

INVITED PAPER

MAGNETIC AND SUPERCONDUCTING PROPERTIES OF UPt_3

J.J.M. FRANSE, A. de VISSER, A. MENOVSKY and P.H. FRINGS *

Natuurkundig Laboratorium der Universiteit van Amsterdam, Valckenierstraat 65, 1018 XE Amsterdam, The Netherlands

Recent results of experimental studies on the thermodynamic and transport properties of the heavy-fermion superconductor UPt_3 are reviewed. These studies include high-magnetic-field and high-pressure experiments in the normal and superconducting states as well as alloying effects on the normal- and superconducting-state properties. The low-temperature properties of UPt_3 are unusual and no satisfactory description in terms of crystal-field effects, spin-fluctuation phenomena, antiferromagnetic order, singlet or triplet superconductivity can be offered.

1. Introduction

The existence of spin fluctuations and superconductivity in the compound UPt_3 has strongly intensified the interest in the intermetallic uranium compounds. The study of both phenomena in this class of compounds started in the late sixties and the early seventies. Magnetism and superconductivity not always had a good reputation in these materials. The superconductivity of pure uranium [1] and the magnetic order of UPt [2,3] may serve as examples. Preparation techniques, the state of stress, the amount of oxides, all have their impact on the measured quantities. In those early studies lattice parameter considerations were thought to provide a hint where to observe superconductivity and in which compounds magnetic order was likely to be found. We now realise that the systematics of the early Hill plot is accidental [4] and that magnetic order and superconductivity in the uranium intermetallics is not governed by a single parameter as the distance, d_{U-U} , between neighbouring uranium ions. Hybridisation between the uranium 5f states and the s, p or d states of the second element prevents localisation of the 5f electrons and may induce superconductivity in compounds at the wrong side of the Hill limit (3.5 Å, approximately). A prominent example nowadays is UPt_3 with $d_{U-U} = 4.13$ Å. The unexpected discovery of its superconducting properties has been set down in a report of the Los Alamos group [5]. Since UPt_3 was already known to be a candidate for a spin-fluctuation compound [6,7], large interest arose in this material, not at

least because good-quality single crystals could be prepared as whiskers, under suitable conditions spontaneously growing from the melt [8], as small needles from a bismuth flux [9], or as bulk material from the melt by the Czochralski method [10].

In table 1 we present a survey on superconductivity, spin fluctuations, magnetic order and crystal-field effects of intermetallic compounds of uranium with Fe, Co, Ni, Pd and Pt. As we mentioned in a previous publication [4], magnetic order and superconductivity do not frequently occur in the intermetallics of uranium with d-transition elements. Whereas, in general, superconductivity is found in the uranium-rich and magnetic order in the uranium-poor compounds, the reverse is true for the uranium–platinum system. The low T_c -values in the superconducting compounds in table 1 are accompanied with relatively large values for the derivatives of the upper critical field to the temperature near T_c , of the order of a few tesla per kelvin, see refs. [12,14,19]. For U_6Fe , Pals et al. [11] provided strong experimental evidence from tunneling experiments that superconductivity in this compound is of singlet type. The large γ -value and the heavy mass of the pairing electrons of about 200 times the free-electron mass in UPt_3 , however, are so unusual that the possibility of p-wave pairing of the superconducting electrons cannot be excluded [20].

In one of our previous publications [19] we presented a survey of experimental data available at that time on spin fluctuations and superconductivity in UPt_3 . In the present work we emphasize the problems that are encountered at classifying UPt_3 in terms of crystal-field effects, antiferromagnetism, spin fluctuations and singlet or triplet superconductivity.

* Present address: Institute Laue-Langevin, Grenoble, France.

Table 1

Superconductivity (I), spin fluctuations (II), magnetic order (III) and crystal-field effects (IV) in intermetallic compounds of uranium with group-VIII d-transition elements; values are given for the coefficient, γ , of the term in the specific heat linear in temperature, for the (high-field) susceptibility, χ , at 4.2 K, for the superconducting transition temperature, T_s , for the spin-fluctuation temperature, T_{sf} , as determined from the logarithmic term in the specific heat at low temperatures, for the magnetic ordering temperature, T_c or T_N , and for the energy splitting between the lowest crystal-field levels, expressed in T_{cf} ; γ in mJ/K²molU, χ in m³/molU and temperatures in K

Comp.	Type	γ	$\chi \times 10^9$	$\chi/\chi(\gamma)$	T_s	T_{sf}	T_c, T_N	T_{cf}	Ref.
U ₆ Fe	I	25	38	1.5	3.86				12
UFe ₂	III	45	29	3.7			160		13
U ₆ Co	I	21	34	1.6	2.3				14
UCO ₂	II	35	17	2.8		18			13
U ₂ Co ₁₁	III						360		15
U ₆ Ni	I				0.5				16
UNi ₂	III	65	14	1.2			27		13
UPd ₃	IV	10	160,300 ^a					24,164 ^b	17
UPd ₄	III						10		18
UPt	III	110	47	2.5			19,27 ^c		13
UPt ₂		77	51	3.8					6
UPt ₃	I	422	57, 107 ^a	1.5	0.5	26 ^d			19
UPt ₅		85	34	2.3					6

^a || and \perp *c*-axis, resp.

^b quasi-cub. and hex. sites, resp.

