

## Magnetism in UPt

This article has been downloaded from IOPscience. Please scroll down to see the full text article.

1998 J. Phys.: Condens. Matter 10 10643

(<http://iopscience.iop.org/0953-8984/10/47/015>)

View [the table of contents for this issue](#), or go to the [journal homepage](#) for more

Download details:

IP Address: 171.66.16.210

The article was downloaded on 14/05/2010 at 17:57

Please note that [terms and conditions apply](#).

## Magnetism in UPt

Karel Prokeš<sup>†‡§</sup>, Jacques C P Klaasse<sup>‡</sup>, Isameldin H Hagnusa<sup>‡</sup>,  
Alois A Menovsky<sup>‡</sup>, Ekkehard H Brück<sup>‡</sup>, Frank R de Boer<sup>‡</sup> and  
Toshizo Fujita<sup>†</sup>

<sup>†</sup> Department of Quantum Matter, ADSM, Hiroshima University, 739 8526 Higashi-Hiroshima, Japan

<sup>‡</sup> Van der Waals–Zeeman Institute, University of Amsterdam, 1018 XE Amsterdam, The Netherlands

Received 10 August 1998, in final form 14 September 1998

**Abstract.** Bulk studies of UPt show clearly that this material orders magnetically below 28 K. In contrast to numerous previous studies we found that the low-temperature properties can be explained only by the presence of an energy gap (22–35 K) in the dispersion relation of magnons which leads to typical exponential temperature dependencies of magnetic, thermal and electrical transport properties. Another magnetic phase transition around 19 K, found by means of magnetization measurements on an as-cast sample, is most likely due to the presence of a minority phase.

### 1. Introduction

Although the itinerant ferromagnetic system UPt has been a subject of intensive studies for three decades [1–5], a big portion of uncertainty about the size of the U magnetic moments, the type of magnetic anisotropy and other physical properties still persists. This compound has attracted considerable attention due to strong pressure dependence of the magnetization whereas the magnetic phase transition temperature of about 27 K was reported to remain unchanged [2, 4, 5]. Several studies have revealed that the saturation magnetization at ambient pressure is strongly sample dependent, scattering significantly around  $0.45 \mu_B/U$  [2, 4] although neutron-diffraction work of Frings *et al* [5] has revealed a much higher moment of  $1.1 \pm 0.4 \mu_B/U$ .

The main obstacle in attempts to determine the physical properties of UPt more precisely is serious metallurgical problems because this system is formed by a solid-state reaction between U and UPt<sub>2</sub> [6]. UPt was first reported to adopt the orthorhombic CrB type of structure [1–4, 7, 8]. Later on, Dommann *et al* [9, 10] suggested that UPt forms, like UIr, in the monoclinic PdBi type of structure. Neutron-diffraction studies of Frings *et al* [5] showed that at room temperature two sets of Bragg peaks are present. One of them is consistent with the PdBi type of structure and the second one can be interpreted in terms of superstructure lines of the CrB type. Finally, recently it has been published that high-resolution neutron-diffraction experiments are not fully in agreement with both the PdBi and the CrB structure [11]. The fact that in most of the cases the samples prepared by melting of constituent elements consist of two structural phases depending on the heat treatment

§ On leave from: Department of Metal Physics, Charles University, 12116 Prague 2, The Czech Republic.

and pressure history is the reason for rather controversial reports on the magnetic properties of this appealing material. Due to the presence of two structural phases, two magnetic phase-transition temperatures to ferromagnetic order at 19 K and 27 K were reported [2, 4]. It has been suggested by Dommann *et al* [9] and later shown by Franse *et al* [6] by means of neutron diffraction that the '27 K' magnetic phase transition is connected with ferromagnetic ordering in PdBi-type ordered UPt. The '19 K' transition was shown to be most probably connected with ordering in some other related structure (probably FeB type). It was suggested that this other structure transforms under pressure to PdBi type, resulting in a decrease of the magnetic moment which is consistent with bulk measurements [5]. Here, we report on magnetic and other physical properties of UPt prepared by a novel method.

## 2. Sample preparation and characterization

As has been mentioned in the introduction, all samples prepared by arc-melting of appropriate amounts of the constituting elements consist of two structural phases depending on the heat treatment and on whether the sample has been exposed to external pressure. The reason for this can be found in the fact that UPt forms in a peritectoid solid-state reaction between U and UPt<sub>2</sub>. We have tried to prepare UPt by a novel method, namely by evaporating In from a melt which contains an appreciable amount of this element. An electrical arc in a continuously gettered Ar atmosphere and a water-cooled copper crucible were used. After evaporation of the In, we have continued in arcing the ingot for about five hours keeping only a small part of it liquid. No additional heat treatment was given to the obtained ingot.

The quality of the resulting material has been inspected by electron microprobe analysis (EPMA). The material was found to consist of two phases. The matrix (97.4 vol.%) was found to be homogeneous with composition deviating from the ideal stoichiometry of UPt by no more than 1 at.% (the resolution limit of EPMA). The second phase (2.6 vol.%) is found to be U-rich, most probably UO<sub>2</sub>, metallic U or U with 4 at.% of Pt (UPt forms, if prepared by an ordinary way, by a solid-state reaction from U(Pt) and UPt<sub>2</sub>). Due to distribution of the second phase within the matrix we were unable to determine unambiguously its composition. We note that no traces of In were found.

