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The heavy-fermion superconductor UPd_2Al_3 at very high pressure

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Abstract. We studied the pressure dependence of the Néel temperature and the superconducting critical temperature of the heavy-fermion superconductor UPd_2Al_3 at pressures up to 13.6 GPa by means of resistivity measurements. The pressure variation of the normal state resistivity is dominated by a large shift of the resistivity maximum to higher temperatures. The Néel temperature decreases with increasing pressure, while the onset of the superconducting state is almost invariant with pressure up to 6.5 GPa. At higher pressures a decrease of T_c has been observed.

1. Introduction

The heavy-fermion superconductor (HFS) UPd_2Al_3 has been the subject of several investigations since its discovery in 1989 [1]. UPd_2Al_3 combines a rather high ordered moment of $0.85\mu_B$ [2] with a superconducting critical temperature of 1.96 K [1], the highest known today among the HFSs. Since the physical properties of the HFSs are found to be very sensitive to changes of the lattice volume, the study of pressure effects may help in the understanding of these compounds. Previous resistivity measurements under pressures below 1 GPa [3–5] have shown that T_c is insensitive to the application of pressure in the low-pressure limit. The findings for the pressure variation of the Néel temperature are more puzzling as values for dT_N/dp from 1 K GPa^{-1} to -1 K GPa^{-1} have been observed [3–5]. Sato *et al* [6] reported a decreasing Néel temperature with a value of 8 K at 8 GPa for a single crystal and $I \perp c$. These results compare to those of thermal expansion experiments [7, 8]. Using Ehrenfest's relation the pressure dependences of T_N and T_c were estimated, giving a value of ~ -0.9 K GPa^{-1} for dT_N/dp and indicating that T_c should be insensitive to pressure. Since up to now no complete and conclusive knowledge of the high-pressure behaviour of UPd_2Al_3 existed, we performed high-pressure resistivity measurements in the temperature range of 1.2–300 K.

2. Experimental details

For the high-pressure experiments we used a 'clamp' device based on the Bridgman technique, with anvils made of sintered diamond, which have flats of 2.2 mm in diameter. Pyrophyllite served as gasket material and steatite as pressure transmitting medium. The sample was prepared by arc melting the pure elements and subsequent annealing at 900 °C for 120 h. For use in the high-pressure cell we cut our sample with the dimensions

$40 \times 100 \times 600\ \mu m^3$ from a larger polycrystal referred to as sample PK4 in [9] and [10]. The pressure was estimated by the T_c of a thin Pb foil next to the sample. The conversion from T_c to pressure values was carried out using the conversion table of Bireckoven and Wittig [11].

3. Results

An overview of our high-pressure resistivity data is given in figure 1. At low pressure we found the characteristic features of the resistivity curve as reported for a polycrystalline sample of the same batch at ambient pressure [4], i.e. a pronounced maximum in $\rho(T)$ near 90 K. As shown in the inset of figure 1, T_{max} increases exponentially with pressure with a slope of $\mathrm{d}\ln T_{max}/\mathrm{d}p \approx 0.06\ \mathrm{GPa}^{-1}$. The ambient temperature resistivity increases from 230 $\mu\Omega$ cm to 280 $\mu\Omega$ cm at 13.6 GPa.

