

HIGH-PRESSURE MAGNETISATION STUDIES OF SOME HEAVY-FERMION URANIUM INTERMETALLICS

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High-pressure magnetisation measurements have been performed on single-crystalline samples of hexagonal UPt_3 along different crystallographic axes at temperatures between 4.2 and 40 K, at pressures up to 4.6 kbar and magnetic fields up to 6.4 T and on tetragonal URu_2Si_2 along the tetragonal axis in the same temperature, pressure and field regions.

1. Introduction

Among the heavy-fermion systems that have been discovered in the past few years, the intermetallic compound UPt_3 (hexagonal MgCd_3 -type of structure) is of special interest because of the coexistence of spin-fluctuation phenomena (indicated by a pronounced $T^3 \ln(T/T^*)$ term in the specific heat) and superconductivity [1, 2, 3]. High-pressure data turn out to be helpful for checking theories that have been developed for describing the normal and superconducting states with the same set of microscopic parameters. Up till now the high-pressure magnetisation studies on UPt_3 have been performed on polycrystalline samples [4, 5]. Due to the large magnetic anisotropy present in this compound, information on single-crystalline material is highly desirable. This information is presented in the present paper. The results of these high-pressure magnetisation measurements will be compared with high-pressure resistivity data, with forced magnetostriction measurements and with a combined analysis of specific heat and thermal expansion data.

A second interesting system, in which magnetism and superconductivity coexist, is the tetragonal compound URu_2Si_2 (ThCr_2Si_2 -type of structure). The compound exhibits a magnetic transition at 17.5 K and superconductivity below 1 K [6, 7, 8]. Previous high-pressure data include the pressure dependence of the magnetic [7, 9] and superconducting [7] transition temperatures

which were found to have different signs. In the present paper we present a further analysis of our high-pressure resistivity data for a monocrystalline sample [9]. In addition recent results of high-pressure magnetisation studies are reported.

2. Experimental methods

Magnetisation measurements were carried out on single-crystalline UPt_3 samples with the magnetic field applied parallel and perpendicular to the hexagonal axis and on a URu_2Si_2 sample with the field along the tetragonal axis only. The mass of the samples amounted to 4.4 (*b*-axis) and 3.9 g (*c*-axis) in the case of UPt_3 , and to 0.9 g for the URu_2Si_2 sample. Samples were prepared by spark erosion from pieces of Czochralski-grown bulk material [10].

In order to measure the magnetisation under pressure, the samples (with sizes limited to 6 mm diameter and 10 mm length) were put in a beryllium-copper pressure cell. To avoid paramagnetic contributions to the measured signal at low temperatures arising from the pressure cell, the construction material of the pressure cell was chosen without cobalt additions. Maximum pressures in this case were limited to 6 kbar. Helium served as the pressure transmitting medium. Special care was taken to freeze the helium gas under constant pressure in order to keep hydrostatic conditions in the solid helium as

good as possible. Pressures in the solid helium region were determined by strain gauges on the pressure cell.

The equipment for magnetisation measurements consists of a set of two oppositely wound pick-up coils in a superconducting solenoid. The high-pressure cell containing the sample is moved up and down between the centres of the pick-up coils. By integrating the induced voltage, the magnetic moment of the sample is determined. The effect of pressure on the resistivity was studied by a four-terminal method in the same way as reported before [11].

3. Experimental results

In fig. 1 we present the magnetisation curves for UPt_3 with the field applied parallel and perpendicular to the hexagonal axis. From the data in this figure we calculate a reduction of the basal plane susceptibility from $(112.8 \pm 1.0) \times 10^{-9}$ to $(98.2 \pm 1.0) \times 10^{-9} \text{ m}^3/\text{molU}$ by applying a pressure of 4.6 kbar (1 molU denotes 1 mole uranium atoms, which is equivalent to 1 mole