The structural parameters have been determined by x-ray diffraction. For this purpose, UPt was ground under inert atmosphere and investigated by Cu K $\alpha$  radiation ( $10^\circ < 2\theta \leq 100^\circ$ ). The diffraction pattern was analysed by means of Rietveld refinement with the program Fullprof [12]. Our refinement indicates that UPt crystallizes in the monoclinic structure with the space group  $P2_1$  (No 4,  $Z = 8$ ). The structural parameters which agree well with literature values [5, 10] are given in table 1. No other reflections were observed in the diffracted pattern which prevented us from identifying the secondary phase mentioned above. On the other hand, it suggests that its content is small, in agreement with the EPMA result. We note that the refinement does not indicate any deviation from the ideal 1:1 stoichiometry.

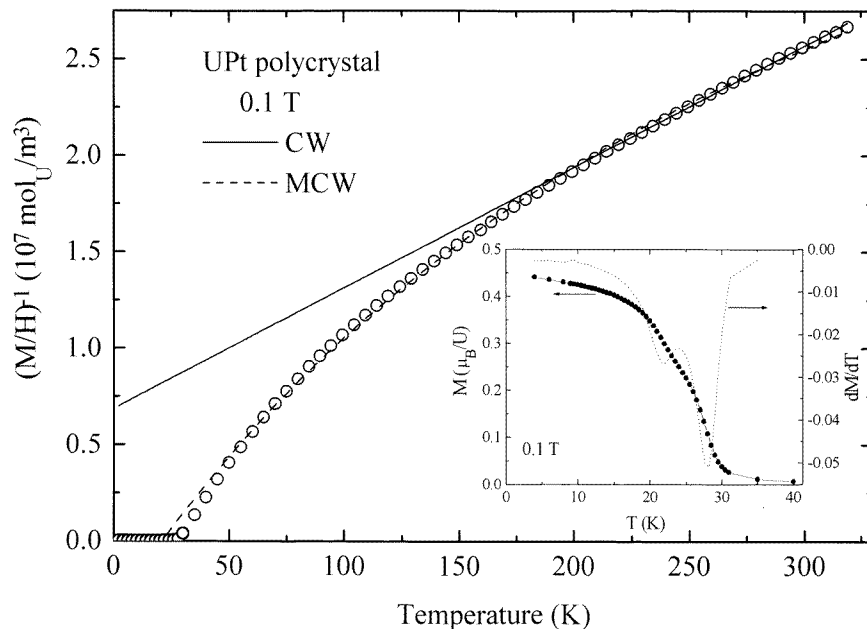
## 3. Results

### 3.1. Magnetic susceptibility and magnetization

The temperature dependence of the magnetic susceptibility  $M/H$  was measured between 2 K and 320 K in fields up to 5 T by means of a Quantum Design SQUID magnetometer. In order to eliminate effects arising from magnetic anisotropy, a fine powdered sample with

**Table 1.** Refined structural parameters for UPt at room temperature.

Space group: $P2_1$					$T = 293$ K
Atom	Site	$x$	$y$	$z$	
U(1)	2(a)	0.118(4)	0(0)	0.113(6)	
U(2)	2(a)	0.609(5)	0.007(5)	0.624(7)	
U(3)	2(a)	0.771(4)	0.728(4)	0.361(4)	
U(4)	2(a)	0.369(5)	0.731(3)	0.878(5)	
Pt(1)	2(a)	0.136(6)	0.271(4)	0.127(7)	
Pt(2)	2(a)	0.559(4)	0.268(4)	0.594(5)	
Pt(3)	2(a)	0.893(5)	0.444(4)	0.394(7)	
Pt(4)	2(a)	0.369(5)	0.456(4)	0.881(7)	
Lattice parameters			$R$ factors		
$a = 570.66 \pm 0.14$ pm	$\alpha = 90^\circ$		$R_p = 3.28\%$		
$b = 1079.81 \pm 0.23$ pm	$\beta = 100.15^\circ \pm 0.01^\circ$		$R_{wp} = 4.61\%$		
$c = 576.72 \pm 0.13$ pm	$\gamma = 90^\circ$		$\chi^2 = 3.48$		



**Figure 1.** The temperature dependence of the inverse magnetic susceptibility ( $\chi = M/H$ ) measured on a fixed-powder sample at 0.1 T. The solid and dashed lines represent best fits to a Curie–Weiss (CW) law and a modified Curie–Weiss (MCW) law, respectively. In the inset, the temperature dependence of the magnetization measured at 0.1 T together with its temperature derivative are shown. Clearly, two maxima are observed.

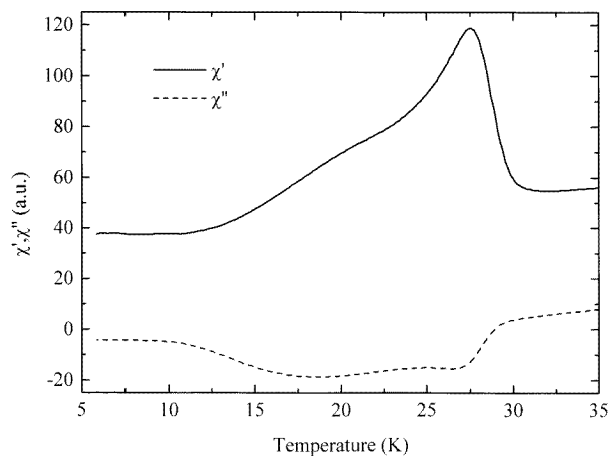
grains fixed in random orientation by diamagnetic glue was used in these measurements. The contributions of the sample holder and glue to the susceptibility have been subtracted.