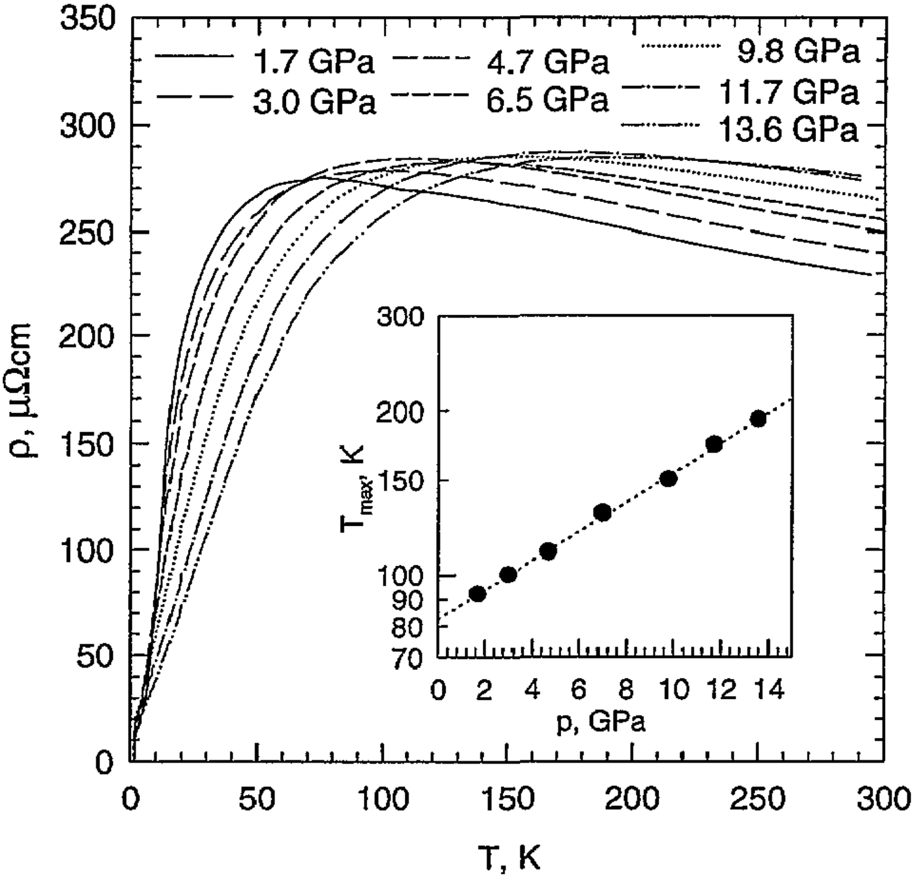

Figure 1. The electrical resistivity of polycrystalline UPd_2Al_3 at pressures up to 13.6 GPa. The inset shows the pressure variation of the temperature of the resistivity maximum.

At low temperatures we focus our interest on the two phase transitions in UPd_2Al_3, the transition to an antiferromagnetic phase with a Néel temperature of 14.5 K at ambient pressure and the transition to the superconducting state at 2 K. For the latter figure 2 shows the various transitions we observed at high pressures, normalized to the normal state residual resistivity of 20 $\mu\Omega$ cm, which did not vary significantly under pressure. Clearly we observe a sharp onset of the transition, which is almost independent of pressure up to 6.5 GPa. For pressures above 6.5 GPa the temperature of the onset decreases with pressure. The transitions at low pressure were sharp, but below 10% of the normal state resistivity we observed a tail in the transition. The origin of this behaviour, which was not reported at ambient pressure, may be attributed to a small impurity phase, which became noticeable due to the preparation procedure for the thin sample. Further investigations with other samples may clarify this point. A fit of the resistivity from T_c to 3.5 K results in a T^2 dependence with A coefficients decreasing from 0.7 $\mu\Omega$ cm K^{-2} at 1.7 GPa to 0.4 $\mu\Omega$ cm K^{-2} at 6.5 GPa. Above 3.5 K the resistivity deviates from simple power law behaviour.

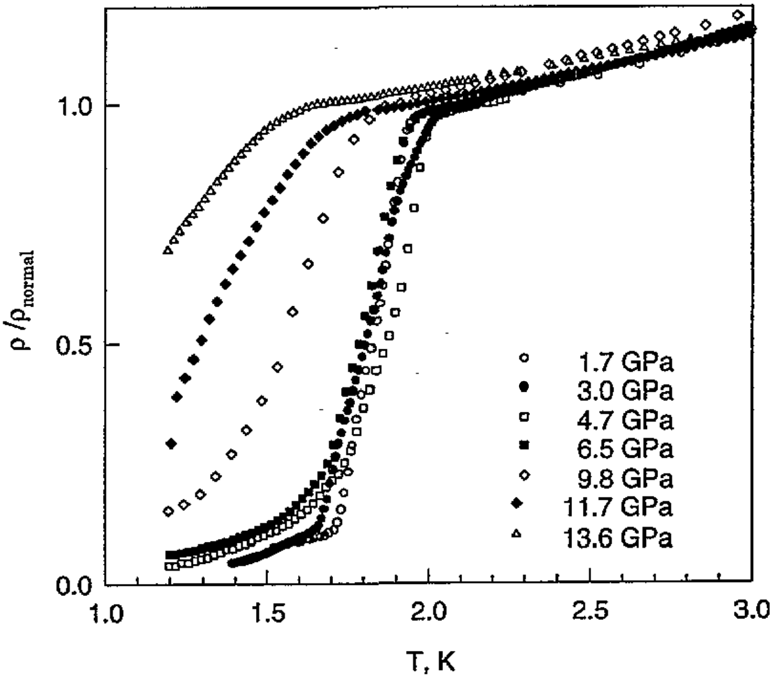

Figure 2. Transitions to the superconducting state of UPd_2Al_3 at elevated pressures. The resistivity data have been renormalized to the resistivity of the non-superconducting state for clarity.

