

## INVITED PAPER

### NORMAL PHASE OF HEAVY-FERMION $UPt_3$

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A review is presented of the normal-state properties of the heavy-fermion superconductor  $UPt_3$  ( $\gamma = 422 \text{ mJ/K}^2 \text{ molU}$ ). At present a large variety of experiments has been performed on good quality single crystals. These studies include specific heat, thermal expansion, susceptibility, electrical resistivity, magnetostriction etc., as well as the application of high-magnetic fields and high pressures. Most of these experiments reveal pronounced anomalies at low temperatures. The thermal properties can be fairly described in a spin-fluctuation model. The magnetic properties are strongly anisotropic. On alloying  $UPt_3$  with Pd, additional anomalies, probably of antiferromagnetic nature, are introduced in the material. In spite of a tremendous effort to unravel the intriguing low-temperature properties of  $UPt_3$ , some of them remain quite puzzling. Fortunately the interest from theoretical side in heavy-fermion compounds is growing. In this respect the application of Fermi-liquid theory to  $UPt_3$  is discussed. In addition, recent data on the temperature dependence of the magnetoresistance and on the pressure dependence of the susceptibility, obtained on single-crystalline  $UPt_3$  samples, and thermal expansion data on polycrystalline  $U(Pt,Pd)_3$  compounds, are presented.

#### 1. Introduction

The heavy-fermion systems are characterized by a huge value for the coefficient of the linear term in the specific heat ( $\gamma \cong 400 \text{ mJ/K}^2 \text{ mol}$ ) [1]. Such a linear term exists, however, only in a limited temperature range. Therefore the  $\gamma$ -value is here merely defined as the ordinate obtained by extrapolating the specific heat curve, plotted as  $c/T$  vs  $T^2$ , to zero temperature. In the class of the heavy-fermion systems  $UPt_3$  takes a special place, being the only compound which specific heat can be described in a theoretical model, that is to say the spin-fluctuation model [2-5]. Superconductivity in  $UPt_3$  is observed near 0.5 K [3,4]. The unusual combination of strong spin fluctuations and superconductivity has led to speculations on odd-parity superconductivity [4,6]. However, this point is far from being settled and the precise nature of the superconducting state is still subject of lively discussions. A survey of experimental data, on both the normal and superconducting state, has been presented in refs [7] and [8]. In ref [8] we emphasized that no satisfactory description of the low-temperature properties of  $UPt_3$  in terms of crystal-field effects, spin-fluctuation phenomena, antiferromagnetic order and singlet or triplet superconductivity can be offered. In order

to unravel the intriguing low-temperature properties of  $UPt_3$ , a number of additional experiments have been performed since then. Among these, alloying experiments provide perhaps the most powerful tool.

In this paper we focus on the normal phase of  $UPt_3$ . We start by reviewing its thermal, magnetic and transport properties. Afterwards we turn to a Fermi-liquid analysis of the thermal properties. In the last section we discuss the alloying experiments and their implications for the normal phase of  $UPt_3$ .

#### 2. Thermal properties

The specific heat of  $UPt_3$  obeys the relationship

$$c = \gamma^* T + \beta T^3 + \delta T^3 \ln(T/T^*) \quad (1)$$

Here the enhanced coefficient of the linear term  $\gamma^*$  and the  $T^3 \ln(T/T^*)$ -term account for the spin-fluctuation effects and  $\beta$  is the usual phonon coefficient. In order to perform a computer fit to the data it is more convenient to express the specific heat as

$$c = \gamma^* T + \beta^* T^3 + \delta T^3 \ln T, \quad (2)$$

where  $\beta^* = \beta + \beta_{sf}$  and where  $\beta_{sf} = -\delta \ln T^*$

Table 1

Coefficients of 3-parameter fits to the measured (I) and phonon corrected (II) specific heat of UPt<sub>3</sub>, according to eq. (2) and various parameters derived in a Fermi-liquid analysis, employing  $k_F = 1.06 \text{ \AA}^{-1}$ . The meaning of the symbols is explained in the text.