In figure 1, the temperature dependence of  $(M/H)^{-1}$  of the fixed-powder sample measured at 0.1 T is shown. The solid line represents a Curie–Weiss (CW) law between 200 and 320 K which yields an effective moment  $\mu_{eff} = 3.18 \pm 0.02 \mu_B/U$  and a

paramagnetic Curie temperature  $\theta_p = -110 \pm 3$  K. We note that even at the highest measured temperatures the  $\chi^{-1}$ - $T$  curve exhibits significant curvature so that the effective moment is likely to be higher. This result is in agreement with literature results of  $3.55 \mu_B/U$  [6, 13] derived from data up to 1000 K. We also performed a modified Curie–Weiss law (MCW) type of fit which includes a temperature-independent term  $\chi_0$  and yields a reasonable agreement with experimental data above 50 K. The fit parameters are  $\mu_{eff} = 1.97 \pm 0.02 \mu_B/U$ , a paramagnetic Curie temperature  $\theta_p = 21.0 \pm 0.8$  K and  $\chi_0 = 1.72 \times 10^{-8} \text{ m}^3 \text{ mol}_U^{-1}$ . It should be emphasized that the description of the susceptibility of polycrystalline, magnetically anisotropic, material in terms of an MCW picture should only be considered as a parametrization of the data and not as a physical description due to a mixing of different CW branches. However, it can serve as clear evidence for presence of strong magnetic anisotropy in UPt and as a parametrization of this anisotropy.

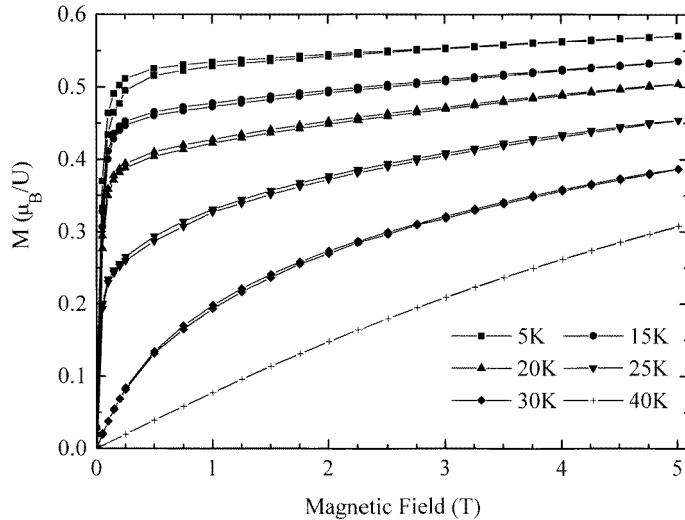
In the inset of figure 1 we show the temperature dependence of the magnetization of UPt measured at 0.1 T. The temperature derivative of magnetization, which is also shown in the inset, reveals maxima at 21.8 K and 27.8 K suggesting existence of two magnetic phase transitions. Two transitions, albeit at 19 K and 27 K (or only one transition), have been observed by several other authors [3, 4, 13, 14]. The anomaly around 22 K is present only in data taken at low fields (smaller than 1 T). When a higher field is applied, no anomaly is seen at this temperature region.

In figure 2, the temperature dependence of the ac susceptibility is shown, measured at low temperatures on a bulk piece. Two transitions, estimated to be situated around 22.5 and 27.7 K, respectively, are visible. These transition temperatures agree well with the dc magnetic susceptibility results.



**Figure 2.** The temperature dependence of the real (solid line) and imaginary (broken line) parts of the ac susceptibility measured on bulk UPt.

The field dependence of magnetization was measured at various temperatures on a fixed-powder sample by means of a Quantum Design SQUID magnetometer in fields up to 5 T and is presented in figure 3. At low temperatures, the magnetization sharply increases, showing a much slower, nearly linear, increase above 0.1 T. At 5 K, the spontaneous magnetization  $\mu_S$ , obtained by extrapolation to zero field, amounts to  $0.52 \mu_B/U$ . This value agrees well with literature values [2, 4, 13]. The magnetization shows a slight hysteresis and the values



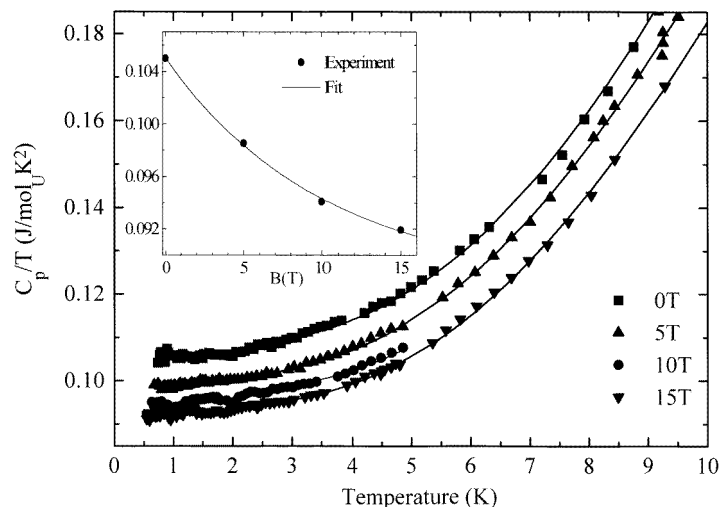
**Figure 3.** The field dependence of magnetization as measured on fixed-powder sample in fields up to 5 T at various temperatures. Note that the hysteresis loop is extremely narrow.

of the magnetization at zero field (about  $0.06 \mu_B/U$ ) and the remanent field (about 0.01 T) are extremely small. As the temperature increases, the saturation magnetization decreases and the hysteresis loop gets even narrower. At 28 K, the spontaneous magnetization equals zero indicating the proximity of the magnetic phase transition, in agreement with magnetic susceptibility measurements.