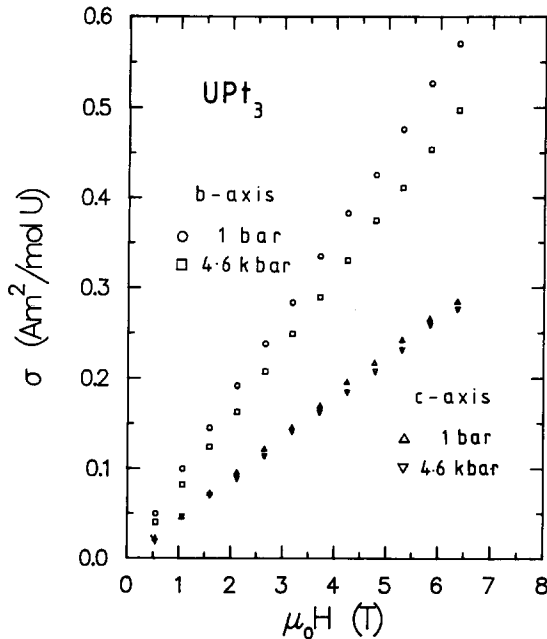


Fig. 1. Magnetisation of single-crystalline UPt_3 at 4.2 K, for field directions and pressures as indicated.

formula units). This reduction is much smaller for the hexagonal axis: χ equals $(57.2 \pm 0.5) \times 10^{-9} \text{ m}^3/\text{molU}$ at 1 bar and $(55.9 \pm 0.5) \times 10^{-9} \text{ m}^3/\text{molU}$ at 4.6 kbar. In figs. 2a and b we

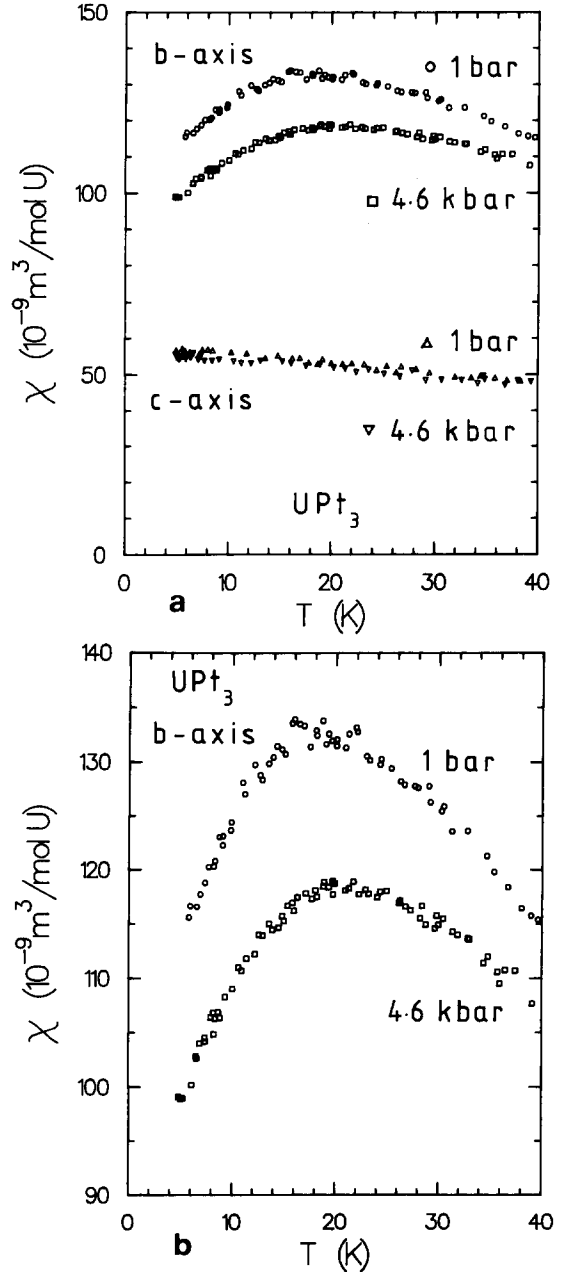


Fig. 2. (a) Susceptibility of single-crystalline UPt_3 for field directions ($B = 5 \text{ T}$) and pressures as indicated; (b) idem b -axis.

show the temperature dependence of the susceptibility for temperatures up to 40 K. The maximum in the susceptibility curve along the b -axis that occurs at 17.9 K at 1 bar shifts under applied pressures of 4.6 kbar with (2.5 ± 0.5) K to higher temperatures. The susceptibility along the c -axis is hardly affected by applying pressures up to 4.6 kbar as follows from the data of fig. 2a.

