K along the a-axis (Δ). The broken curve represents a fit to eq.1 with values for the parameters given in the text [data from ref.13].

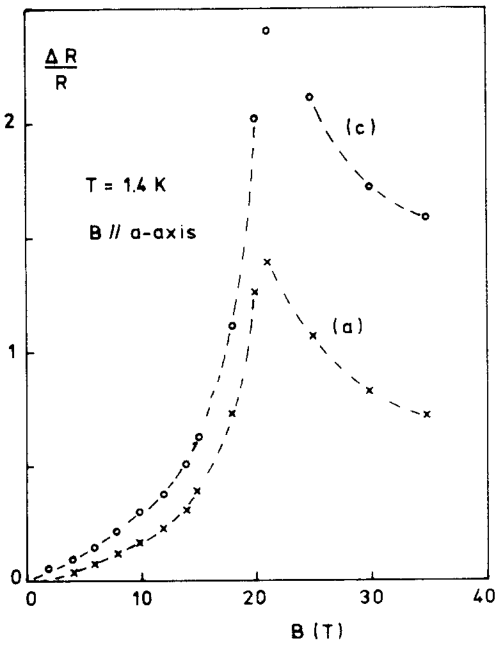

FIGURE 4
Magnetoresistance of UPt_3 at 1.4 K with the field along the a-axis and the current in the a-direction (x) and c-direction (o) [data from ref. 14].

TABLE I

Specific heat of UPt_3 below 20 K; c/T fitted to: $\gamma^* + \beta^* T^2 + \delta T^3 \ln T$; $\gamma^* = \gamma m^*/m$, $\beta^* = \beta - \delta \ln T^*$, c in mJK^{-1}/mole f.u., T in K [data from ref.8 and 13].

sample	temp. range	γ^*	β^*	δ
polycr.	1.3-16.5	422	-4.18	1.54
single crystal	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	422	-3.45	1.30

analyzing the experimental data.

In order to facilitate the comparison between theory and experiment the high-field magnetization data for field directions in the basal plane have been fitted to the expression:

$$\partial\sigma/\partial H = \chi_0 \{1 + aH^2 + bH^4 + cH^6\} \qquad (1)$$

For fields up to 20 T the values for $10^9\chi_0$, 10^5a, 10^7b and 10^8c amount to 112(1)m^3/mole f.u., 135(25)m^3/T^2 mole f.u., 0(16)m^3/T^4 mole f.u. and 3.0(3)m^3/T^6 mole f.u., respectively.

Finally, in fig.7, the coefficient of thermal expansion is shown along different crystallographic directions.

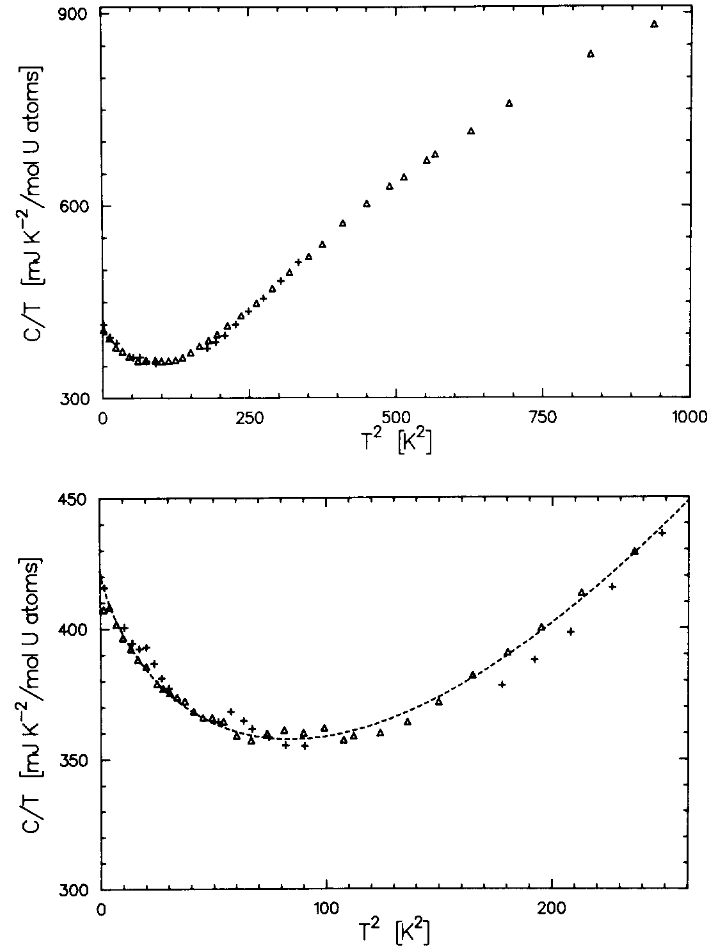

FIGURE 5
Specific heat of polycrystalline UPt$_3$ in a plot of c/T versus T^2 with (+) and without (Δ) a magnetic field of 5 T. The broken curve represents the three parameter fit of the zero field data, see Table I |data from ref.13|.