### 3.2. Specific heat

The specific heat was measured by a standard semi-adiabatic method using two experimental set-ups. The first one was used to measure the specific heat in zero field between 2 and 90 K while the second has been used for measurements between 0.4 and 40 K in zero field and in magnetic fields up to 15 T. The zero-field results agree well in the region of overlap. It should be noted that at low temperatures in the former equipment the signal due to the sample was about ten times larger than the signal due to the addenda. However, at 30 K the signal of the sample amounts to only 10% of the total measured heat capacity which accounts for appreciable scatter of data at higher temperatures (and in magnetic fields) obtained in this equipment [15, 16].

In figure 4, the low-temperature part of the temperature dependence of  $C_p/T$  measured in fields up to 15 T is shown.  $C_p/T$  cannot be satisfactorily described by a  $\gamma + aT^2$  dependence, which contains only the electronic part  $\gamma$  and the phonon contribution and neglects the contribution due to magnetic order. For a good description a term which involves magnetic excitations with a gap in the dispersion relation has to be introduced. Such a term has been used in the literature to describe the contribution of magnetic excitations with an energy gap to the low-temperature specific heat properties of rare-earth elements [17, 18]. The best fits of  $C_p/T$  against  $T$  between 0.4 K and 9 K at various fields to  $\gamma + aT^2 + fT^{1/2} \exp(-\Delta/T)$ , where  $\Delta$  denotes a gap in the (otherwise isotropic) dispersion relation of ferromagnetic magnons, are represented in figure 4 by solid lines. It is quite clear that experimental data and fits agree rather well. The low-temperature specific-heat



**Figure 4.** The low-temperature part of the temperature dependence of  $C_p/T$  of bulk UPt measured in fields up to 15 T. Solid lines through symbols are the best fit to the expression given in the text involving a gap in the dispersion relation of ferromagnetic magnons. In the inset we show the field dependence of the Sommerfeld coefficient  $\gamma$  (points) together with the best fit as described in the text.

coefficient  $\gamma$  found for zero field is either comparable with values published in literature [3, 4, 13], slightly smaller than a published value (by  $10 \text{ mJ mol}^{-1} \text{ K}^{-2}$ ) [6] or higher (by  $25 \text{ mJ mol}^{-1} \text{ K}^{-2}$ ) [14]. From the numerical results given in table 2, it follows that while the numerical factors  $a$  and  $f$  and the value of the gap  $\Delta$  ( $\sim 22 \text{ K}$ ) are practically unaffected by applied magnetic field, the low-temperature specific-heat coefficient decreases with increasing field. At 15 T, the decrease amounts to 12.5% of the zero-field value. Such a decrease of  $C_p/T$  with increasing field, which has not been reported until the present, suggests that at least a part of the low-temperature specific heat is of magnetic origin. The dependence of  $\gamma$  which is shown in the inset of figure 4 on magnetic field  $\mu H = B$  can be approximated by exponential decay  $\gamma(B) = \gamma(\infty) + b \exp(-B/\Delta_B)$ , where  $\gamma(\infty)$  represents the low-temperature specific-heat coefficient in the infinite-field limit, and  $\Delta_B$  describes how fast  $\gamma$  decays with increasing field  $B$ . The fitted parameters are  $\gamma(\infty) = 87.91 \pm 1.32 \text{ mJ mol}^{-1} \text{ K}^{-2}$ ,  $b = 17.11 \pm 1.27 \text{ mJ mol}^{-1} \text{ K}^{-2}$  and  $\Delta_B = 10.17 \pm 1.50 \text{ T}$ . By taking into account experimental errors, the value of  $\gamma(\infty) = 87.91 \text{ mJ mol}^{-1} \text{ K}^{-2}$  agrees well with the zero-field value determined by De Podesta *et al* [14].

In figure 5, the temperature dependence of the specific heat is shown in the  $C_p/T-T$  representation in zero field (open points) and at 15 T (closed symbols). As can be seen, zero-field data are dominated by a very well defined peak at 28 K suggesting magnetic phase transition. Nothing is seen around 19 K which suggests that the amount of the ‘19 K’ phase is small. It is remarkable that the fit, which is shown in figure 5 by a full line through the open symbols, introduced in the preceding paragraph, describes the data quite well nearly up to the magnetic phase transition. The fitting parameters obtained between 2 and 20 K are given in table 2. The peak which appears at zero field at 28 K is very sensitive to magnetic field. It is smeared out already at 5 T and the magnetic entropy is shifted to higher temperatures in agreement with findings of other authors [14].