^c differing ord. temp. exist

^d see also table 2

2. Normal-state properties

Experiments on the normal-state properties of UPt_3 include thermal and elastic properties (specific heat [7,9,19,21], thermal expansion [22], elastic constants [22,23]), magnetic properties (susceptibility [19], high-field magnetisation [24] magnetoresistance [25]) and transport properties (electrical [26] and thermal [27] conductivity, thermopower [27]) besides XPS [28], photoemission [29–31], reflectivity [32], synchrotron radiation [33] and neutron investigations [23,34]. DHvA experiments at 1.5 K in fields up to 40 T along different crystal directions prove to be not successful [19] just as NMR measurements are not. Susceptibility measurements below room temperature have been performed along different crystallographic directions and for a polycrystalline sample between 300 and 1000 K [7], see fig. 1. The anisotropy in the susceptibility between directions parallel and perpendicular to the hexagonal axis as well as the maximum in the susceptibility that occurs for field directions in the hexagonal plane have some resemblance with the crystal-field compound $PrNi_5$ [35]. Extending this resemblance and considering within the L - S coupling scheme the ground state to be $^3H_4(U^{4+})$ as is also suggested for UPd_3 [17], one deduces from the susceptibility data a value for the

crystal-field parameter B_0^0 of 7.0 K, quite close to the value of 5.82 K for $PrNi_5$ [35]. The effective moment that follows from the susceptibility data below 300 K amounts to $2.6\mu_B$ and does not exceed a value of $2.9\mu_B$ at temperatures up to 1000 K, clearly deviating from that of the most probable $5f^2$ ($3.58\mu_B$) or $5f^3$ ($3.62\mu_B$) configurations and as such an indication of a large overall crystal-field splitting. In case of UPd_3 a value of $2.8\mu_B$ has been reported for the temperature interval 70–300 K [36]. Magnetisation curves at 1.5 and 4.2 K for UPt_3 reveal non-linearities above 10 T for field directions in the basal plane, resulting in a peak in the differential susceptibility at 20 T [19,24], see fig. 2a. At nearly the same value for the component of the field in the basal plane the magnetoresistivity has a sharp maximum [19,25], see fig. 2b. The positive magnetoresistance and its peak at 21 T remind to some type of antiferromagnetism. The initial field dependence of the magnetoresistivity, however, is certainly not quadratic. Moreover, the magnetisation curves are not compatible with a simple type of antiferromagnetic order [7]. Again, crystal-field effects do not seem to be inconsistent with these high-field data. Finally, the thermal expansion is strongly anisotropic in UPt_3 with extrema in the linear thermal expansion coefficient along the different crystallographic axes between 10 and 15 K [19,22]. The

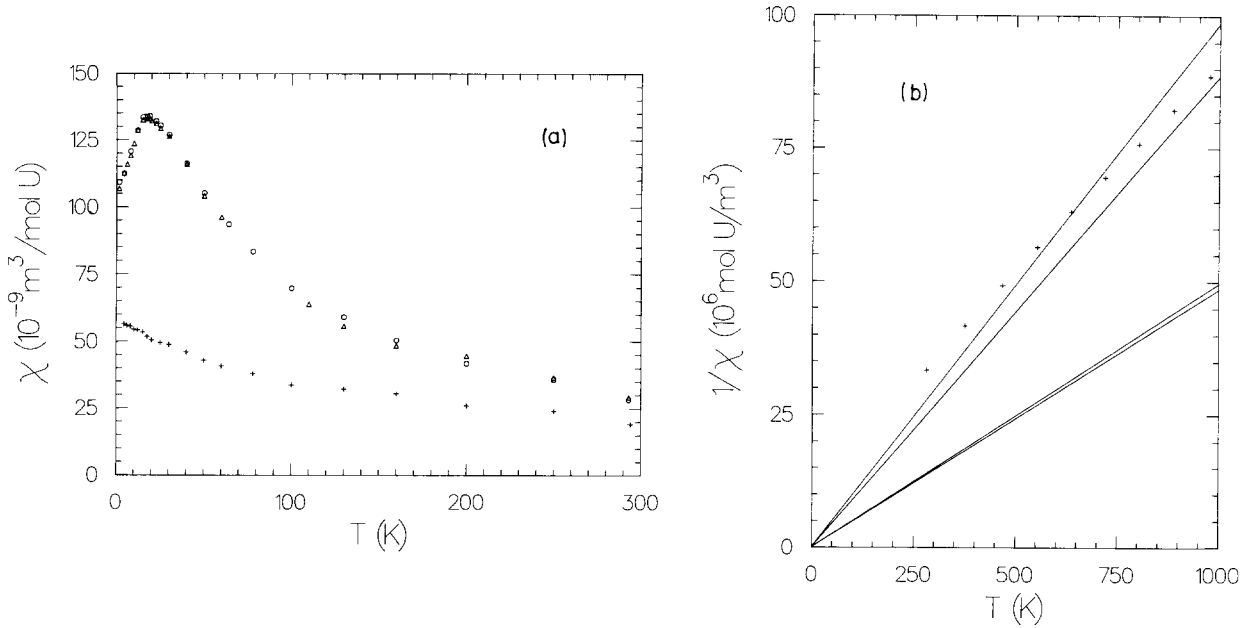


Fig. 1. The susceptibility, χ , of UPt_3 between 1.5 and 300 K for a single-crystalline sample along the a -(\circ), b -(Δ) and c -($+$) directions (a) and the inverse susceptibility, χ^{-1} , between 300 and 1000 K for a polycrystalline sample (b); the value for the effective magnetic moment derived from these curves ranges from $2.6\mu_B$ (below 300 K) to $2.9\mu_B$ (between 700 and 1000 K); full lines in (b) represent the effective moments $2.54\mu_B$ ($5f^1$), $2.68\mu_B$ ($5f^4$), $3.58\mu_B$ ($5f^2$) and $3.62\mu_B$ ($5f^3$); data from refs. [7,24].