For pressures up to 6.5 GPa we could estimate the Néel temperature as the cusp of the temperature derivative of the resistivity $d\rho(T)/dT$ (figure 3). Above 6.5 GPa the resistivity increases linearly from T_c up to 20 K, and a Néel temperature could not be estimated from the data (see the 9.8 GPa data in figure 3). Figure 4 shows the pressure variation of the Néel temperature and of T_c^{onset} for the whole pressure range up to 13.6 GPa.

4. Discussion

The interpretation of our data will concentrate on three main features: the temperature

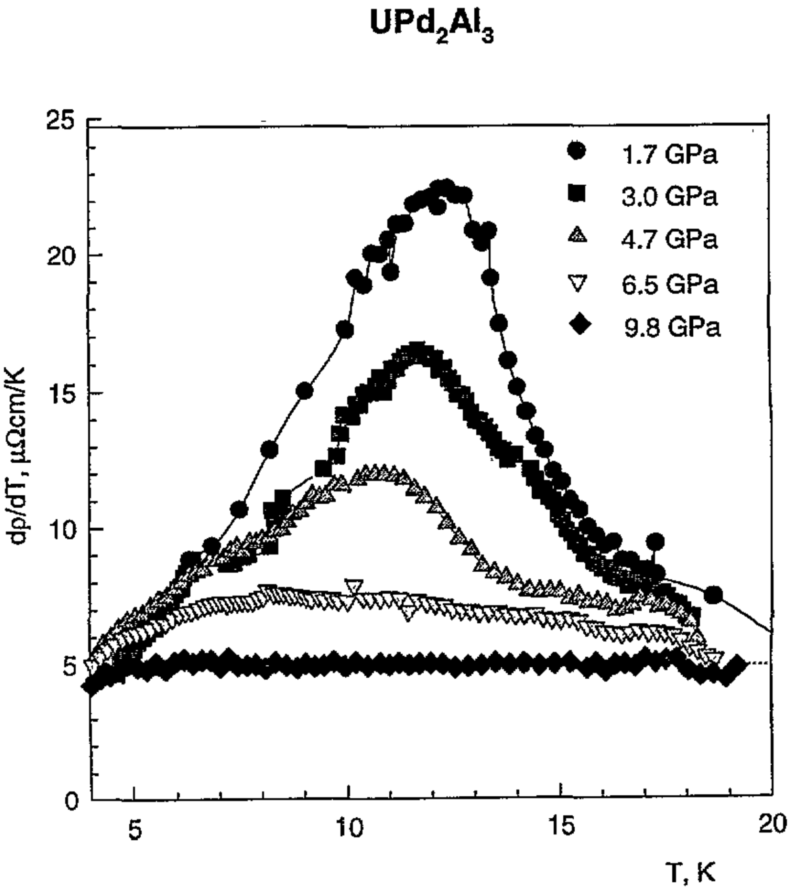

Figure 3. The determination of the Néel temperature from $d\rho/dT$ data at pressures up to 6.5 GPa. Note the absence of a significant maximum for the 9.8 GPa data.

increase of T_{max}, as the dominant effect of high pressure on the normal state resistivity, and the variation of T_N and of T_c^{onset} with pressure.

From the inset in figure 1 we can see the linear pressure dependence of T_{max} on a logarithmic temperature scale, yielding a value of 0.06 GPa^{-1} for $d\ln T_{max}/dp$, valid for the whole investigated pressure range. Such an increase of T_{max} with pressure is very common in the heavy-fermion compounds and is usually interpreted by an increase of T_K, the characteristic temperature of the Kondo scattering. With a compressibility of ~ 0.85 Mbar^{-1} obtained from ultrasound experiments [12] we estimate a Grüneisen parameter $\Omega \approx 7$.

Several investigations of polycrystalline and single-crystalline UPd$_2$Al$_3$ at low pressure revealed variations of dT_N/dp from 1 K GPa^{-1} [10] observed for a single crystal with $I \perp c$ to -0.9 K GPa^{-1} [10] for a single crystal with $I \parallel c$. Sato *et al* [6] observed for single-crystalline UPd$_2$Al$_3$ -0.6 K GPa^{-1} at pressures up to 8 GPa with $I \perp c$. Our value for dT_N/dp of -0.9 K GPa^{-1} appears to be in agreement with the values for single-crystal samples with $I \parallel c$ and the value of Sato *et al* for higher pressures. At low pressure for the same sample from the same batch a value for dT_N/dp of only -0.1 K GPa^{-1} had been found. Modler *et al* [8] estimated from the thermal expansion data using the Ehrenfest relation a value of dT_N/dp in the low-pressure limit for hydrostatic conditions of -0.9 K GPa^{-1}, i.e.