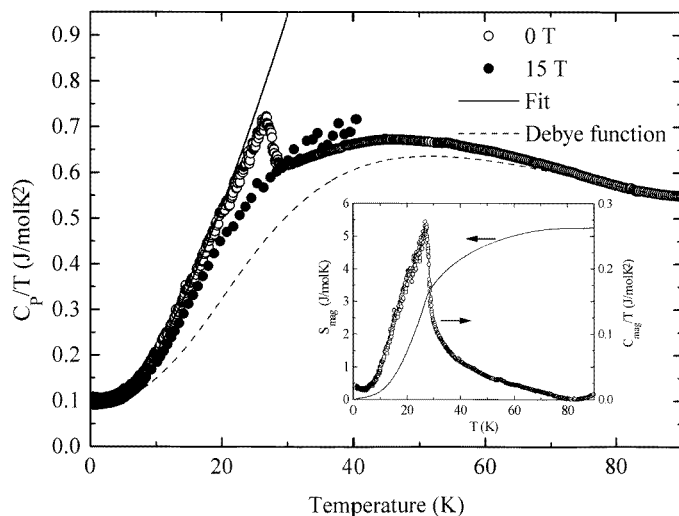
In order to determine the magnetic entropy we need to estimate the phonon and electronic contributions to specific heat. Several attempts have been proposed in the literature

**Table 2.** Fitted parameters of the low-temperature specific-heat of UPt at various magnetic fields together with Debye temperature calculated according to  $\theta_D^3 = 2 \times 12\pi^4 R/5a$ .

Field (T)	$\gamma$ ( $\text{mJ mol}^{-1} \text{K}^{-2}$ )	$a$ ( $10^{-4} \text{ J mol}^{-1}$ )	$f$ ( $\text{J mol}^{-1} \text{K}^{-5/2}$ )	$\Delta$ (K)	$\theta_D$ (K)
0 <sup>a</sup>	104.77(21)	4.66(18)	0.159(4)	22.1(2)	203
0	104.99(15)	5.07(18)	0.165(4)	23.4(4)	197
5	98.54(14)	4.60(18)	0.143(4)	21.9(2)	203
10	94.10(10)	5.08(27)	b	b	197
15	91.93(11)	4.85(13)	0.148(3)	22.0(2)	200

<sup>a</sup> Fit between 2 and 20 K obtained in different set-up.

<sup>b</sup> No reliable values obtained due to lack of data above 5 K; at low temperatures the exponential term does not contribute substantially to the specific heat.



**Figure 5.** The temperature dependence of the specific heat in zero field in the  $C_P/T$ - $T$  representation measured in a different experimental set-up than the results shown in figure 4 and measured up to 90 K. The solid line through the symbols is the best fit to the expression given in the text (the same type of fit was used to fit the data in figure 4). The broken line denotes the sum of the Debye function which was used to approximate the phonon contribution and of the electronic term  $\gamma$  as described in the main text. The temperature dependence of the magnetic part  $C_{mag}/T$  and the resulting magnetic entropy  $S_{mag}$  are given in the inset.

[3–6, 13, 14]. The non-magnetic isostructural compounds ThPt, NiPt or the Debye function have been used to estimate the lattice contribution. However, it is not clear what value should be used for the electronic contribution which may lead to substantially different values of the magnetic entropy. It seems quite reasonable to use for the electronic contribution the value  $\gamma(\infty) = 87.91 \text{ mJ mol}^{-1} \text{K}^{-2}$  derived in the preceding paragraph. To account for the phonon contribution, which is shown in figure 5 by a broken line, we have used the Debye function with  $\theta_D = 190 \text{ K}$  which provides, together with the electronic contribution, good agreement with the experimental data in the high-temperature limit. Debye temperatures between 197 and 203 K have been calculated according to the formula  $\theta_D^3 = 2 \times 12\pi^4 R/5a$  for different magnetic fields (see table 2) which are close to the  $\theta_D = 190 \text{ K}$  used in the analysis and in the literature. The temperature dependence of the magnetic entropy  $S_m$ ,

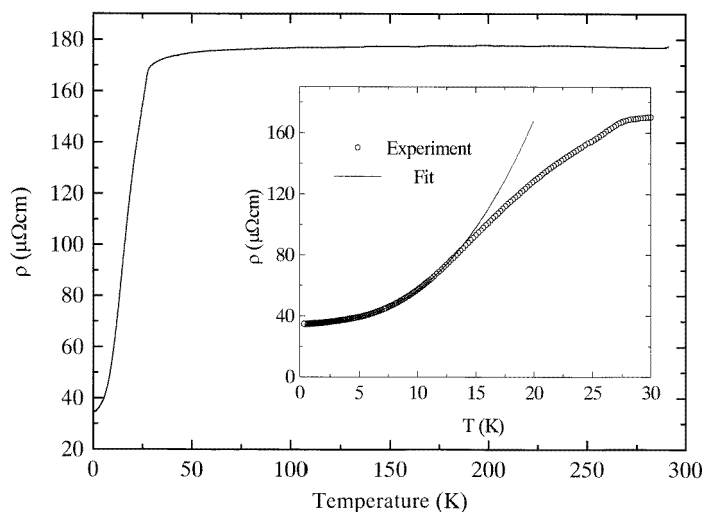


which is shown in the inset of figure 5, has then been calculated by integrating magnetic part of  $C_P/T$  up to 85 K and amounts to  $5.2 \pm 0.3 \text{ J mol}^{-1} \text{ K}^{-1}$ . This value which can be expressed as  $0.9R \ln 2$ , accidentally agrees well with the value derived by Luengo *et al* [3] but it is significantly higher than values derived by other authors [4–6, 13, 14]. In applied magnetic fields, the magnetic entropy is shifted towards higher temperatures. At 15 T, 91% of magnetic entropy is attained at 40 K, the highest temperature measured in high fields. A similar result has been obtained by De Podesta *et al* [14].