Fit	Interval (K)	$\gamma^*$ (a)	$\beta^*$ (b)	$\beta_{ph}$ (b)	$\beta_{sf}$ (b)	$\delta$ (b)	$m^*$ (m <sub>e</sub> )	$T_f$ (K)	$T^*$ (K)	$B^*$	$A_0^+$	$F_0^+$	$g_1$
I	1.2–20	422	-3.72	0.85	-4.57	1.38	180	275	27	83.74	-3.71	-0.788	-0.452
I	1.2–10	421	-3.53	0.85	-4.38	1.30	180	276	29	79.45	-3.64	-0.785	-0.440
II	1.2–20	424	-3.44	-	-3.44	0.965	181	274	35	57.73	-3.24	-0.764	-0.373
II	1.2–10	418	-2.77	-	-2.77	0.716	179	278	48	44.71	-2.95	-0.747	-0.325

Units: (a) mJ/K<sup>2</sup> mol U, (b) mJ/K<sup>4</sup> mol U

represents the contribution from spin-fluctuation effects to the  $T^3$ -term. The coefficients deduced from such a fit to the specific heat data of a single-crystalline sample [3] are listed in table 1 (see also fig. 1). The  $\gamma^*$ -value enables one to determine the effective mass of the electrons, provided the Fermi wavenumber,  $k_F$ , is known. The latter can be obtained from an analysis of the temperature derivative of the upper critical field (-4.4 T/K) [3],  $k_F = 1.06 \text{ \AA}^{-1}$ , or from

configurational arguments,  $k_F = (3\pi^2 Z/\Omega)^{1/3} = 1.08 \text{ \AA}^{-1}$ , assuming that 6 f-electrons ( $Z = 6$ ) per unit cell volume ( $\Omega = 140.9 \text{ \AA}^3$ ) take part in the formation of the electron liquid. With the first value of  $k_F$ , we obtain the effective mass  $m^*/m_e = 3\hbar^2 \gamma^*/(k_B^2 k_F V_m m_e) = 180$ , the Fermi temperature  $T_F = \hbar^2 k_F^2 / 2k_B m^* = 275 \text{ K}$ , and the Fermi velocity  $v_F^* = \hbar k_F / m^* = 6800 \text{ m/s}$ . Note that the Fermi velocity has the same order of magnitude as the sound velocity [9]. From band-structure calculations  $\gamma$ -values ranging from 14.9 to 22.5 mJ/K<sup>2</sup> mol U have been deduced [10–14]. Although these calculations reveal a certain enhancement they cannot account for the huge electronic specific heat that is observed at low temperatures.

In order to determine the purely electronic contribution to  $c$ , knowledge of the phonon contribution is indispensable. Unfortunately, no information on a non-magnetic analog to UPt<sub>3</sub> is available. From the published phase diagrams the existence of, for instance, ThPt<sub>3</sub> is not clear. Nevertheless two groups performed XPS-measurements on a ThPt<sub>3</sub> sample, but gave no details about its structure [15,16]. Frings [17] estimated  $\beta$  at 0.85 mJ/K<sup>2</sup> mol U ( $\theta_D = 210 \text{ K}$ ), a value derived from a two parameter fit to the specific heat of a polycrystalline UPt<sub>3</sub> sample (with  $\gamma_0 = 225 \text{ mJ/K}^2 \text{ mol U}$ ). This value for  $\beta$  nicely agrees with the one obtained from sound velocity measurements at room temperature 0.76 mJ/K<sup>2</sup> mol U ( $\theta_D = 217 \text{ K}$ ) [9]. This enables us to calculate the characteristic temperature  $T^*$ , which is often called the spin-fluctuation temperature. Mathematically it is the temperature at which the  $\delta T^3 \ln T$ -contribution and the  $\beta_{sf} T^3$ -

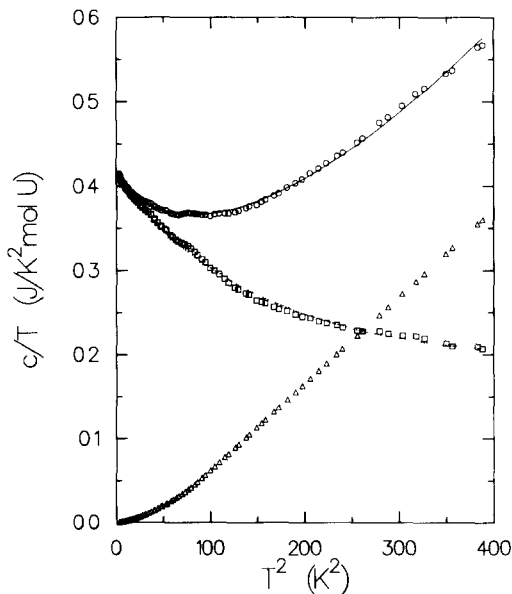


Fig. 1. Measured (○) from ref. [3], calculated phonon (△) from ref. [18], and phonon-corrected (□) specific heat of UPt<sub>3</sub>. The full and dotted lines represent 3-parameter fits to the measured and phonon-corrected specific heat, with coefficients listed in table 1.