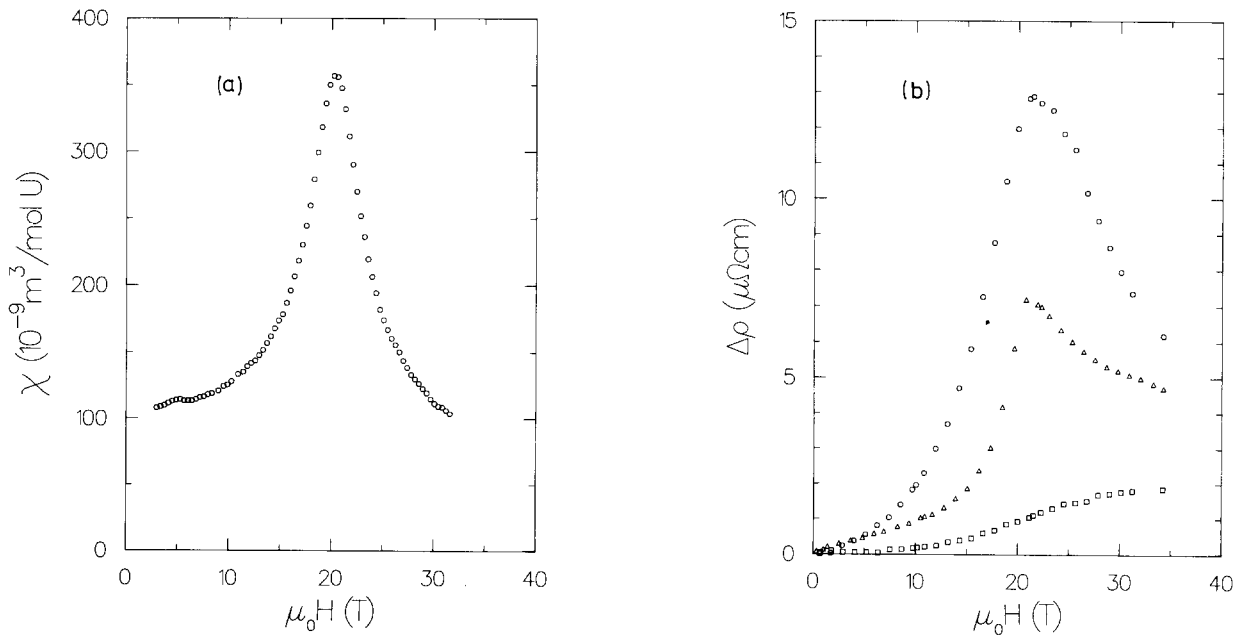


Fig. 2. The high-field maximum in the differential susceptibility along the b -axis at 4.2 K (a) and the magnetoresistivity with the current along the c -axis and the field parallel (4.2 K, \square) and perpendicular (4.2 K, \circ ; 1.5 K, Δ) to the hexagonal axis (b) of a single-crystalline UPt_3 sample; the maxima in the susceptibility only occur at low temperatures for field directions in the hexagonal plane; maxima in the magnetoresistivity are observed at low temperatures for all current directions at a value of 21 T for the component of the field in the hexagonal plane; data from refs. [24,25].

temperature at which these extrema are observed as well as the values for the thermal expansion coefficients along and normal to the hexagonal c -axis are again very similar in UPt_3 [22] and $PrNi_5$ [37].

Irrespective of this fairly large amount of evidence for crystal-field effects playing a dominant role in UPt_3 we have to discard this possibility on the basis of the low-temperature thermal properties. In particular the specific heat does not provide any argument to consider a level scheme with a splitting between the lowest levels of the order of 25 K as in case of $PrNi_5$, since no Schottky peak could be noticed for UPt_3 below 30 K. Instead, a logarithmic term at low temperature is observed together with an enhanced value of $422 \text{ mJ/K}^2 \text{ mol}$ for the coefficient of the linear electronic term. A remarkable feature is the weak field dependence of the specific heat below 20 K down to the lowest temperatures. The low-temperature thermal expansion is also inconsistent with a crystal-field type of description, because of the large upturn in the α/T vs. T^2 plot at low temperatures, see fig. 3. The Grüneisen parameter that is proportional to the ratio of the thermal expansion to the specific heat, exceeds its usual value of about 2 by almost two orders of magnitude [22] and reflects the strong coupling between the lattice and the spin-fluctuation phenomena. High-pressure experiments on resistivity [26,38] and susceptibility [39] in the normal state point to a strong depression of spin fluctuations with pressure and yield a value for the relative pressure dependence of the characteristic temperature for spin fluctuations of 25 Mbar^{-1} . Subsequent forced magnetostriction measurements [40] support this result and show that it is only the basal plane susceptibility which exhibits these large pressure effects. For a field direction along the hexagonal axis the pressure effect on the susceptibility is at least one order of magnitude smaller. The value for the characteristic temperature, T_{sf} , depends on the way it is extracted from the different types of experiments and ranges between 6.5 K (maximum in the temperature derivative of the resistivity) and 26 K (analysis logarithmic term in the specific heat), see table 2. The differences between the low-temperature properties of UPt_3 and UPd_3 are remarkable. The coefficient of the linear term in the specific heat of UPd_3 is no larger than 5 [41] to 10 [36] $\text{mJ/K}^2 \text{ mol}$. In UPd_3 two phase transitions have been observed at 5 and 7 K [42], both non-magnetic in origin. Evidence for crystal-field states comes from neutron-scattering experiments [17], from which a $5f^2$ configuration with an $L-S$ ground state, 3H_4 , is concluded. For both uranium sites in dhcp UPd_3 , the hexagonal and the quasi-cubic sites, a singlet ground state with an energy distance to

the first excited (doublet) level of 164 and 24 K, respectively, has been derived [17]. Which energy splitting belongs to which site, however, has not firmly been established. The temperature dependence of the susceptibility below 300 K could be reproduced fairly well with the proposed level scheme. Although there are small differences in the lattice parameters of these two hexagonal compounds (table 3), one cannot understand on the basis of the pressure dependence of the spin-fluctuation phenomena such drastic changes in the low-temper-