### 3.3. Electrical resistivity

The electrical resistivity was measured on a small bar-shaped sample by the standard AC four-point method in the 260 mK–290 K temperature range. For the measurements below 4 K, a  $^3\text{He}$  refrigerator was used. The electrical contacts were established with silver paint.

The temperature dependence of the electrical resistivity is shown in figure 6. It is very similar to results reported earlier [4, 6, 11]. At high temperatures, the electrical resistivity slightly increases with lowering temperature. Around 200 K, it starts to decrease and below approximately 27 K it drops drastically due to appearance of ferromagnetic order. We do not see any sudden change in the slope around 19 K suggesting that amount of the ‘19 K’ phase is small. Instead of that a smooth change is found. The  $\rho_{290 \text{ K}}/\rho_{260 \text{ mK}}$  ratio amounts to 5.2. This value is slightly lower than the ratio of 6.3 found by Rodríguez-Fernández *et al* [11] for  $\rho_{290 \text{ K}}/\rho_{4.2 \text{ K}}$  but higher than about 3.5 found by Franse *et al* [6].



**Figure 6.** The temperature dependence of the electrical resistivity. The low-temperature part is shown in the inset. The full line through symbols is the best fit (between 0 and 12 K) to the expression given in the text.

The low temperature part has been reported to behave according to  $\rho = \rho_0 + a_\rho T^2$  with  $a_\rho = 0.19 \mu\Omega \text{ cm K}^{-2}$  [4] and  $0.45 \mu\Omega \text{ cm K}^{-2}$  [19]. Our analysis clearly indicates that the low-temperature part cannot be satisfactorily described by this type of dependence. Instead of that, we found that it can be well fitted by the expression  $\rho = \rho_0 + aT^2 + E\Delta T(1 + 2T/\Delta) \exp(-\Delta/T)$ , where  $E$  depends on the spin-disorder resistivity and the electron–magnon coupling constant and  $\Delta$  denotes the gap in the dispersion relation of magnons. This expression was derived for describing the electron–magnon interaction

in rare-earth elements [17, 18] and we have used a similar expression, derived by Andersen and Smith [18] for description of the specific heat. In fact, the values of  $\Delta$  derived from the specific heat data and the electrical resistivity data should be equal. The best fit to data below 12 K yields  $\rho_0 = 34.663 \pm 0.003 \mu\Omega \text{ cm}$ ,  $a_\rho = 0.1827 \pm 0.0004 \mu\Omega \text{ cm K}^{-2}$ ,  $E = 0.226 \pm 0.005 \mu\Omega \text{ cm K}^{-2}$  and  $\Delta = 34.2 \pm 0.4 \text{ K}$ . The data (points) and the fit (solid line) are shown in the inset of figure 6. It should be, however, kept in mind that our sample is polycrystalline and a strong magnetic anisotropy reflects very often also in an anisotropy of transport properties, which would require to modify substantially the testing function (residual resistivity, the gap and the dispersion relation of magnons etc are isotropic).

#### 4. Discussion

The electronic structure of actinides is characterized by a large spatial extent of the 5f wavefunctions. Therefore, the 5f electron states in many actinides participate in chemical bonding and strongly interact with the environment. In this respect, we distinguish direct 5f–5f interaction on one side and hybridization of the 5f states with the valence states of neighbouring atoms (5f–ligand hybridization) in the crystal lattice on the other. Clearly, the most important parameter concerning the two-ion (5f–5f or 5f–ligand) interaction is the spacing between the U atom and the atom in question and on geometrical conditions around the U atom. The significance of the former parameter has been pointed out by Hill [20]. The effect of both mechanisms on the magnetic order is twofold. On one side it promotes magnetic ordering because both mechanisms effectively contribute to interaction between magnetic moments (if we look to this problem from the localized point of view). On the other, however, it leads, in the strong hybridization limit, to destruction of the 5f magnetic moments. The former feature leads to higher ordering temperatures in actinide systems as compared to rare-earth isostructural compounds containing, except for U, the same elements. As a consequence of the latter fact, the 5f states in actinides are, as a rule, to a certain extent delocalized and the magnetic moments are smaller than expected for a free  $\text{U}^{3+}$  or  $\text{U}^{4+}$  ion. It is evident that the hybridization contributes also to the enhancement of magnetic anisotropy of a type depending on the crystal structure. In this respect, one can speak about the hybridization-induced magnetic anisotropy. Moreover, highly correlated 5f-electron states are expected to form a narrow band pinned at the Fermi surface. The high density of states at  $E_F$  is projected into high values of the specific heat and into anomalous transport properties.