contribution to the specific heat are equal but opposite in sign  $T^* = e^{-\beta_c/\delta}$ . With the first value for  $\beta$  we deduce  $T^* = 27$  K. Recently, inelastic neutron experiments enabled Renker et al [18] to determine the phonon density of states of UPt<sub>3</sub>, and hence to calculate the lattice contribution to the specific heat in a more sophisticated way. At temperatures between 9 and approximately 20 K the Debye temperature amounts to 200 K (close to the value we quoted above), whereas below 9 K it gradually increases to a value of 320 K at 1 K. The electronic specific heat, after correcting for this lattice contribution, is shown in fig. 1 as well. The parameters of a fit to the purely electronic specific heat according to eq. (2), with  $\beta^* = \beta_{st}$ , are listed in table 1. This fit is as good as the fit to the non-phonon corrected specific heat, an important difference, however, is the 30% smaller  $\delta$ -value.  $T^*$  equals in this case 35 K.

The linear thermal-expansion coefficient,  $\alpha$ , of UPt<sub>3</sub> (hexagonal MgCd<sub>3</sub>-type of structure [19]) is strongly anisotropic [9]. The dilatation in the basal plane is positive and passes through a maximum at roughly 10 K, whereas along the hexagonal axis the dilatation is negative up to 40 K and has a minimum at 15 K. Assuming proper Gruneisen parameters, i.e. assuming that the free energy can be written as  $F = f(T/T_i)$ , where  $T_i$  represents a characteristic temperature like the Fermi temperature (for  $\Gamma_e$ ) or the Debye temperature (for  $\Gamma_{ph}$ ), one expects identical terms in the specific heat and in the thermal expansion (see also section 5). A fit to the volume expansion,  $\alpha_v$ , in the temperature range 1.2–20 K, including a  $T^3 \ln(T/T^*)$ -term, works, however, not as good as for the specific heat. The  $T^*$ -value deduced from this fit equals 29 K. The Gruneisen parameter, as deduced from the measured linear terms in  $c$  and  $\alpha$ , amounts to the large value of 69 at 1 K [9,20].

At applying a magnetic field, the specific heat at low temperatures is hardly affected [2]. In a field of 8 T a small increase of the  $\gamma$ -value is observed (2%) [21], as expected from thermodynamics, since  $\partial^2 \chi / \partial T^2 > 0$  [2,15]. The specific heat of UPt<sub>3</sub> under pressure has been measured by Brodale et al [21]. These authors find a strong suppression of the coefficients  $\gamma^*$ ,  $\beta_{st}$  and  $\delta$ .

### 3. Magnetic properties

The magnetic properties of UPt<sub>3</sub> are strongly anisotropic. The susceptibility reveals a maximum near 17 K, for field directions in the basal plane [2,15] (see also fig. 2). Although this maximum first has been interpreted as a sign of antiferromagnetic order [15], it is more likely to associate it with spin-fluctuation effects [22]. At 17 K no sign of magnetic order has been observed in the specific heat. The effective moment, deduced from a Curie-Weiss analysis, amounts to  $2.9 \mu_B$  per U-atom. Such a value cannot be explained in the Russell-Saunders coupling scheme. On one hand this might be caused by the presence of crystal fields at high temperatures, on the other hand the large centrifugal forces on the electron orbits ask for the application of intermediate coupling. The high-field magnetisation reveals a strong increase of the (induced) moment at fields between 15 and 25 T, for field directions in the basal plane and temperatures below 20 K [2,5,23]. At 35 T the magnetic moments in the basal plane are rather large, amounting to  $1 \mu_B$  per U-atom. The forced magnetostriction, measured on a single-crystalline sample, for different field and dilatation