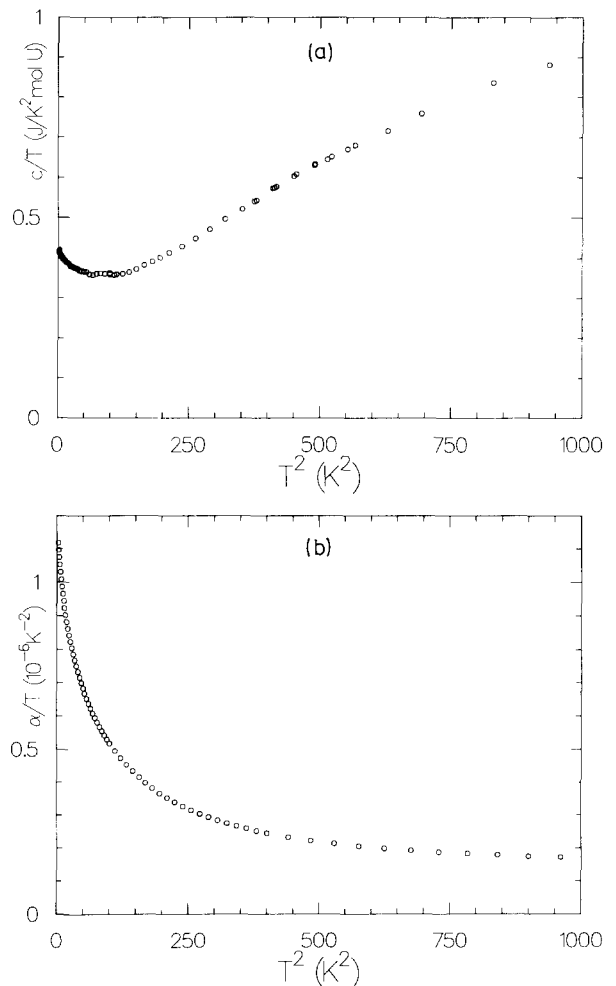


Fig. 3. The specific heat (a) and the thermal expansion (b) of UPt_3 plotted as c/T and α/T vs. T^2 , respectively; the limiting values in the normal state for c/T and α/T at T approaching zero, result in a value for the corresponding Grüneisen parameter of 75; a more sophisticated analysis leads to a value of 159 for the Grüneisen parameter that is associated with the spin-fluctuation phenomena; data from refs. [21,22].

Table 2

Values for the characteristic temperature for spin fluctuations, T_{sf} , in UPt₃ as derived from specific heat (c) and thermal expansion (α) measurements and values for the temperature at which a maximum in thermal expansion, susceptibility (χ), thermo-electric power (S) and resistivity (ρ) and their derivatives to the temperature are observed; data from refs. [19,22,24,26,27]

$T^3 \ln T/T_{sf}$ -term		Temperature (K) at which maximum is observed				
c	α	$\alpha(T)$	$\chi(T)$	$S(T)$	$\partial\rho/\partial T$	$\partial\chi/\partial T$
26	11	11	16	7	6.5	10

ature properties. An evident difference between UPt₃ and UPd₃ is the concentration of hexagonal sites which in UPd₃ is half that of UPt₃. In order to better understand the effect of a substitution of Pt by isoelectronic Pd, a series of pseudo-binary compounds, U(Pt_{1-x}Pd_x)₃, has been prepared with x -values of 0.01, 0.02, 0.05, 0.10, 0.15, 0.20 and 0.30 [43]. Specific heat measurements reveal that the spin-fluctuation phenomena are rapidly lost for x -values exceeding 0.1 [43,44], see fig. 4, the γ -values reaching a value almost equal to that of UPd₃ for $x = 0.3$. A maximal value of about 590 mJ/K²mol is observed at $x = 0.05$ for the coefficient γ of the specific heat. The increase in γ is accompanied by a decrease in T_{sf} as determined from specific heat measurements.

3. Superconducting-state properties

Superconductivity in UPt₃ occurs below 0.5 K [9]. Meissner-effect measurements [45,46] prove the superconductivity to be a bulk property. The transition temperature shows a quite large sample dependence. The highest values for T_s and the smallest transition regions are obtained for well-annealed single-crystalline samples [47]. Apparently, stresses destroy superconductivity as is evident from experiments on unannealed powdered samples [9]. In annealed polycrystalline samples the anisotropic thermal expansion coefficient below 100 K

can induce stresses between neighbouring crystallites up to several Kbars, causing a lowering of the transition temperature and a broadening of the transition region. The application of hydrostatic pressure on single-crystalline samples depresses the transition temperature and results in a value for $\partial \ln T_s / \partial p$ of -25 Mbar^{-1} [48], almost the opposite value as was found for $\partial \ln T_{sf} / \partial p$, see above. These high-pressure results for the two characteristic temperatures for superconductivity and spin fluctuations are not in favour of electron pairing in the superconducting state intermediated by spin fluctuations and seem to reject the suggestions for triplet superconductivity [49] in this compound. Nevertheless, triplet superconductivity in UPt₃ is not inconsistent with upper critical field studies as is discussed in another contribution to this conference [50], with the absence of a Josephson current in vacuum tunneling experiments with Al or Nd as counterelectrode [51] and with the T^2 -dependence of specific heat [27,52], thermal conductivity [27] and ultrasonic attenuation [53] in the superconducting state. An analysis of the temperature dependence of the upper critical field near the superconducting transition temperature provides us with values for the Fermi velocity ($v_F = 6.8 \times 10^3 \text{ m/s}$) and the effective electron mass ($m^* = 180m_e$) [21]. The Fermi velocity of the heavy-fermion superconductors is extremely low and for UPt₃ comparable in magnitude with the sound velocity ($v_s = 3.9 \times 10^3 \text{ m/s}$ [22,53]). The value for the effective mass of the electrons at the Fermi level of about 180 times that of a free electron does not imply that the mass enhancement due to spin fluctuations is of that order of magnitude. In fact, at writing $m^* = m_0(1 + \lambda_{sf})$, where λ_{sf} is the enhancement factor of the linear electronic term in the specific heat, a previous analysis [19] of the specific heat data in the normal state leads to a λ_{sf} -value of 0.88, resulting in a mass enhancement by spin fluctuations of 1.88. Band effects, electron-phonon enhancement etc. have to account for the remaining difference between m_0 and the free-electron mass. Band-structure calculations result in density of states values at the Fermi level that