Pressure results for the magnetisation data of URu_2Si_2 along the tetragonal axis are presented in fig. 3. Applying a pressure of 4.6 kbar the susceptibility reduces from the 1 bar value of $(70.0 \pm 1.0) \times 10^{-9}$ to a value of $(60.8 \pm 1.0) \times 10^{-9} \text{ m}^3/\text{molU}$. In addition the temperature dependence of the magnetisation of URu_2Si_2 along the tetragonal axis in a field of 5.01 T has been measured (not shown). At 17.6 K we observe an anomaly in the magnetisation curve indicating the Néel temperature that has been determined on a different sample of the same melt in resistivity measurements to be equal to 17.4 K [9]. Apparently the Néel temperature is shifted under pressure to higher temperatures. This

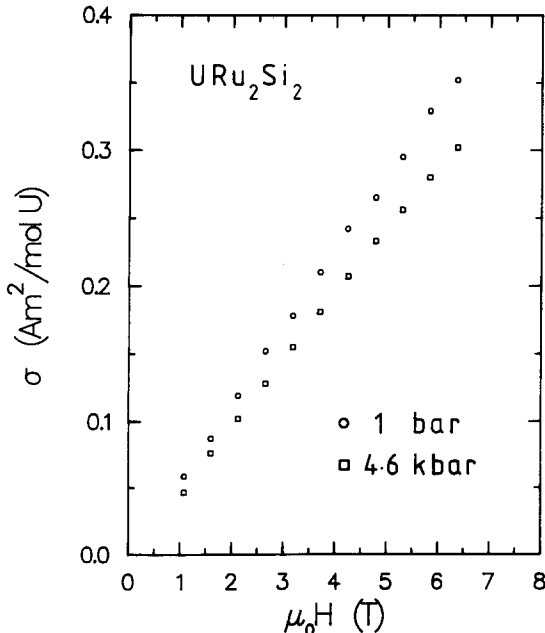


Fig. 3. Magnetisation of single-crystalline URu_2Si_2 at 4.2 K, for a field along the c -axis at pressures as indicated.

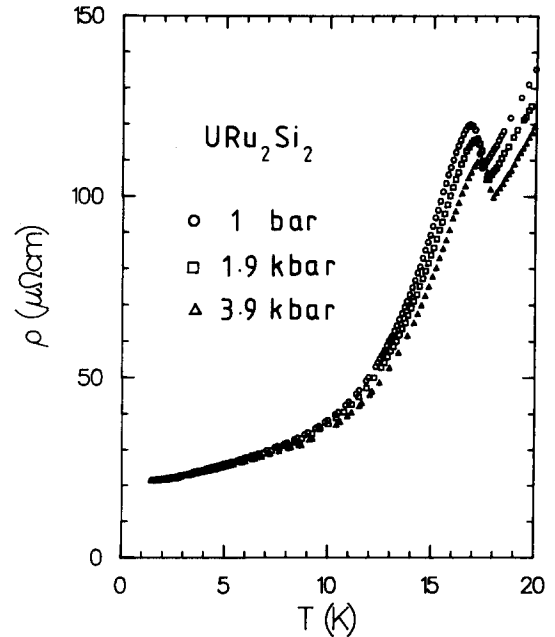


Fig. 4. Electrical resistivity of single-crystalline URu_2Si_2 with the current along the a -axis and pressures as indicated.

pressure-induced shift cannot be determined from these magnetisation data with the same accuracy that we reached in the resistivity measurements in which the ordering temperature manifests itself by a sharp minimum (see fig. 4). A value of (130 ± 40) mK/kbar follows for the pressure dependence of the ordering temperature from the magnetisation data, compared to a value of (118 ± 2) mK/kbar from the resistivity measurements. Experiments on the susceptibility of URu_2Si_2 with the field applied perpendicular to the tetragonal axis are in progress.