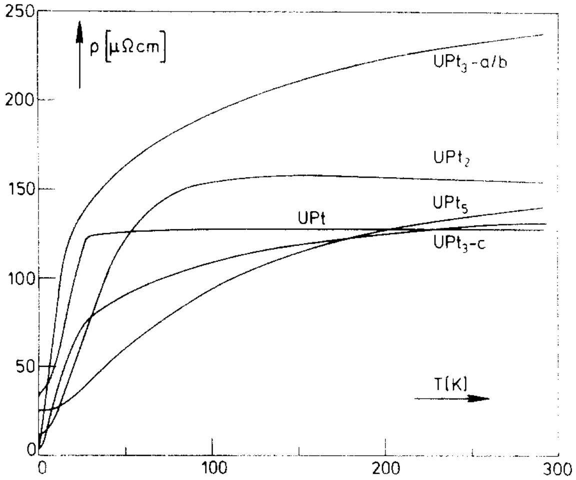

FIGURE 6
Temperature dependence of the electrical resistivity of UPt_3 along different crystallographic directions. For comparison the results for UPt, UPt_2 and UPt_5 are shown [data from ref.6].

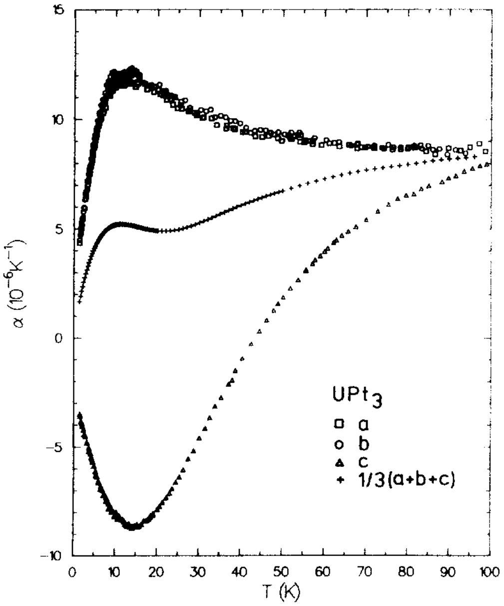

FIGURE 7
The coefficient of thermal expansion of UPt_3 along different crystallographic directions [data from ref.14].

4. SUPERCONDUCTIVITY

Superconductivity in the intermetallic compound UPt_3 was far from expected for several reasons. For more than one decade the Hill plot (22) served as a guide for observing magnetic order and superconductivity in the actinide compounds. According to the systematics of this plot, the distance between neighbouring actinide ions has a critical value beyond which no superconductivity and below which no magnetic order is believed to exist. For UPt_3 this distance amounts to 4.12 Å, considerably larger than the critical value of 3.5 Å. Furthermore, spin fluctuations which occur in nearly magnetic substances are thought to suppres normal S-state superconductivity and, as we discussed in the preceding section, spin-fluctuation phenomena are very pronounced in this compound. At the discovery of superconductivity by Stewart et a. directly the possibility of P-state superconductivity was mentioned and a lot of effort is put in proving this special type of superconductivity to be present in this intermetallic compound.

Another remarkable feature is that superconductivity is easily destroyed by damaging the sample as was also reported by Stewart et al These damaging effects arose the question whether superconductivity in this compound should be considered as a bulk property. Evidence for bulk superconductivity was derived from specific heat experiments, although the values for the relative jump in the specific heat at T_c are 1/3 to 1/4 of the BCS result.
Palstra et al. recently investigated the superconducting properties of UPt_3 by determining the Meissner effect. A serious problem in the investigation of the Meissner effect by cooling the sample in field and measuring the flux expulsion are flux spinning effects. In fig.8 we show some of the flux expulsion results which lead to a superconducting volume fraction of 30% at 332 mK. This expelled flux will set a lower limit to the superconducting volume fraction. In order to avoid these difficulties with flux pinning, the virgin magnetization curve in low fields was measured on the same sample, in this case cooled below T_c in zero field. In this experiment, however, it is impossible to discern normal regions embedded in superconducting regions. This method will set for that reason an upper limit to the superconducting volume fraction. The virgin magnetization curve at 352 mK is also shown in fig.8 and yields a superconducting volume fraction of 80%, see also fig.10. Due to the large flux pinning effects this latter result is believed to give the most reliable estimate of the superconducting volume fraction. The superconducting volume fraction derived by these two methods are given in fig.9 as a function of temperature. In fig.10 the virgin magnetization curve and a magnetization loop at 352 mK are shown. A