Table 3

Lattice parameters, distance between neighbouring uranium ions, d_{U-U} , and molar volume, V_{mol} , of UPt₃ (hexagonal MgCd₃-type of structure) and UPd₃ (double-hexagonal TiNi₃-type of structure) at 300 K; data from refs. [10,36], respectively

	a (Å)	c (Å)	d_{U-U} (Å)	V_{mol} (m ³ /molU.)
UPt ₃	5.752	4.897	4.13	42.3×10^{-6}
UPd ₃	5.770	9.631	4.11	41.8×10^{-6}

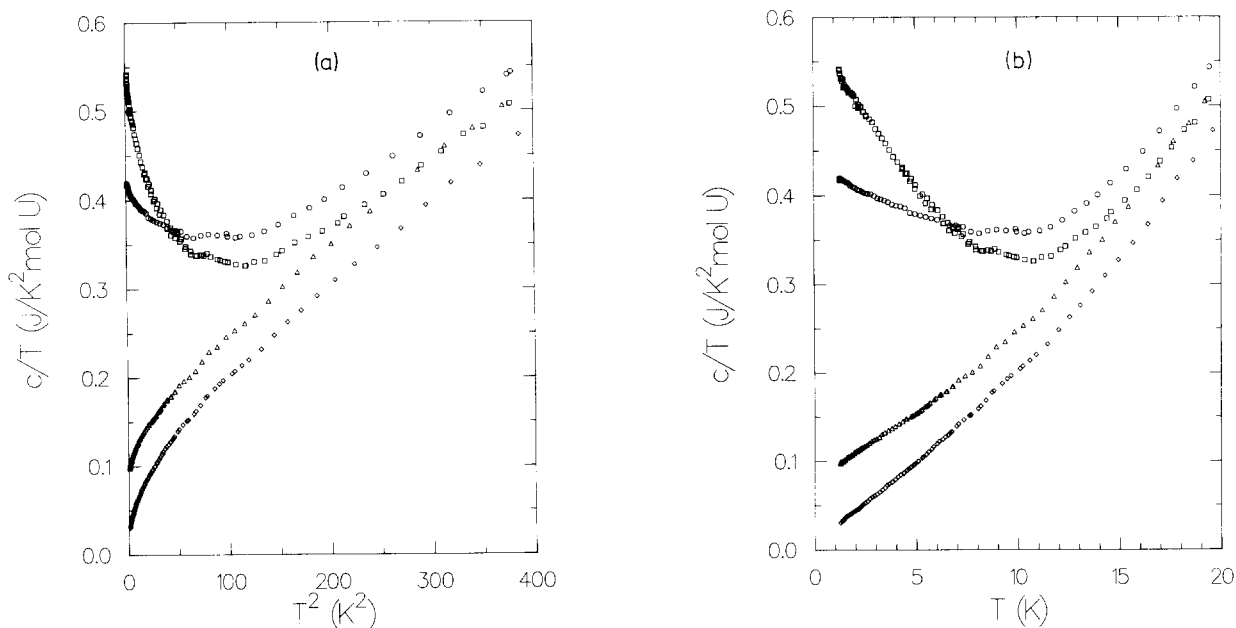


Fig. 4. Specific-heat data for $U(Pt_{1-x}Pd_x)_3$ compounds for $x = 0$ (\circ), 0.1 (\square), 0.2 (\triangle) and 0.3, (\diamond) in a plot of c/T vs. T^2 (a) and of c/T vs. T (b); the entropy difference between the curves for $x = 0.1$ and 0.2 in fig. 4b below 16 K amounts to 2.8 J/KmolU; data from ref. [43].

correspond with unenhanced γ -values of 14.9 [54] and 17.5 [55] mJ/K²mol, suggesting that the total enhancement due to spin-fluctuation and electron-phonon effects is equal to a factor of 30 or 24, respectively. Since the magnetic and transport properties of UPt_3 are strongly anisotropic, one can not exclude anisotropies in the temperature dependence of the upper critical field for the different crystallographic directions. The study of these anisotropies is considerably hampered by the sample dependence of the superconducting properties. Non-linearities in the temperature dependence of the upper critical field near T_c have been observed in ac susceptibility [47] and resistivity [50,56,57] measurements. The origin of these non-linearities is not understood, and is presently under study [52].

The specific-heat data on UPt_3 reveal a broad anomaly just below the resistive transition temperature with values for the relative jump in the specific heat of 1/4 to 1/3 of the BCS result [9,21,27]. At using entropy considerations it has been argued [27] that the coefficient of the linear term in the specific heat is at most a few percent of that in the normal state indicating the pairing of the heavy-mass electrons in the superconducting state. If the T^2 -dependence of the specific heat holds down to the lowest temperatures, however, a

larger percentage can not be excluded, see also refs. [50,52].

The $U(Pt_{1-x}Pd_x)_3$ alloys, mentioned above, have also been investigated for their superconducting behaviour. No superconductivity has been observed, even not in the $x = 0.01$ sample in measurements down to 40 mK [43]. These results do not point to a close relation between superconductivity and spin fluctuations which one would expect in case of triplet superconductivity.

