

RESISTIVITY OF SINGLE-CRYSTALLINE UPt_3 AND ITS PRESSURE DEPENDENCE; INTERPRETATION BY A SPIN-FLUCTUATION MODEL

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Resistivity measurements on a series of uranium–platinum compounds were carried out in the range 1.3 up to 300 K and pressures up to 5 kbar. The resistivity of single-crystalline UPt_3 is strongly anisotropic and points to pronounced spin-fluctuation effects that are partly suppressed at a pressure of 4 kbar. In paramagnetic UPt_2 and UPt_5 spin-fluctuation effects are less pronounced, whereas in ferromagnetic UPt the spin-disorder scatter is dominant above 27 K.

1. Introduction

Since recently superconductivity has been observed in UPt_3 [1] it becomes clear that this compound (hexagonal $MgCd_3$ -type of structure) belongs to a rapidly growing new class of materials which are indicated as “heavy fermion” superconductors. Other members of this class are $CeCu_2Si_2$ [2], UBe_{13} [3], U_6Fe [4] and U_6Co [5]. Unusual low-temperature properties are observed in UPt_3 : the susceptibility [6] is anisotropic and shows a low-temperature maximum along the a - and b -direction near 16 K; magnetization curves [6,7] at 4.2 and 1.5 K exhibit a strongly non-linear behaviour, whereas at 20 and 77 K a linear relationship between magnetic moment and applied field is observed; the specific heat [1,6] does not give any evidence for magnetic order, that was originally suggested for this compound, and shows a $T^3 \ln T$ contribution below 15 K, together with an unusually high value for the coefficient of the electronic term; superconductivity [1] has been observed below 0.54 K.

In this contribution we report on resistivity measurements along different crystallographic directions at zero pressure and applied pressures of 4–5 kbar in the temperature interval 1.5–300 K. For comparison the resistivity data of the remaining uranium–platinum compounds (orthorhombic UPt and UPt_2 , cubic UPt_5) are also presented.

2. Experimental

Single-crystalline UPt_3 was prepared in a tri-arc melting equipment by the Czochralski method [8], yielding a cylinder of approximately 6 mm diameter and 20 mm long. Cylindrical samples, with typical sizes of 1.5 mm diameter and a length of 6 mm, were machined out of the bulk along the different crystal axes by means of spark erosion. Measurements were performed before and after annealing at 900 °C during one day. Polycrystalline compounds (UPt , UPt_2 and UPt_5) were prepared by arc melting the appropriate amounts of constituents in a titanium gettered argon atmosphere. After casting of the melt into a water-cooled copper crucible, cylindrical samples were obtained, with a diameter of 1.5 mm and a length of 15 mm. Finally, all samples were annealed in evacuated sealed silica tubes at 900 °C for a period of 7 days.

The overall temperature picture and the pressure dependence of the electrical resistivity were obtained by a four-point low-frequency ac-method. Current and voltage contacts were realised by clamping the samples onto an anodized aluminium sample holder, using small pieces of razor blades, welded to copper springs. The sample holder could be placed in a pressure vessel, constructed out of a beryllium–copper alloy. Helium was used as hydrostatic pressure transmitting medium. Apart from some irreproducibilities, that occurred around

40 K at solidification of the helium at pressures of about 4 kbar, this method appeared to satisfy quite well. The resistivity values are accurate within 3 and 7% for poly- and single-crystalline samples, mainly due to geometrical uncertainties. Relative accuracies were about $0.1 \mu\Omega\text{cm}$. The residual resistivity and the temperature dependence of the resistivity at low temperature and zero pressure were determined in addition by a standard four-point dc-method.