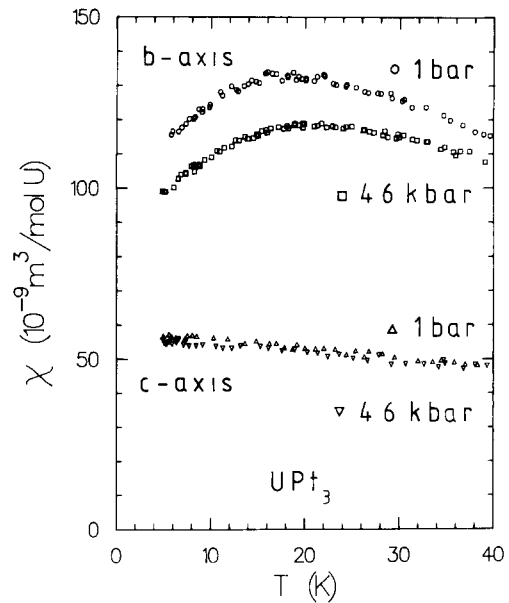


Fig. 2 Susceptibility of single-crystalline UPt<sub>3</sub> for field directions ( $B = 5$  T) and pressures as indicated

directions, is also strongly anisotropic [24]. The volume magnetostriction is roughly one order of magnitude larger for a field direction in the basal plane than for a field along the hexagonal axis. Applying thermodynamic relations we could predict from these magnetostriction data values of  $-23$  and  $-3 \text{ Mbar}^{-1}$  for the pressure dependence of the susceptibility,  $\partial \ln \chi / \partial P$ , with the field applied in the basal plane and along the hexagonal axis, respectively. This has been confirmed by pressure experiments on single-crystalline samples [25]. From the data in fig. 2 we deduce values for  $\partial \ln \chi / \partial P$  of  $(-28 \pm 3)$  and  $(-5 \pm 3) \text{ Mbar}^{-1}$  at  $4.2 \text{ K}$ , respectively. The pressure effect on  $T_{\text{max}}$  is positive,  $\partial \ln T_{\text{max}} / \partial P = (30 \pm 6) \text{ Mbar}^{-1}$ , quite close to the value obtained on a polycrystalline sample [26]. These data imply a suppression of the spin-fluctuation effects at applying hydrostatic pressures. The occurrence of spin-fluctuation effects in UPt<sub>3</sub> is furthermore supported by inelastic neutron experiments [27].

#### 4. Transport properties

The anisotropic electrical resistivity,  $\rho$ , of UPt<sub>3</sub> can be described in a spin-fluctuation model as well [28]. It reveals a  $T^2$ -dependence below  $2 \text{ K}$  [28,26,29], with coefficients of  $1.6$  and  $0.7 \text{ } \mu\Omega \text{ cm/K}^2$  for the basal plane and hexagonal axis, respectively, several orders of magnitude larger than observed for an ordinary metal. The sharp increase of  $\rho$  at low temperatures is followed by a tendency to saturate at values of  $240$  (basal plane) and  $130 \text{ } \mu\Omega \text{ cm}$  (hexagonal axis) at room temperature. UPt<sub>3</sub> is the only heavy-fermion system that reveals a monotonically rising  $\rho(T)$ , even up to  $1000 \text{ K}$  [29]. All other heavy-fermion systems reveal a Kondo-like resistance [1]. From resistivity under pressure (upto  $200 \text{ kbar}$ ) a strong suppression of the spin-fluctuation phenomena has been concluded [28,26,30].

The anomaly in the magnetisation near  $20 \text{ T}$  is reflected as a pronounced maximum in the magnetoresistance [7,8,31]. The magnetoresistance in a field of  $8 \text{ T}$  ( $B \parallel I \parallel a$ -axis) is positive below  $14 \text{ K}$ , and negative above this temperature (see fig. 3). The maximum at  $7 \text{ K}$  shifts towards lower temperatures with decreasing field. A positive  $\Delta\rho$  at

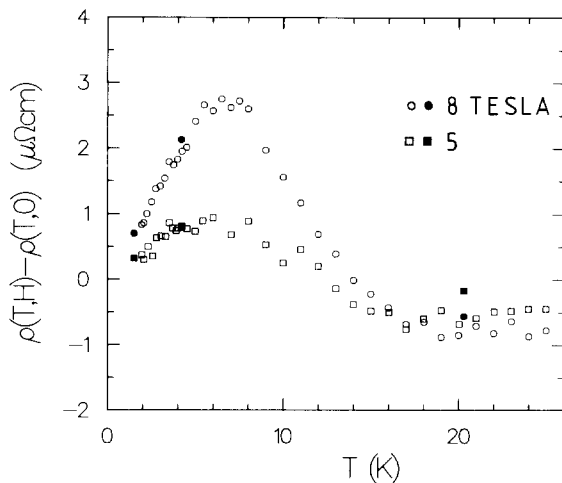


Fig. 3 Magnetoresistance of single-crystalline UPt<sub>3</sub> ( $B \parallel I \parallel a$ -axis) as function of temperature in a field of  $5 \text{ T}$  ( $\square$ ) and  $8 \text{ T}$  ( $\circ$ ). Closed symbols are taken from ref. [31].