4. Discussion

Experiments on the pressure dependence of the susceptibility of polycrystalline UPt_3 at 4.2 K have previously been reported in refs. 4 and 5. The resulting value for the relative pressure dependence of the susceptibility amounts to -25 Mbar^{-1} , quite close to the value of $-28 \pm 3 \text{ Mbar}^{-1}$ that we deduce from the data of fig. 1

for the b -axis. For the c -axis a value of $-5 \pm 3 \text{ Mbar}^{-1}$ results. Note that throughout this paper we approximate the relative pressure dependence of a quantity A , $\partial \ln A / \partial P$, by $(A_0)^{-1} \Delta A / \Delta P$, where A_0 represents the 1 bar value of A . In analysing the pressure results for the polycrystalline sample it was already realised that due to preferential orientations the pressure effect is largely governed by the susceptibility of the basal plane of UPt_3 . This conclusion was further supported by the results of forced magnetostriction measurements along different crystallographic axes and for different directions of the applied field with respect to the crystallographic axes [12]. By applying thermodynamic relations the relative pressure dependence of the susceptibility in the basal plane was predicted to be equal to -23 Mbar^{-1} for fields up to 3 T along the a -axis and equal to -3 Mbar^{-1} for a field along the c -axis; both results hold for T equal to 4.2 K. In previous publications the temperature, T_m , at which the maximum in the χ vs. T curve occurs has been considered as indicative for the spin-fluctuation temperature T^* . Furthermore it was noticed that the product of the susceptibility value at 4.2 K and the temperature T_m is almost pressure-independent ($\chi T_m \approx \chi T^* = \text{const.}$). In this way various results for $\partial \ln T^* / \partial P$ were obtained. In addition this pressure derivative was also determined from the coefficient of the T^2 -term in the resistivity at low temperatures ($\rho = AT^2$ with $A \propto (T^*)^{-2}$). The present values for $\partial \ln T^* / \partial P$ as determined from the pressure dependence of the susceptibility at 4.2 K and from the pressure-induced shift of T_m are equal to (28 ± 3) and $(30 \pm 6) \text{ Mbar}^{-1}$, respectively, and fall inside the error limits that have previously been derived on the basis of susceptibility measurements on polycrystalline UPt_3 samples and of resistivity measurements on single-crystalline material [4, 5, 11, 13].

The relative pressure dependence of χ and T_m can be transformed into $\partial \ln \chi(4.2 \text{ K}) / \partial \ln V$ and $\partial \ln T_m / \partial \ln V$ values by using the compressibility value, $\kappa = 0.479 \text{ Mbar}^{-1}$, derived from sound velocity measurements performed by de Visser et al. at 300 K [14]. This value agrees nicely with the value 0.477 Mbar^{-1} , that can be derived from

sound velocity measurements of Yoshizawa et al. [15], taking one of the elastic constants, c_{13} (not measured by Yoshizawa et al.), from ref. 14. Under the assumption that c_{13} is (nearly) temperature independent the data of Yoshizawa provide a compressibility of 0.476 Mbar^{-1} at 4.2 K. The resulting values for $\partial \ln \chi(4.2 \text{ K}) / \partial \ln V$ and $\partial \ln T_m / \partial \ln V$ are (58 ± 6) and $-(63 \pm 13)$, respectively. The first value agrees quite closely with the value for the Grüneisen parameter, $\Gamma = 60$, as derived from a combination of specific heat and thermal expansion data on UPt_3 [15]. Since the crystal structure of UPt_3 is hexagonal we should in fact not expect an isotropic value for the compressibility. Following the analysis of the sound velocity measurements on UPt_3 at room temperature we deduce values for the linear compressibility, κ_a and κ_c , along the a -axis and c -axis, of 0.1641 and 0.1508 Mbar^{-1} , respectively. From these values we calculate a relative increase in c/a ratio with pressure, $\partial \ln(c/a) / \partial P \approx \kappa_a - \kappa_c = 0.013 \text{ Mbar}^{-1}$. The 4.2 K-value for this quantity is found to be equal to 0.012 Mbar^{-1} . Although there is no direct evidence that a change in the c/a ratio of UPt_3 plays a dominant role in the pressure-induced shifts in the superconducting and spin-fluctuation temperatures, there are indications that at substituting in UPt_3 platinum by palladium [16, 17], or uranium by thorium [17, 18], it is mainly the decrease in the c/a ratio which causes new ordering phenomena (Ramirez et al. in ref. 18). According to the c/a value for the pseudobinary (U, Th) Pt_3 and U(Pt, Pd) $_3$ compounds and using the information that is provided by the anisotropic compressibility data we expect a full suppression of these ordering phenomena in the 5% Pd and 5% Th samples at a pressure of roughly 30 kbar. At this pressure the decrease in the c/a ratio on alloying with respect to pure UPt_3 is compensated by the increase in the c/a ratio with pressure. Experiments to verify these hypotheses are currently in progress.