3. Results

The results of the resistivity measurements with the ac-method at zero pressure are shown in fig. 1, as smooth curves through the data points (data on UPt_3 are taken on unannealed samples). All runs were taken with increasing and decreasing temperatures; no hysteresis effects were observed. The curves were slightly shifted in a vertical direction, to let the residual resistance, ρ_0 , coincide with the value obtained from the dc-method. The ρ_0 -values are 34, 11.2 and $25.7 \mu\Omega\text{cm}$ for polycrystalline UPt , UPt_2 and UPt_5 , respectively, and 7.2, 7.2 and 3.6 for the a -, b - and c -axis of single-crystalline UPt_3 , respectively.

In hexagonal UPt_3 a large anisotropy was observed between the basal plane and the hexago-

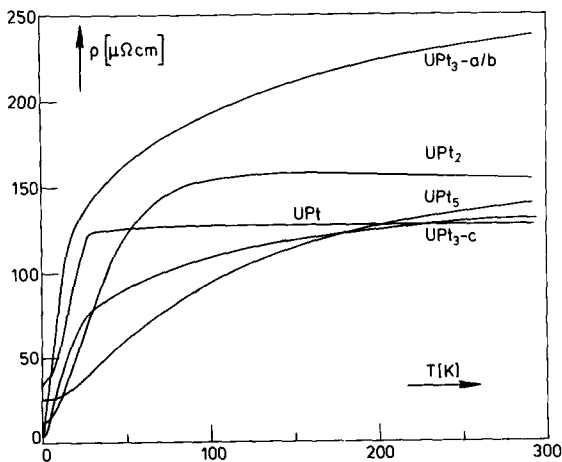


Fig. 1. Temperature dependence of the electrical resistivity at zero pressure. Data on single-crystalline UPt_3 are taken on unannealed samples.

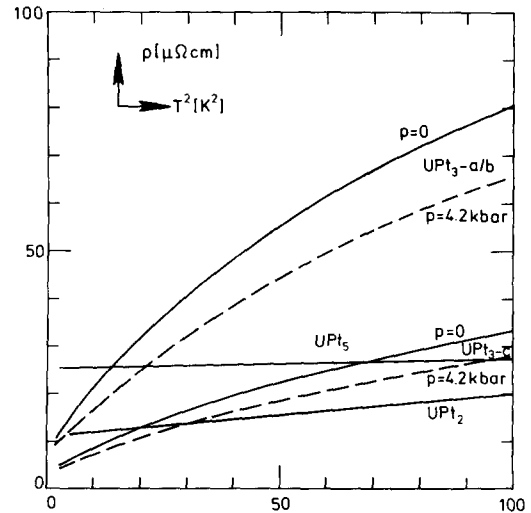


Fig. 2. Electrical resistivity versus T^2 at zero pressure (full line) and at a pressure of 4.2 kbar (dashed line).

nal axis. Data taken along the a - and b -axis show no significant difference within the experimental accuracy. Residual resistance ratio's (i.e. $\rho(300 \text{ K})/\rho_0$) were 33, 33 and 37 for the a -, b - and c -direction, before annealing; after annealing the ρ_0 -values decreased to 6.2, 3.0 and 1.7, leading to residual resistance ratio's of 38, 79 and 78, respectively. The differences in ρ_0 -values, possibly due to small deviations from stoichiometry, might be introduced by cutting the samples from different parts from the single-crystalline batch. The resistivity curve of a polycrystalline UPt_3 sample, prepared in the above described way, nearly coincides with the curves along the a - and b -axis, indicating that preferred orientations occur (the crystallites have the c -axis mainly perpendicular to the cylinder axis, as was confirmed by magnetization measurements on this sample).

The low-temperature results of the resistivity measurements at applied pressure of 4.2 kbar are given in figs. 2 and 3a for the compound UPt_3 in a plot of ρ versus T^2 .