low temperatures, in combination with a maximum as function of field, has also been observed in several CeCu<sub>2</sub>Si<sub>2</sub> and CeAl<sub>3</sub> samples [32]. An extrapolation of the data in fig. 3 to  $0 \text{ K}$  results in a field independent  $\rho_0$ . Sulpice et al. [33], however, observed a linear field dependence  $\rho_0(H) = \rho_0 + aH$ , with  $a = 0.22 \text{ } \mu\Omega \text{ cm/T}$ . This implies a  $\Delta\rho_0$  of  $1.76 \text{ } \mu\Omega \text{ cm}$  at  $8 \text{ T}$ , which is not observed in our case. This discrepancy might be due to the differences in the  $\rho$ -values of the samples:  $3.2 \text{ } \mu\Omega \text{ cm}$  [33] and  $9 \text{ } \mu\Omega \text{ cm}$  (this work) at  $1 \text{ K}$ .

Anomalies have also been observed in other transport properties, such as heat conductivity [29], thermopower [29], and Hall effect [34–36].

#### 5. Fermi-liquid analysis

The large electronic specific heat that is observed in the heavy-fermion systems points to a strongly interacting electron liquid. Therefore it is tempting to apply Fermi-liquid theory [37]. Pethick and Carneiro [38] have shown that small momentum transfer processes, such as spin-fluctuations, lead to a  $T^3 \ln(T/T^*)$ -term in the specific heat. With this term they could successfully account for the specific heat of the eminent Fermi-liquid <sup>3</sup>He. In close analogy herewith Coffey and Pethick [39] and Pethick et al. [40] applied a Fermi-liquid model to UPt<sub>3</sub>. An im-

portant role in their approach plays the coefficient of the  $T^3 \ln(T/T^*)$ -term, which is given by  $\delta = 3\pi^2 \gamma^* B^s / 10 T_F^2$ . The quantity  $B^s$  is a function of the Landau scattering amplitudes,  $A_l^\lambda = F_l^\lambda / (1 + F_l^\lambda / (2l + 1))$ , where  $\lambda = s$ ,  $a$  refers to spin-symmetric and spin-antisymmetric terms of the interaction  $F_l^\lambda$  represent the Landau parameters. Retaining only terms with  $l = 0$  and taking  $A_0^s = 1$  [40],  $B^s$  depends on  $A_0^a$  only. The coefficients  $\delta$  and  $\gamma^*$  enable one then to determine  $A_0^a$ , provided  $k_F$  is known (see section 2). Results for the different parameters, obtained from various specific heat fits, are listed in table 1. Pethick et al subsequently use the value for  $A_0^a$  in an expression for the superconducting transition temperature that was first applied to the superfluid transition in <sup>3</sup>He by Patton and Zaringhalam [41],

$$T_c^{(j)} = 1.13 \epsilon T_F e^{1/g_j} \quad (3)$$

Here  $\epsilon$  is a numerical constant,  $T_F$  is the Fermi temperature and  $g_j$  represents a pseudopotential. If  $g_j < 0$ , the potential is attractive,  $j = 0$  or 1 indicates singlet or triplet pairing. The  $g_j$ 's can also be written as a function of the Landau scattering amplitudes. Assuming only small momentum transfer processes, neglecting components with  $l \geq 2$  and with estimates of  $A_l^\lambda$  from various normal state properties one finds a negative  $g_1 = (1 + A_0^a)/6$  and  $g_1 < g_0$  (see table 1). Hence triplet pairing is favoured. The same conclusion was reached by Valls and Tešanović within a somewhat simplified Fermi-liquid model [42]. Unfortunately, it is not possible to calculate  $T_c$  from the normal state properties, since  $\epsilon$  is not known. However, one may test eq (3) by considering its pressure derivative assuming a pressure independent  $\epsilon$ ,

$$\partial \ln T_c / \partial P = \partial \ln T_F / \partial P - 6(1 + A_0^a)^{-2} \partial A_0^a / \partial P \quad (4)$$