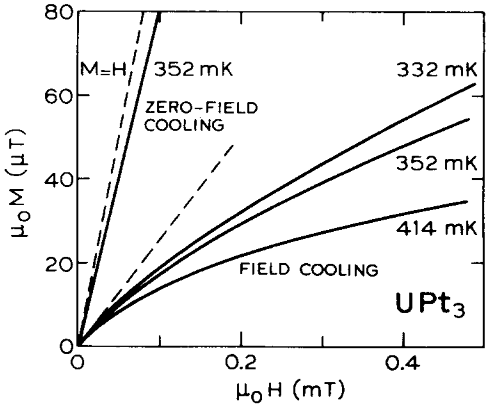

FIGURE 8
Magnetization versus magnetic field of UPt$_3$. Zero-field cooling denotes the virgin magnetization curve.The curves denoted by "field cooling" were drawn through the data points (not shown) obtained by measuring the flux expulsion at constant magnetic field as a function of temperature. The dashed lines represent the full Meissner effect (M=H) and the initial slope of the 332 mK curve.

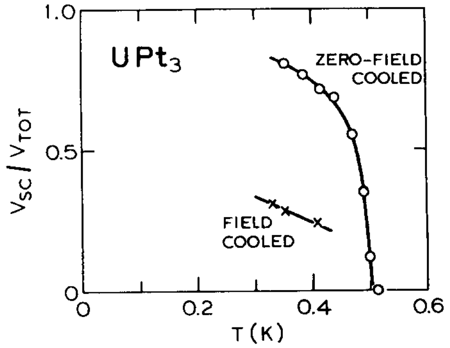

FIGURE 9
Superconducting volume fraction of UPt$_3$ versus temperature as obtained by the field cooling (x) and the zero-field cooling (o) methods |data from ref.12|.

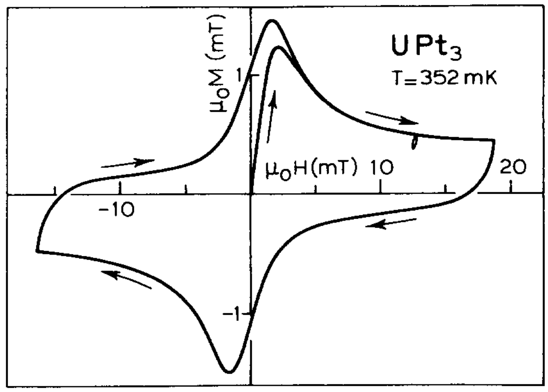

FIGURE 10
Virgin magnetization curve and magnetization loop of UPt$_3$ at 352 mK |data from ref. 12|.

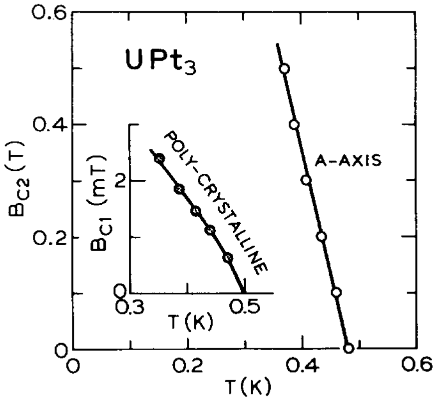

FIGURE 11
Temperature dependence of the critical fields B_{c1} (insert) for a polycrystalline sample, and B_{c2} for a single crystal along the a-axis |data from ref.8 and 12|.

value for the lower critical field, B_{c1}, of 2.2 mT is derived from this experiment. The upper critical field, B_{c2}, has been studied by De Visser et al. in ac susceptibility measurements on polycrystalline and single-crystalline samples. Values for B_{c1} and B_{c2} as a function of temperature are shown in fig. 11. From $B_{c2}/B_{c1} \cong 2\kappa^2/\ln\kappa$ one calculates for the Ginzburg-Landau parameter κ a value of 20. The results of these upper critical field studies have been used by De Visser et al. to estimate some important parameters that characterize the superconducting state of UPt_3: the coherence length $\xi_0 \cong 2.0 \times 10^{-8}$m, the mean free path $l \simeq 3.6 \times 10^{-8}$m and the London penetration depth $\lambda \simeq 3.6 \times 10^{-7}$m. The Ginsburg-Landau parameter κ is found to be 23, in good agreement with the value of 20 obtained from the ratio of the upper and lower critical fields. The same analysis results in an effective mass of the pairing electrons of 180 m_0, with m_0 the free electron mass, thereby classifying this material as a heavy fermion superconductor. Nearly identical results for these superconducting parameters have been reported by Chen et al. with the current along the hexagonal c-axis and by Willis et al. with the current along the c-axis as well as along a direction in the basal plane. For perpendicular fields non-linearities are observed in the temperature dependence of the upper critical field. The resulting anisotropies in the upper critical field cannot arise, according to Varma (23), for S-state superconductors and follow the behaviour expected for triplet pairing. Non-linearities in the upper critical field as a function of temperature have also been reported by De Visser et al. in ac susceptibility measurements with the magnetic field applied along the c-axis.