4. Discussion

The resistivity data of single-crystalline UPt_3 and polycrystalline UPt_2 and UPt_5 display an

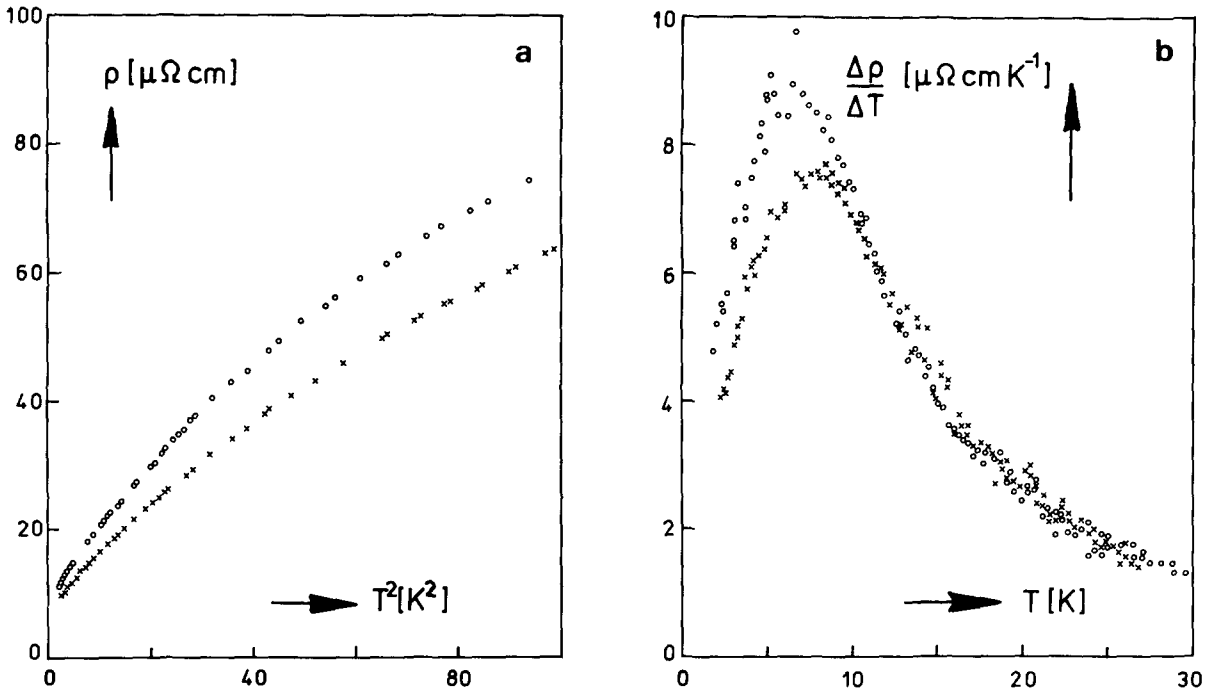


Fig. 3. Electrical resistivity versus T^2 (a) and differential resistivity versus T (b) at zero pressure (open circles) and at a pressure of 4.2 kbar (crosses) for the compound UPt_3 along the b -axis.

S-shape type of curve that is characteristic for systems in which spin-fluctuation phenomena occur [9]. A sharp rise at low temperatures is followed by a negative curvature towards the temperature axis (i.e. $\partial^2\rho/\partial T^2 < 0$) and a tendency to saturate at large values in the room-temperature region. The overall large electrical resistivities are ascribed to scattering of the conduction electrons by the spin-fluctuations of the $5f$ band electrons. The electron-electron and electron-phonon scattering terms in the resistivity are assumed to be negligible at the low-temperature side, whereas they play a minor role in the high-temperature region, as can be concluded by comparing previous measurements [10,11] on UPt and isostructural $ThPt$.

According to theory [12] the spin-fluctuation contribution to the resistivity can be expressed by an AT^n -term, with $n = 2$ at low temperatures and $n = 1$ at elevated temperatures. A plot of ρ versus T^2 (see fig. 2) clearly shows a value less than 2 for the exponent n in case of UPt_3 at temperatures

above 1.5 K. In a double logarithmic plot of the resistivity data (see fig. 4) we observe that below 1.5 K the exponent possibly approaches to a value of two for UPt_3 , yielding large values for the coefficient A : 1.6 and 0.7 $\mu\Omega\text{cm}/\text{K}^2$ for the basal plane and hexagonal axis, respectively. In the other non-magnetic compounds UPt_2 and UPt_5 , an $n = 2$ value holds over a considerably larger temperature interval; the coefficient A for these compounds takes values of 0.09 and 0.01 $\mu\Omega\text{cm}/\text{K}^2$, respectively.