Data on the pressure dependence of the specific heat [21] may serve to determine the pressure dependence of  $A_0^a$  and  $T_F$ . Comparing the thus calculated value for  $\partial \ln T_c / \partial P$ ,  $-44 \text{ Mbar}^{-1}$  [40], with the one derived from experiments,  $-26 \text{ Mbar}^{-1}$  [26], one finds a reasonable agreement. One can also determine the right hand side of eq (4) from the thermodynamic relationship between the specific heat at zero pressure and

the thermal expansion. Writing the thermal expansion conform

$$\alpha = \gamma_\alpha^* T + \beta_\alpha T^3 + \delta_\alpha T^3 \ln(T/T^*), \quad \text{one has} \quad (5)$$

$$\gamma_\alpha^* = -\frac{1}{3V} \frac{\partial \gamma}{\partial P} \quad \text{and} \quad \delta_\alpha = -\frac{1}{9V} \frac{\partial \delta}{\partial P} \quad (6)$$

Using the coefficients obtained by fitting the uncorrected specific heat data (see table 1), and fitting the thermal expansion data to eq 5 in the temperature range 1.2–20 K ( $\gamma_\alpha^* = 1.08 \times 10^{-6} \text{ K}^{-2}$ ,  $\beta_\alpha = -1.77 \times 10^{-8} \text{ K}^{-4}$  and  $\delta_\alpha = 5.27 \times 10^{-9} \text{ K}^{-4}$ ), we obtain  $\partial \ln T_c / \partial P = -19 \text{ Mbar}^{-1}$ , in good agreement with the experimental value. With the  $\gamma^*$ ,  $\beta$  and  $\delta$  values deduced from the phonon corrected specific heat we obtain  $\partial \ln T_c / \partial P = -125 \text{ Mbar}^{-1}$ . This large value for the calculated relative pressure dependence of  $T_c$  is mainly caused by the lower  $\delta$ -value derived for this fit. Also, at reducing the temperature range for which the various fits are made, a considerably larger value for  $\partial \ln T_c / \partial P$  is observed.

## 6. Alloying experiments

In order to further elucidate the low-temperature properties of UPt<sub>3</sub>, useful information might be obtained from alloying experiments. A first candidate for substitutions at the Pt site can be found in iso-electronic Pd. Specific heat [8,43–46], susceptibility [44], high-field magnetisation [44] and resistivity [47] measurements have been performed on a number of polycrystalline U(Pt<sub>1-x</sub>Pd<sub>x</sub>)<sub>3</sub> compounds, for  $x \leq 0.30$ . For low Pd concentrations the  $\gamma^*$ -value increases with respect to pure UPt<sub>3</sub>, amounting upto 600–700 mJ/K<sup>2</sup>mol U for a 10% sample. This has been interpreted as an enhancement of the spin-fluctuation phenomena ( $T^* \approx 10 \text{ K}$  for the 10% sample). At higher Pd content the  $\gamma^*$ -value rapidly drops and the spin-fluctuation phenomena are suppressed. In accordance herewith we observe a shift with increasing Pd concentration of the anomalies, present for pure UPt<sub>3</sub> at 17 K in the susceptibility, and at 21 T (at 4.2 K) in the magnetisation, towards lower temperatures and fields, respectively. An interesting feature of these compounds is the occurrence of additional