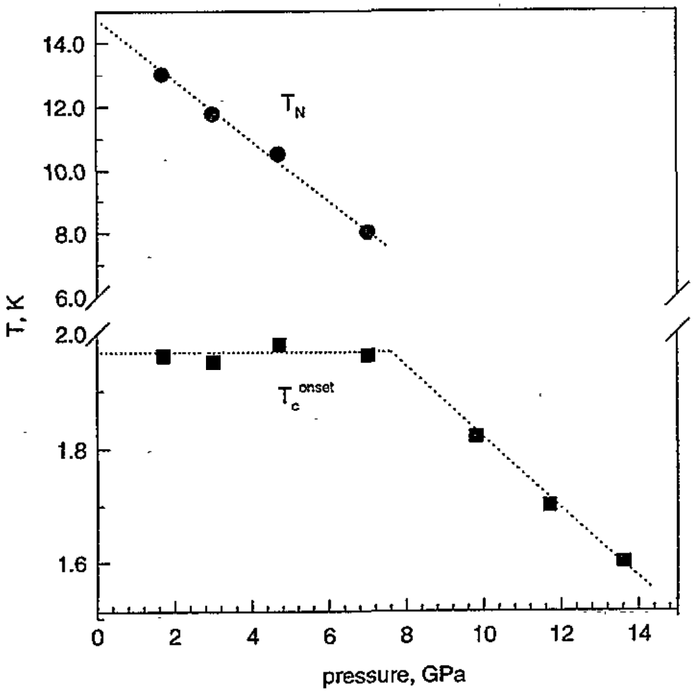

Figure 4. The pressure dependence of the Néel temperature and the onset of the superconducting transitions. Dotted lines are guides for the eye.

the value we found for our sample. They also found an enormous anisotropy of dT_N/dp for uniaxial stress. Thus one would tentatively ascribe the differences found in dT_N/dp to be due to non-hydrostatic conditions. Slight differences in the composition of the samples may also lead to different initial pressure dependences. One should further note that the method used to define T_N, i.e. the cusp in $d\rho/dT$ or a maximum in $d^2\rho/dT^2$, may influence the observed initial values. Above 6.5 GPa we could not estimate T_N, because the maximum in $d\rho/dT$ disappeared. The absence of the signature of T_N above 6.5 GPa may have two origins: (i) T_N decreases further with the same rate, but the magnetic moment has become so small that in resistivity data the anomaly is washed out or (ii) the antiferromagnetic ordering is suppressed at these high pressures. Measurements of the magnetic properties under high pressure are necessary to settle this question finally.

Modler *et al* [8] also suggested from their thermal expansion data that T_c should be very insensitive to hydrostatic pressure and indeed our observation of $dT_c/dp \approx 0$ up to 6.5 GPa confirms this estimation. However, in figure 4, which shows the pressure dependences of T_N and T_c^{onset}, a remarkable effect is visible. Just at about 6.5 GPa, where T_N becomes undetectable, T_c^{onset} starts to decrease with increasing pressure. Above 6.5 GPa we estimate dT_c/dp to be -0.05 K GPa^{-1}. Also the transitions become broader, which may reflect the pressure gradient in the pressure cell. A comparison to other U-based HFSs reveals that the T_c values of all other U-based HFSs decrease with increasing pressure. The exceptional behaviour of UPd_2Al_3 is a further hint that this compound is, at ambient pressure, farther away from the critical point corresponding to the transition from the magnetic to a non-magnetic regime. Therefore pressure initially does not affect the superconducting properties. At higher pressure, once the magnetic moment has been sufficiently quenched, superconductivity also begins to be influenced and T_c decreases.

5. Conclusion

The resistivity measurements under pressure presented here allow us to suggest a temperature–pressure phase diagram of UPd_2Al_3 up to 13.6 GPa as presented in figure 4. Around 6.5 GPa T_N became undetectable for our method. The question of whether it drastically decreases to zero or is just washed out remains open and might be a subject for future experiments. Our measurements confirm the pressure dependences for T_N and T_c for the low-pressure limit as suggested from thermal expansion experiments. At high pressures the behaviour of UPd_2Al_3 changes and shows a decreasing T_c as observed already for other HFSs.

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