The characteristic change to a negative curvature towards the temperature axis in the resistivity curve is clearly reflected by a peak in a plot of $d\rho/dT$ versus T (see fig. 5) and is most pronounced for the data along the a - and b -axis of UPt_3 . It is considered to reflect the spin-fluctuation effects most clearly. The sharp peak in $d\rho/dT$, or the inflection point in the ρ versus T curve, shifts from 6.5 K for UPt_3 to higher temperatures for UPt_2 (26 K) and UPt_5 (30 K), and becomes weaker in structure for the latter compounds.

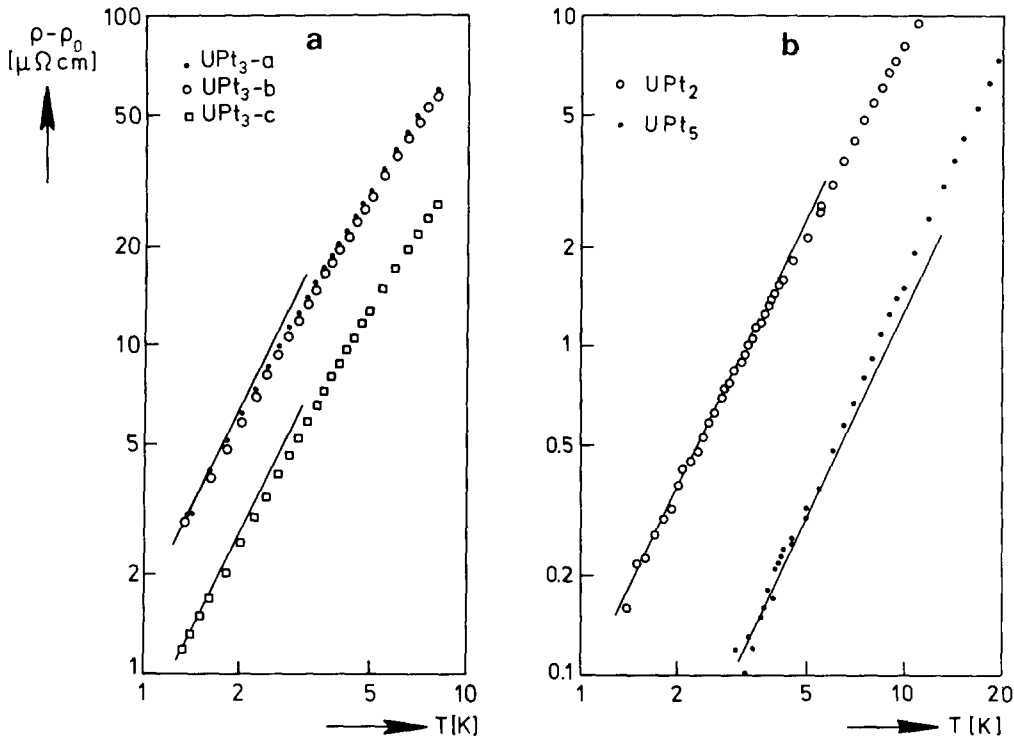


Fig. 4. Double logarithmic plot of $\rho - \rho_0$ versus T for annealed single-crystalline UPt_3 (a), and UPt_2 and UPt_5 (b). Note the differences in vertical scale. The full lines represent $n = 2$.