4. Additional considerations

The low-temperature physical properties of UPt_3 are unexampled. Large anomalies occur in nearly all investigated properties in the liquid helium temperature region. Although these anomalies are in most cases well-established from an experimental point of view, their interpretation does not suit the existing models for crystal-field effects, spin fluctuations or singlet superconductivity. In this last section we shall dwell on some inconsistencies that arise at analysing the experimental data. Let us start with the concept of spin fluctuations. Evidence for spin-fluctuation phenomena to be present in UPt_3 comes most evidently from the T^3

In T/T_{sf} term [56] in the specific heat. A fit of the normal-state data to the sum of a linear, a cubic and this logarithmic term holds from 0.5 up to 20 K. From analysing the specific heat data at temperatures above 16 K, the unenhanced value for the coefficient of the linear term and the phonon coefficient have been derived, resulting in a T_{sf} value of 26 K. A similar description of the thermal expansion, however, breaks down above 6 K and yields a value for T_{sf} of 11 K. According to spin-fluctuation theories, this break-down has to be expected at temperatures much lower than the characteristic temperature. In other words, a description of the specific heat with only the additional logarithmic term works too well and leads to the conjecture that this description must be accidental in the higher temperature region. Support for the concept of spin fluctuation in UPt₃, although in a qualitative way only, comes from the resistivity measurements: the quadratic temperature dependence at the lowest temperatures and the large value of the coefficient of this quadratic term, see fig. 5. In a magnetic field the spin-fluctuation contribution to the resistivity is expected to be depressed leading to a negative magnetoresistivity as far as spin fluctuations

are involved. The experiments on UPt₃ at 1.5 and 4.2 K show the opposite; a large and positive magnetoresistivity for field directions in the basal plane with a sharp peak in the magnetoresistivity at 21 T. If there is any negative term in the magnetoresistivity, it is completely immersed in this huge and positive term. A negative magnetoresistivity is observed indeed at 77 K [25] and amounts for the current along the hexagonal axis and the field in the basal plane to -1% at 35 T. As the origin of this huge and positive term is concerned we already mentioned the concurrence with the peak in the differential susceptibility and its possible explanation in terms of crystal-field effects. Down to the lowest temperature, however, no Schottky-type of term in the specific heat or thermal expansion is seen. A satisfactory description of the susceptibility in terms of crystal-field effects is also lacking. The value for the susceptibilities at 4.2 K along the different crystallographic directions are small compared to those of UPd₃. These reduced values could be a consequence of the large splitting (164 K) between the nonmagnetic singlet ground state and the first excited level for the hexagonal sites. A peak in the differential susceptibility at 20 T,

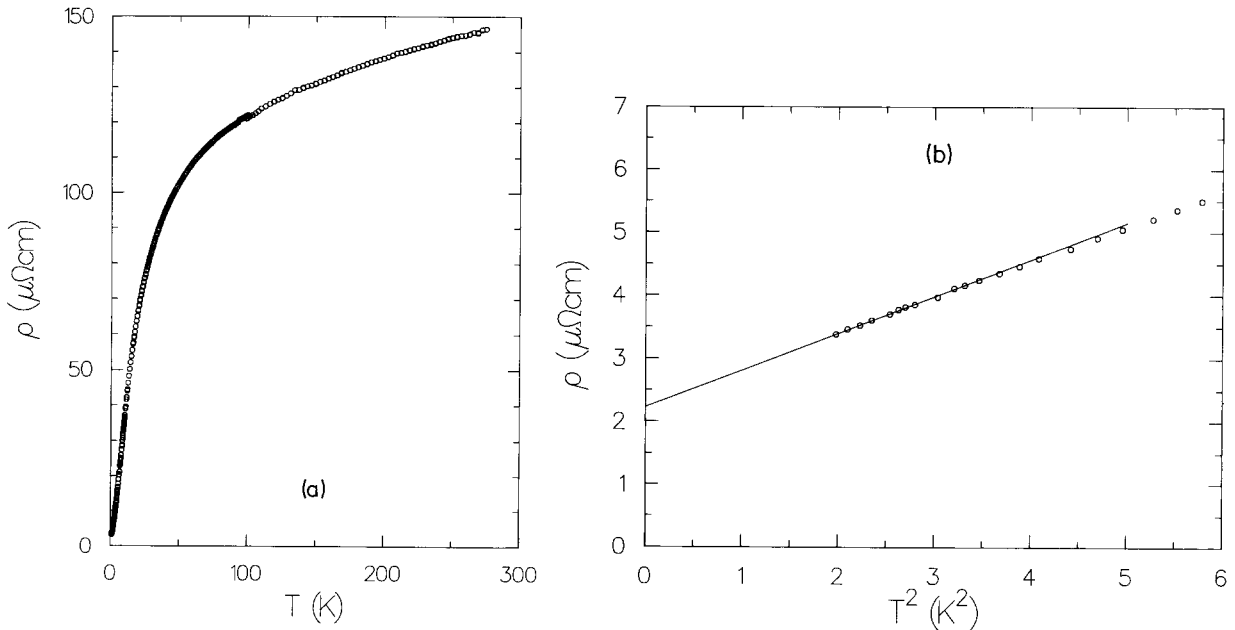


Fig. 5. Temperature dependence of the resistivity of an unannealed UPt₃ whisker (current along the *c*-axis) in a plot of ρ vs. T below 300 K (a) and in plot of ρ vs. T^2 between 1.4 K and 2.5 K; the residual resistance ratio, $\rho(300\text{ K})/\rho_0$ amounts to 66, the room-temperature value being equal to $148\ \mu\Omega\text{ cm}$; the uncertainty in the resistivity values due to geometrical factors is estimated to be $\pm 10\%$; the superconducting transition in the whisker occurs at 0.48 K; the whisker dimensions are approximately: $20\ \mu\text{m}$ (*a*-axis) $\times 40\ \mu\text{m}$ (*b*-axis) $\times 1000\ \mu\text{m}$ (*c*-axis), see ref. [8].