Whereas hardly any effect of the sample condition is noticed in specific heat measurement in the normal state, large annealing effects are observed for Czochralski-grown single-crystalline UPt_3 on the superconducting transition. Taking into account the different definitions of the superconducting transition temperature used in references 7 and 8 nearly identical results are obtained for flux-grown needle-like and Czochralski-grown bulk single crystals after the appropriate annealing. Powdered and polycrystalline samples closely agree with these results. In unannealed Czochralski-grown single crystals, however, the superconducting transition temperature is 0.05 to 0.1 K lower. Stresses and/or structural defects apparently influence the superconducting transition. More systematic studies in the relation between sample condition and superconductivity are needed.

5. CONCLUDING REMARKS

The intermetallic compound UPt_3 shows an unusual combination of pronounced spin-fluctuation phenomena and superconductivity. Spin-fluctuation effects are strongly anisotropic. Special low-temperature features in the high-field magnetization and resistivity curves only occur for external fields applied along any direction in the basal plane. For arbitrary field directions the projection of the field in the basal plane governs the resistance curve. Large anisotropies are also observed in the $\chi(T)$ and $\rho(T)$ curves.

Superconductivity in UPt_3 is a bulk property. A temperature of (0.53 ± 0.02)K for the onset of superconductivity is reproduced in annealed single-crystalline and poly-crystalline samples. More systematic studies on the relation between sample quality and superconducting properties are required, especially when tiny effects in the superconducting properties play a role in speculations about a new type of superconductivity in intermetallic compounds.

Resistivity and specific heat measurements on polycrystalline UPt_2 and UPt_5 indicate spin-fluctuation effects in these compounds to be present too, although considerably less pronounced than in UPt_3. Superconductivity in UPt_2 and UPt_5 has not been observed so far in experiments down to 0.3 K, suggesting that the absence of superconductivity and the presence of less pronounced spin-fluctuation phenomena are connected. This connection, however, is not supported by annealing effects that hardly influence the spin fluctuations but largely change the superconducting transition temperature.

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) A.Menovsky and J.J.M.Franse, J.Cryst.Growth 65 (1983) 286.

(4) J.J.M.Franse, J.Magn.Magn.Mat. 31-34 (1983) 819.

(5) J.J.M.Franse, Helv.Phys.Acta 56 (1983) 131.

(6) A. de Visser, J.J.M.Franse and A.Menovsky, J.Magn.Magn.Mat. 43 (1984) 43.

(7) G.R.Stewart, Z.Fisk, J.O.Willis and J.L. Smith, Phys.Rev.Lett. 52 (1984) 679.

(8) A. de Visser, J.J.M.Franse, A.Menovsky and T.T.M.Palstra, Proceedings of the 4th General Conference of the Condensed Matter Division of the European Physical Society, The Hague (1984), to be published in Physica B.

(9) A. de Visser, J.J.M.Franse, A.Menovsky and T.T.M.Palstra, J.Phys.F, in print.

(10) J.O.Willis, Z.Fisk, J.L.Smith, J.W.Chen, S.E.Lambert and M.B.Maple, this conference.

(11) J.W.Chen, S.E.Lambert, M.B.Maple, Z.Fisk, J.L.Smith, G.R.Stewart and J.O.Willis, Phys.Rev.B in print.

(12) T.T.M.Palstra, P.H.Kes, J.A.Mydosh, A. de Visser, J.J.M.Franse and A.Menovsky, Phys.Rev.B., in print.

(13) P.H.Frings and J.J.M.Franse, to be published.

(14)A. de Visser et al., to be published.
(15)A. de Visser and J.J.M.Franse, Conference on Rare Earth and Actinides, St.Pölten, September 3-6 (1984).
(16)H.Capellman, S.M.Johnson, K.R.A.Ziebeck, J.J.M.Franse, A.Benoit and J.Flouquet, Fourth International Conference on Valence Fluctuations, Cologne, August 27-30 (1984).
(17)J.Schoenes and J.J.M.Franse, Conference on Rare Earth and Actinides, St.Pölten, September 3-6 (1984).
(18)T.J.Heal and G.I.Williams, Acta Cryst. 8 (1955) 494.
(19)R.B.Helmholdt, private communication.
(20)A.Menovsky, J.J.M.Franse and A.C.Moleman, J.Cryst.Growth 67 (1984) 31.
(21)P.H.Frings, Thesis Amsterdam (1984).
(22)H.Hill, Plutonium 1970, W.N.Miner ed. (AIME, New York) (1970) 2.
(23)C.M. Varma, to be published.