The relative pressure dependence of the susceptibility of URu_2Si_2 along the tetragonal axis of $(-29 \pm 5) \text{ Mbar}^{-1}$ is almost equal to the corresponding value for UPt_3 in the basal plane. Above the ordering temperature of 17.5 K this

relative pressure derivative is slightly reduced and amounts to $-(21 \pm 5) \text{ Mbar}^{-1}$ between 20 and 30 K. The relative pressure derivative of the magnetic ordering temperature of 7 Mbar^{-1} is different in magnitude compared to the spin-fluctuation temperature in UPt_3 . Its sign is the same as observed for some antiferromagnetic heavy-fermion compounds, like U_2Zn_{17} and UCd_{11} , with $\partial \ln T_N / \partial P$ -values of 1.8 and 14 Mbar^{-1} , respectively [19]. The anomalies in the specific heat [6, 7, 8] and resistivity of URu_2Si_2 around 17.5 K [7, 8, 9, 20] are suggestive for the formation of an energy gap at 17.5 K by which part of the energy states at the Fermi level are removed. This suggestion has been put forward by Maple et al. [8] in analysing the specific heat and has been adopted by Palstra et al. [20] in discussing the temperature dependence of the resistivity of single-crystalline URu_2Si_2 below 17 K.

The following expression has been used in the latter case:

$$\rho(T) = \rho_0 + BT(1 + 2T/\Delta) \exp(-\Delta/T) + AT^2, \quad (1)$$

where Δ represents the energy gap and where the quadratic term is added in order to account for the electron-electron interactions. We performed fits to the experimental data using eq. (1) in the temperature intervals 1.4–15 K and 5–15 K, in both cases taking a fixed value for parameter B . Resulting values for the parameters ρ_0 , Δ and A at different pressures are collected in table I. According to these data we derive values for the relative pressure derivatives of the parameters ρ_0 , Δ and A of (4 ± 2) , (11 ± 1) and $(-48 \pm 6) \text{ Mbar}^{-1}$, respectively. A remarkable feature of the high-pressure results is that $\partial \ln \chi^2(4.2 \text{ K}) / \partial P$ and $\partial \ln A / \partial P$ are within the error limits found to be equal. The same result is obtained for the corresponding parameters of UPt_3 , where the susceptibility result in this latter case refers to the basal plane. Finally we remark that the relative pressure derivatives of Δ and T_N are of the same order of magnitude.

In summary we have performed high-pressure

Table I

ρ_0 , Δ and A -values obtained from a 3-parameter fit to the electrical resistivity of URu_2Si_2 , using eq. (1), fixing B at $477 \mu\Omega\text{cm/K}$, at pressures and temperature intervals as indicated

Interval	Pressure	ρ_0 ($\mu\Omega\text{cm}$)	Δ (K)	A ($\mu\Omega\text{cmK}^{-2}$)
1.4–15 K	1 bar	21.5	84.6	0.16
1.4–15 K	3.9 kbar	21.7	88.3	0.13
5.0–15 K	1 bar	22.1	84.1	0.15
5.0–15 K	1.9 kbar	22.2	86.0	0.14
5.0–15 K	3.9 kbar	22.4	87.7	0.12
*1.4–17 K	1 bar	33.0	90.0	0.17

*From ref. 20, with $B = 800 \mu\Omega\text{cm/K}$.

magnetisation and resistivity measurements on single-crystalline samples of the heavy-fermion compounds UPt_3 and URu_2Si_2 . The anisotropy in the pressure effects in the case of UPt_3 , as expected from previous forced magnetostriction measurements, is confirmed. The relative pressure dependence of the characteristic spin-fluctuation temperature, as derived from the basal plane susceptibility data, amounts to $28 \pm 3 \text{ Mbar}^{-1}$. A further analysis of the resistivity data under pressure in the case of URu_2Si_2 results in a modest increase in the value for the energy gap near 17 K: Δ equals 84 K at 1 bar and 88 K at 4.6 kbar.

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