anomalies [43,44] These appear as pronounced peaks in the specific heat at 3.6, 5.8 and 5.5 K for the 2, 5 and 7% Pd sample, respectively. At applying a magnetic field of 5 T these anomalies shift to slightly lower temperatures. In resistivity measurements, on polycrystalline samples, as well as on single-crystalline whiskers [47], these anomalies turn up as a minimum in  $\Delta\rho/\Delta T$ . In fig. 4 we present thermal expansion measurements on some selected polycrystalline  $U(Pt_{1-x}Pd_x)_3$  compounds. The maximum observed for pure  $UPt_3$  at 12 K [9,48], shifts towards lower temperatures with increasing Pd content. The thermal expansion of a 15% Pd compound resembles that of the *c*-axis of  $UPt_3$ . Clearly, the additional anomalies at 3.6 and 5.8 K for the 2 and 5% Pd sample, are also reflected in the thermal expansion. Since the thermal expansion coefficient is strongly anisotropic, and preferred orientations are present in these samples, a more quantitative analysis awaits data on single crystals. Susceptibility and high-field magnetisation data on a single crystal of the  $U(Pt_{0.95}Pd_{0.05})_3$  compound can be found in ref. [45]. Recently, neutron experiments have revealed that the anomaly in a 5% Pd sample at 5.8 K is associated with the onset of antiferromagnetic order [49]. This suggests that the compound  $UPt_3$  is close to an antiferromagnetic instability. Specific heat [46,50], susceptibility [50] and resistivity [50] measurements on Th doped  $UPt_3$  reveal rather similar anomalies. However, these have been interpreted as caused by Fermi surface instabilities (spin density waves), due to doping induced strains. It would be interesting to see how far antiferromagnetism in the Pd and spin density waves in the Th doped compounds are related, as is the case, for instance, in Chromium. From resistivity measurements on the 5% Pd compound the relative pressure dependence of the Néel temperature,  $\partial \ln T_N / \partial P$ , has been determined at  $-55 \text{ Mbar}^{-1}$  [51]. Note that the temperature, at which the maximum in the susceptibility of pure  $UPt_3$  is observed, increases with pressure (see fig. 2), indicating a different nature of  $T_{\text{max}}$  and  $T_N$ . The occurrence of superconductivity in the  $U(Pt_{1-x}Pd_x)_3$  compounds depends strongly on the Pd content, as follows from the  $T_c$ 's of the 0.1 and 0.2% Pd samples [45].

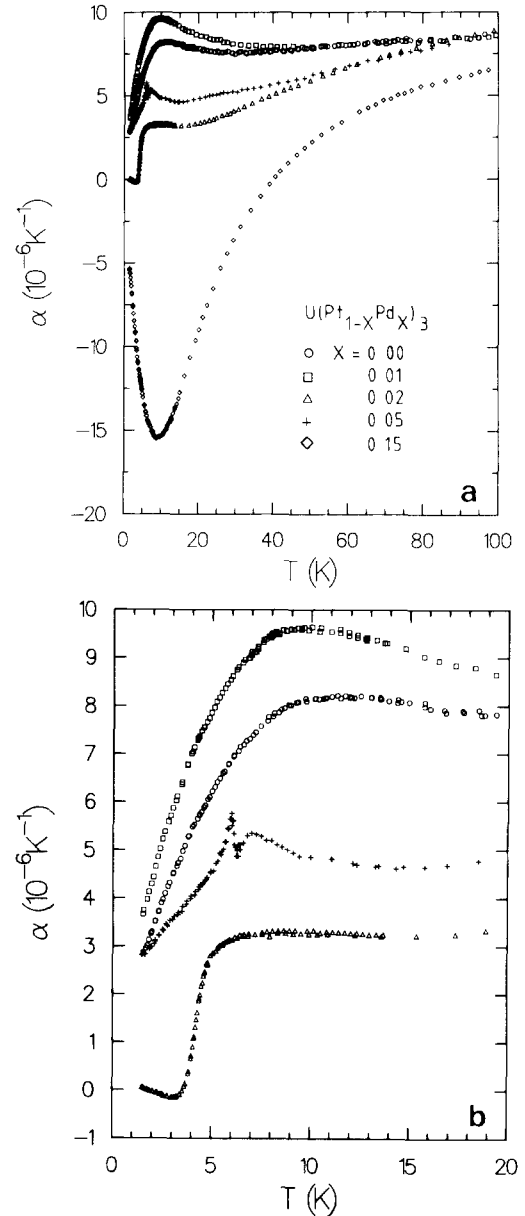


Fig. 4 (a) and (b) Linear thermal expansion coefficient of polycrystalline  $U(Pt_{1-x}Pd_x)_3$  compounds, for  $x = 0.00$  ( $\circ$ ),  $0.01$  ( $\square$ ),  $0.02$  ( $\triangle$ ),  $0.05$  ( $+$ ) and  $0.15$  ( $\diamond$ )

Superconductivity has not been observed in a 0.5% Pd sample [44]. What the precise correlation between the occurrence of superconductivity and antiferromagnetism in these pseudobinary  $U(Pt, Pd)_3$  compounds is, asks, however, for more detailed investigations.

It is a pleasure to thank F E Kayzel for experimental assistance at performing the thermal expansion measurements. This work is part of the research program of the Dutch "Stichting FOM"

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