however, does not fit into this picture, nor a maximum value for the susceptibility at 16 K. Another possibility at explaining the susceptibility data is a much lower value for the crystal-field splitting, comparable to the quasi-cubic sites in UPd_3 or the hexagonal sites in $PrNi_5$. On this latter compound we performed high-field experiments at 4.2 K on the susceptibility of a polycrystalline sample with fields up to 32 T in order to study the field dependence of the differential susceptibility of a crystal-field compound. The results for $PrNi_5$ differ markedly from those for UPt_3 : the differential susceptibility of $PrNi_5$ is almost constant up to 10 T and starts to decrease at higher fields resulting in a value for the differential susceptibility at 30 T of about 30% of the low-field value; the UPt_3 -data are shown in fig. 2a. We are inclined for that reason to consider the maxima in the susceptibility to be related not to an excited crystal-field level but rather to the same many-body effects that cause the anomalies in the low-temperature thermal and transport properties. High-pressure experiments support this view since there is good agreement between the pressure effects on the resistivity and on the temperature at which the maximum in the susceptibility occurs. At considering the pressure dependence of the coefficient A of the term in the resistivity, quadratic in temperature, to represent the pressure dependence of the many-body phenomena, and at taking A proportional to T^{*-2} , the result for $\partial \ln T/\partial p$ is 25 Mbar^{-1} . Susceptibility measurements under pressure arrive at $\partial \ln \chi (4.2 \text{ K})/\partial p = -\partial \ln T_m/\partial p = -25 \text{ Mbar}^{-1}$, where T_m is the temperature at which the maximum in the susceptibility along the a - and b -directions occurs. Working in the spin-fluctuation model, the low-temperature Grüneisen parameter, Γ_c^{enh} , introduced in ref. [22] and connecting the spin-fluctuation contributions to specific heat with those to thermal expansion, is equal to the relative volume derivative of λ_{sf} , the spin-fluctuation enhancement factor in the specific heat, where λ_{sf} is proportional to $\ln S$, S being the Stoner factor. Taking T_{sf} inverse proportional to S we arrive at the expression: $\Gamma_c^{\text{enh}} = -(\ln S)^{-1} \partial \ln T_{sf}/\partial \ln V$. With values for S of 1.5, for the compressibility of 0.48 Mbar^{-1} and for Γ_c^{enh} of 159, taken from refs. [19,21], we find $\partial \ln T_{sf}/\partial p = 31 \text{ Mbar}^{-1}$. So we observe a close relation in the pressure derivative of the different characteristic temperatures as measured on the magnetic and thermal properties of UPt_3 , suggesting that they have a common base. Although in our present analysis the absolute values of the logarithmic pressure derivatives of the superconducting-transition temperature and the spin-fluctuation temperature are perhaps more deviating than indicated in ref. [48], there is no doubt

about their opposite signs. And as already mentioned before, this result seems to exclude electron pairing intermediated by spin fluctuations.

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References

- [1] D.E. Gardner and T.F. Smith, *Phys. Rev.* 154 (1967) 309.
- [2] J.G. Huber, M.B. Maple and D. Wohlleben, *J. Magn. Magn. Mat.* 1 (1975) 58.
- [3] J.J.M. Franse, P.H. Frings, A. van der Liet, A. Menovsky and A. de Visser, *Physica* 123B (1983) 11.
- [4] J.J.M. Franse, *J. Magn. Magn. Mat.* 45 (1984) 54.
- [5] G.R. Stewart, Z. Fisk, J.O. Willis and J.L. Smith, *Los Alamos Science* 10 (1984) 59.
- [6] J.J.M. Franse, *J. Magn. Magn. Mat.* 31–34 (1983) 819.
- [7] P.H. Frings, J.J.M. Franse, F.R. de Boer and A. Menovsky *J. Magn. Magn. Mat.* 31–34 (1983) 240.
- [8] A. Menovsky, J.J.M. Franse and A.C. Moleman, *J. Cryst. Growth* 67 (1984) 31.
- [9] G.R. Stewart, Z. Fisk, J.O. Willis and J.L. Smith, *Phys. Rev. Lett.* 52 (1984) 679.
- [10] A. Menovsky and J.J.M. Franse, *J. Cryst. Growth* 65 (1983) 286.
- [11] J.A. Pals, W. van Haeringen and M.H. van Maaren, *Phys. Rev.* B15 (1977) 2592.
- [12] L.E. De Long, J.G. Huber, K.N. Yang and M.B. Maple, *Phys. Rev. Lett.* 51 (1983) 312.
- [13] P.H. Frings, Thesis (Amsterdam, 1984).
- [14] A. Menovsky, J.C.P. Klaasse, R. Verhoef and J.J.M. Franse, 14-èmes Journées des Actinides, ed. J. Schoenes (Davos, 1984) p. 131.
- [15] W. Trzebiatowski, in: *Ferromagnetic Materials I*, ed. E.P. Wohlfarth (North-Holland, Amsterdam, 1980) p. 438.
- [16] B.S. Chandrasekhar and J.K. Hulm, *J. Phys. Chem. Solids* 7 (1958) 259.
- [17] A.F. Murray and W.L. Buyers, in: *Crystalline Electric Fields and Structural Effects in f-electron Systems*, eds. J.E. Crow, R.P. Guertin and T.W. Mihalisin (Plenum, New York, 1980) p. 257.
- [18] A. Musarik, J. Leciejewics and J. Zygmunt, *Phys. Stat. Sol.* (a) 28 (1975) K107.
- [19] J.J.M. Franse, P.H. Frings, A. de Visser, A. Menovsky, T.T.M. Palstra, P.H. Kes and J.A. Mydosh, *Physica* 126B (1984) 116.
- [20] G.R. Stewart, *Rev. Modern Phys.* 56 (1984) 755.