Spacing between U atoms in UPt falls into the critical region for uranium compounds (3.4–3.6 Å) [20]. Therefore, we may anticipate the majority of properties mentioned in the preceding paragraph. It is evident that UPt does exhibit anomalous magnetic, thermal and electrical-transport properties. The high-temperature magnetic susceptibility can be described in terms of a Curie–Weiss law with an effective moment close to  $5f^2$  or  $5f^3$  electron configuration, i.e. in the framework of localized moments. At lower temperatures, however, this picture is inadequate. The saturation magnetization, magnetic entropy and other physical properties point to the itinerant nature of ferromagnetic order in UPt. Similar behaviour has been observed for isostructural UIr [9]. From the temperature dependence of the specific heat a value of  $0.9R \ln 2$  has been derived for magnetic entropy for UPt. This value is too small for a localized system but too large for a system with magnetic order purely of itinerant origin [21]. To elucidate this point more, we can evaluate the Kadowaki–Woods ratio  $a_\rho/\gamma^2$  [22], where  $a_\rho$  denotes the coefficient of the  $T^2$  term of the electrical resistivity and the Wilson ratio  $R_W = (\chi_0/\gamma)(\pi^2 k_B^2/3\mu_B^2)$  [23], where  $\mu_B$  is a Bohr magneton,  $\chi_0$  denotes the temperature independent part of the magnetic susceptibility

expressed in  $\text{emu mol}^{-1}$  and  $\gamma$  is expressed in  $\text{erg mol}^{-1} \text{K}^{-2}$ ). It is expected that the former coefficient will be close to a universal value of  $1.0 \times 10^{-5} \mu\Omega \text{ cm} (\text{mJ mol}^{-1} \text{K}^{-1})^{-2}$  if the same quasiparticles participate in both the electrical conduction and the low-temperature specific heat properties. The latter coefficient,  $R_W$ , is of the order of unity for a free electron gas and can be enhanced due to electron–electron correlations if the enhancement of  $\chi_0$  and  $\gamma$  has the same origin. From the values  $a_\rho/\gamma^2 = 1.65 \times 10^{-5} \mu\Omega \text{ cm} (\text{mJ mol}^{-1} \text{K}^{-1})^{-2}$  and  $R_W = 1.0$  we can conclude that the magnetism in UPt is governed mainly by itinerant electron states which participate in both transport and magnetic properties. The slightly negative temperature derivative of the electrical resistivity at high temperatures points to a strong hybridization between the 5f electron states and conduction electrons. The high absolute value of the spin-disorder electrical resistivity can be taken as further support for this statement. Rodríguez-Fernández *et al* [11] showed that in  $\text{U}_{1-x}\text{La}_x\text{Pt}$  compounds, both the residual and the spin-disorder resistivity are strongly dependent on the content of La. This points to the fact that the scattering of conduction electrons is critically dependent on the actual position and shape of 5f bands in the vicinity of the Fermi level. Finally, the presence of 5f electron states at the Fermi limit can be deduced also from the enhanced low-temperature specific-heat coefficient which amounts in zero field to  $105 \text{ mJ mol}^{-1} \text{K}^{-2}$ .

Let us consider a possible influence of secondary phases on the physical properties of our sample. From the x-ray diffraction and electron microprobe analysis we were able to conclude that our sample contains about 2.6 vol.% of U-rich phase. We think that  $\text{UO}_2$ , metallic U or U with 4 at.% of Pt are the most probable candidates. It is well known that metallic U does not order magnetically [24] with a rather low value of the low-temperature specific-heat coefficient. Alpha-U exhibits three phase transitions at 43, 37 and 22–23 K which are connected with charge-density waves (CDWs). Around 43 and 37 K we do not observe any anomalies in our data and the only coincidence is found around 22 K. The transition in  $\alpha$ -U around 22–23 K is associated with a lock-in of one the incommensurate components of CDW to a commensurate value [24]. Correspondingly, very little if any influence on the magnetic properties of our sample is expected.  $\text{UO}_2$ , on the other hand, orders antiferromagnetically at 30.8 K with U moments of  $1.74 \mu_B/\text{U}$  [25]. Clearly, at this temperature, no anomaly is visible in our data. Therefore, we have to conclude that we cannot explain the anomaly seen in the temperature dependence of the magnetic susceptibility around 22 K by the presence of impurities suggested. One can argue, however, that the anomaly at 22 K can be identified as the anomaly seen previously at 19 K by other authors [2–6]. This is supported by the fact that usually two transitions, at 19 K and 27 K, were seen in studies of various UPt samples. Although this conclusion is very tempting we do not have any solid piece of evidence for this statement. The situation is complicated by the fact that there exists probably another structural phase of UPt which is neither monoclinic neither orthorhombic of the CrB type [11]. This phase might be responsible for the 22 K anomaly. Clearly, more work is necessary to elucidate this point.

From previously published works it was shown that the low-temperature properties of UPt can be described in the framework of a Fermi liquid system with an indication of spin waves known as magnons. Our measurements of the temperature dependence of the specific heat and the electrical resistivity, however, provides in addition to that a clear indication that an energy gap in the dispersion relation of magnons is present. The former temperature dependence leads to an energy gap of about 22 K while the latter one to about 35 K. Moreover, the energy gap seems to be insensitive to the applied magnetic field as evidenced by the specific heat measurements.

The existence of an energy gap can explain rather controversial observations of the low-temperature properties of UPt published earlier. If we accept that it is very difficult

to prepare UPt as single phased system, then it is not at all surprising that the size of the gap will be strongly sample dependent. In some cases, the gap  $\Delta$  can be nearly negligible, so that the low-temperature properties can be described well by ordinary ferromagnetic magnons. In some samples, however, it can be rather high, and the physical properties exhibit characteristic exponential temperature dependencies.