In the high-temperature region saturation at large values becomes noticeable; room temperature values are: 154 and 141 $\mu\Omega\text{cm}$ for UPt_2 and

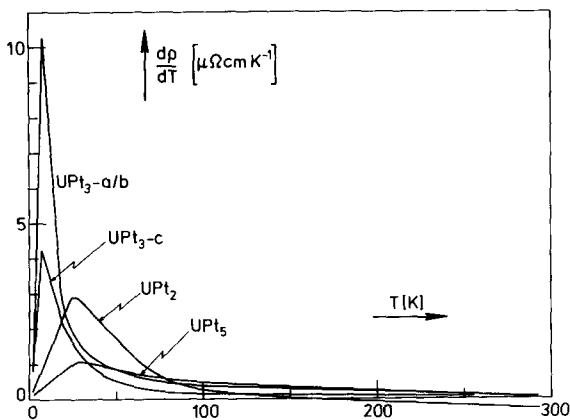


Fig. 5. Temperature dependence on the derivative of the electrical resistivity with respect to temperature.

UPt_5 , and 132 (hexagonal axis) and 238 $\mu\Omega\text{cm}$ (basal plane) for UPt_3 . In particular the latter value is unusually large, and consistent with a strong electron-spin-fluctuation scattering. In the uranium-platinum series only the compound UPt_2 displays a weak maximum, around 165 K, as follows from a negative $d\rho/dT$ in fig. 5.

Equiatomic UPt orders magnetically below 27 K. This compound reveals a complex magnetic behavior, reflected in two magnetic transitions at 27 and 19 K, leading to ferromagnetic arrangement of the magnetic moments below 19 K. The large resistivity values in the paramagnetic region are ascribed to spin-disorder scatter; the resistivity curve of UPt exhibits additional anomalies [13] around 19 K, that were not reported in previous papers [10,11]. A detailed analysis of the magnetic properties of this compound will be published elsewhere [13].

At applying hydrostatic pressures on the com-

pound UPt₃ the resistivity decreases, although the ρ_0 -values are pressure independent as follows from figs. 2 and 3a. At room temperature the resistivity decreases by a few percent only at applying a pressure of 5 kbar. Low-temperature changes in the resistivity with pressure are considerably larger. In a description of the resistivity data at the lowest temperatures by the expression $\rho = \rho_0 + AT^2$ a decrease of the coefficient A by 40% over 4.2 kbar is found. A plot of $\Delta\rho/\Delta T$ -values versus temperature for the basal plane (see fig. 3b) leads to a shift of the peak towards higher temperature (7.5 K); a considerable increase in $\partial^2\rho/\partial T^2$ near 16 K, coincident with a peak in the susceptibility, can also be observed. Above 45 K the pressure effect on the resistivity is temperature independent and amounts to a value of $-1.4 \mu\Omega\text{cm/kbar}$. Along the c -axis the pressure effects show the same features as those indicated by fig. 3 for the b -axis although less pronounced.

In summary we conclude that the resistivity curves of UPt₂ and UPt₃ vary initially according to a T^2 -law, consistent with a simple model in which the conduction electrons are scattered by the spin fluctuations of the 5f band electrons via the exchange interaction. The temperature range, however, in which this T^2 -dependence is observed is very limited, indicating that other mechanisms, involving a more complex theory, play an important role. The maximum in UPt₂ might be due to de-hybridization of the s - f band; scattering becomes less effective at high temperatures leading to a decrease in the resistivity. Although for UPt₃ a T^2 -term can be observed only at approaching 1.3 K, we believe, taking into account the overall resistivity behaviour, that these data are indicative of pronounced spin-fluctuation effects, in consistence with specific heat, susceptibility and magnetization data. Resistivity is strongly anisotropic as are the magnetic parameters. By applying hydrostatic pressures between 4 to 5 kbar the spin-fluctuation effects in the resistivity are partly suppressed in the low-temperature region.

Note added in proof

Our Czochralski-grown single-crystalline UPt₃ samples along the a -, b - and c -axis reveal a superconducting transition temperature of 0.48 K. For details see: A. de Visser et al., in: Proc. 4th General Conf. of the Condensed Matter Division of the European Physical Society, 19–23 March 1984, Den Haag, The Netherlands (to be published in Physica B).

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