- [21] A. de Visser, J.J.M. Franse, A. Menovsky and T.T.M. Palstra, *J. Phys. F* 14 (1984) L191.
- [22] A. de Visser, J.J.M. Franse and A. Menovsky, *J. Phys. F* 15 (1984) L53.
- [23] M. Yoshizawa, B. Lüthi, T. Goto, T. Suzuki, B. Renker, A. de Visser, P.H. Frings and J.J.M. Franse, *J. Magn. Magn. Mat.* 52 (1985) 413.
- [24] P.H. Frings and J.J.M. Franse, *Phys. Rev.* B31 (1985) 4355.
- [25] A. de Visser, R. Gersdorf, J.J.M. Franse and A. Menovsky, *ICM85*.
- [26] A. de Visser, J.J.M. Franse and A. Menovsky, *J. Magn. Magn. Mat.* 43 (1984) 43.
- [27] J.J.M. Franse, A. Menovsky, A. de Visser, C.D. Bredl, U. Gottwick, W. Lieke, H.M. Mayer, U. Rauchschalbe, G. Sparn and F. Steglich, *Z. Phys.* B59 (1985) 15.
- [28] W.D. Schneider and C. Laubschat, *Phys. Rev.* B23 (1981) 997.
- [29] R.D. Parks, M.L. den Boer, S. Raaen, J.L. Smith and G.P. Williams, *Phys. Rev.* B30 (1984) 1580.
- [30] A.J. Arko, C.G. Olsen, D.M. Wieliczka, Z. Fisk and J.L. Smith, *Phys. Rev. Lett.* 53 (1984) 2050.
- [31] J.W. Allen, S.J. Oh, L.E. Cox, W.P. Ellis, M.S. Wire, Z. Fisk, J.L. Smith, B.B. Pate, I. Lindau and A.J. Arko, *Phys. Rev. Lett.* 54 (1985) 2635.
- [32] J. Schoenes and J.J.M. Franse, *Physica* 130B (1985) 69.
- [33] J. Ghijsen, R.L. Johnson, J.C. Spirlet and J.J.M. Franse, to be published.
- [34] H. Capellmann, S.M. Johnson, J.J.M. Franse, F. Steglich and K.R.A. Ziebeck, *ICM85*.
- [35] K. Andres, S. Darack and H.R. Ott, *Phys. Rev.* B19 (1979) 5475.
- [36] P. Zaplinski, D. Meschede, D. Plümacher, W. Schlabit and H. Schneider, in: *Crystalline Electric Field and Structural Effects in f-electron Systems*, eds. J.E. Crow, R.P. Guertin and T.W. Mihalisin (Plenum, New York, 1980) p. 295.
- [37] B. Lüthi and H.R. Ott, *Solid State Commun.* 33 (1979) 717.
- [38] M.S. Wire, J.D. Thompson and Z. Fisk, *Phys. Rev.* B30 (1984) 5591.
- [39] J.J.M. Franse, P.H. Frings, A. Menovsky and A. de Visser, *Physica* 130B (1985) 180.
- [40] A. de Visser, E. Louis, J.J.M. Franse and A. Menovsky, *ICM85*.
- [41] K. Andres, D. Davidov, P. Dernier, F. Hsu, W.A. Reed and G.J. Nieuwenhuys, *Solid State Commun.* 28 (1978) 405.
- [42] H.R. Ott, K. Andres and P.H. Schmidt, *Physica* 102B (1980) 148.
- [43] A. de Visser, J.C.P. Klaasse, M. van Sprang, J.J.M. Franse, A. Menovsky and T.T.M. Palstra, *ICM85*.
- [44] G.R. Stewart and A.L. Giorgi, *J. Low. Temp. Phys.* 59 (1985) 185.
- [45] T.T.M. Palstra, P.H. Kes, J.A. Mydosh, A. de Visser, J.J.M. Franse and A. Menovsky, *Phys. Rev.* B30 (1984) 2986.
- [46] J.L. Tholence, J. Flouquet, J.J.M. Franse and A. Menovsky, *ICM85*.
- [47] A. de Visser, J.J.M. Franse, A. Menovsky and T.T.M. Palstra, *Physica* 127B (1984) 442.
- [48] J.O. Willis, J.D. Thompson, Z. Fisk, A. de Visser, J.J.M. Franse and A. Menovsky, *Phys. Rev.* B31 (1985) 1654.
- [49] O.T. Valls and Z. Tesanović, *Phys. Rev. Lett.* 53 (1984) 1497.
- [50] F. Steglich, U. Ahlheim, J.J.M. Franse, N. Grewe, D. Rainer and U. Rauchschalbe, *J. Magn. Magn. Mat.* 52 (1985) 54.
- [51] F. Steglich, U. Rauchschalbe, U. Gottwick, H.M. Mayer, G. Sparn, N. Grewe, U. Poppe and J.J.M. Franse, *J. Appl. Phys.* 57 (1985) 3054.
- [52] A. Sulpice, J. Gandit, J. Chaussy, J. Flouquet, D. Jaccard, P. Lejay and J.L. Tholence, to be published.
- [53] D.J. Bishop, C.M. Varma, B. Batlogg, E. Bucher, Z. Fisk and J.L. Smith, *Phys. Rev. Lett.* 53 (1984) 1009.
- [54] P. Strange and B.L. Gyorffy, *Physica* 130B (1985) 41.
- [55] J. Sticht and J. Kübler, *Solid State Comm.* 54 (1985) 389.
- [56] J.W. Chen, S.E. Lambert, M.B. Maple, Z. Fisk, J.L. Smith, G.R. Stewart and J.O. Willis, *Phys. Rev.* B30 (1984) 1583.
- [57] D. Jaccard, J. Flouquet, P. Lejay and J.L. Tholence, *Ann. de Chim.*, to be published.
- [58] S. Doniach and S. Engelsberg, *Phys. Rev. Lett.* 17 (1966) 750.