A sample-dependent saturation magnetization scattering significantly around  $0.45 \mu_B/U$  can be found in the literature [2,4]. Frings *et al* [5], however, published a much higher moment of  $1.1 \pm 0.4 \mu_B/U$  on the basis of neutron diffraction. At the same time they argued that UPt exhibits a uniaxial type of magnetic anisotropy which would explain the discrepancy between moment determined in different ways and the temperature dependence of the magnetic susceptibility. In the view of our studies, we came to the same conclusions. Measurements of magnetic susceptibility made on polycrystalline samples then would lead inevitably to curvature in  $\chi^{-1}$  against  $T$  which is also encountered experimentally and the strongly sample-dependent  $\mu_S$  values published in the literature are likely to be due to texture in bulk material.

## 5. Conclusions

By means of x-ray diffraction at room temperature we confirmed that UPt forms in monoclinic structure with space group  $P2_1$  (No 4,  $Z = 8$ ), as UIr does. Bulk measurements point to ferromagnetic order below  $T_C = 28$  K. The additional magnetic phase transition observed in an as-cast sample is most probably due to a minority phase with different crystal structure. From the temperature dependencies of bulk properties a strong influence of ferromagnetic magnons with a gap ( $\sim 22$ – $35$  K) in their dispersion relation can be concluded. The low-temperature specific-heat coefficient  $\gamma$  decreases exponentially with increasing field. The  $\chi^{-1}$ - $T$  curve measured on a polycrystalline sample exhibits a strong curvature which can be explained by a strong magnetic anisotropy of this material. At high temperatures it leads to an effective moment close to values expected for a free  $U^{3+}$  or  $U^{4+}$  ion. Saturation magnetization  $\mu_S = 0.52 \mu_B/U$  agrees well with literature values. UPt can be classified as an itinerant ferromagnetic material.

## Acknowledgments

This work was sponsored in part by the ‘Japanese Society for Promotion of Science (JSPS)’, by a grant-in-aid for scientific research from the Ministry of Education, Science, Sports and Culture of Japan (Mombusho) and by the ‘Stichting voor Fundamenteel Onderzoek der Materie’ (FOM).

## References

- [1] Matthias B T, Chu C W, Corenzwit E and Wohlleben D 1969 *Proc. Natl Acad. Sci. USA* **64** 459
- [2] Huber J G, Maple M B and Wohlleben D 1975 *J. Magn. Magn. Mater.* **1** 58
- [3] Luengo C A, Maple M B and Huber J G 1976 *J. Magn. Magn. Mater.* **3** 305
- [4] Frings P H and Franse J J M 1985 *J. Magn. Magn. Mater.* **51** 141
- [5] Frings P H, Vettier C, Dommann A, Hullinger F and Menovsky A A 1989 *Physica B* **156/157** 832
- [6] Franse J J M, Menovsky A A, de Visser A, Frings P H and Vettier C 1987 *J. Magn. Magn. Mater.* **70** 351
- [7] Dwight A E 1969 *Developments in the Structural Chemistry of Alloys Phases* ed B C Giessen (New York: Plenum) p 181
- [8] Lawson A C, Williams A, Huber J G and Roof R B 1986 *J. Less-Common Met.* **120** 113
- [9] Dommann A, Hullinger F, Sigriest T and Fisher P 1987 *J. Magn. Magn. Mater.* **67** 323

- [10] Dommann A and Hulliger F 1988 *Solid State Commun.* **65** 1093
- [11] Rodríguez-Fernández J, Argüelles M A, Espeso J I, Gómez Sal J C and McEwen K A 1997 *Physica B* **230–232** 86
- [12] Rodríguez-Carvajal J, *FULLPROF* version 3.1c, Jan96, ILL
- [13] Frings P H, Franse J J M, De Boer F R and Menovsky A A 1983 *J. Magn. Magn. Mater.* **31–34** 240
- [14] De Podesta M, Lester C, McEwen K A, Espeso J and Gómez Sal J C 1995 *J. Magn. Magn. Mater.* **140–144** 1359
- [15] Klaasse J C P 1997 *Rev. Sci. Instrum.* **68** 89
- [16] Klaasse J C P, Hagmusa I H and Bruck E 1997 *Rev. Sci. Instrum.* **68** 4208
- [17] Mackintosh A R 1963 *Phys. Lett.* **4** 140
- [18] Andersen N H and Smith H 1979 *Phys. Rev. B* **19** 384
- [19] Lawson A C 1970 *Phys. Lett.* **33A** 231
- [20] Hill H H 1970 *Plutonium and Other Actinides* ed W N Miner (New York: AIME) p 1
- [21] Luengo C A, Huber J G, McCallum R W and Maple M B 1975 *Proc. 14th Int. Conf. on Low Temperature Physics (Otaniemi, 1975)* vol 3, ed M Krusius and M Vuorio (Amsterdam: North-Holland) p 192
- [22] Kadowaki K and Woods S B 1986 *Solid State Commun.* **58** 507
- [23] Wilson K G 1975 *Rev. Mod. Phys.* **47** 773
- [24] Marmeggi J C, Lander G H, van Smaalen S, Brückel T and Zeyen C M E 1990 *Phys. Rev. B* **42** 9365
- [25] Faber J and Lander G H 1976 *Phys. Rev. B* **14** 1151