

## HIGH-MAGNETIC-FIELD AND HIGH-PRESSURE EFFECTS IN MONOCRYSTALLINE URu<sub>2</sub>Si<sub>2</sub>

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High-field magnetisation and high-pressure resistivity measurements have been performed on monocrystalline samples of the superconducting compound URu<sub>2</sub>Si<sub>2</sub> just above  $T_c$ . At 1.4 K, well below the magnetic ordering temperature of about 17.5 K, two sharp transitions at 36 and 40 T have been observed in the magnetisation versus field curves along the tetragonal  $c$ -axis, whereas the  $a$ -axis magnetisation curve remains linear up to 40 T. These transitions point to a complex magnetic behaviour. Above the second transition the magnetic moment per uranium atom exceeds a value of  $1.3 \mu_B$ . The resistivity versus temperature curve is distinguished by two characteristic temperatures of 75 and 17.4 K, corresponding with the onset of coherent scattering and the magnetic transition, respectively. Both characteristic temperatures increase with pressure.

### 1. Introduction

Among the heavy-fermion superconducting compounds with uranium, two typical compounds can be discerned: the Kondo-lattice compound UBe<sub>13</sub> and the spin-fluctuation compound UPt<sub>3</sub>. The characteristic differences between these two compounds are clearly demonstrated by the temperature dependence of the resistivity. In UBe<sub>13</sub> a low-temperature maximum is observed around 2 K with a flat region near 10 K and a negative temperature coefficient at higher temperatures. Below 2 K there is a steep decrease in resistivity before the start of superconductivity around 0.8 K [1]. In UPt<sub>3</sub> the resistivity in the normal state begins with a quadratic temperature dependence and develops a maximum in the temperature derivative around 7 K. The resistivity slowly saturates at temperatures well above 100 K [2]. The maximum in  $\rho(T)$  around 2 K for UBe<sub>13</sub> is considered as the onset of coherent scattering of the conduction electrons. For UPt<sub>3</sub> the maximum in the temperature

derivative of the resistivity at 7 K reflects the low temperature anomalies that have been observed in several other physical properties below 30 K, among them the thermoelectric power [3]. These phenomena can be considered to be indicative for spin-fluctuations.

Recently, it was established by Schlabitz et al. [4] and by Palstra et al. [5] that the ternary tetragonal compound URu<sub>2</sub>Si<sub>2</sub> exhibits the fascinating combination of magnetic order and superconductivity. Large anisotropies were found in both the magnetic and superconducting properties [5], similar to UPt<sub>3</sub> [6]. The resistivity is also anisotropic with a room temperature value along the  $a$ -axis about twice as large as along the  $c$ -axis [7]. The resistivity versus temperature curve for URu<sub>2</sub>Si<sub>2</sub>, however, is in some sense intermediate between the curves for UBe<sub>13</sub> and UPt<sub>3</sub>, discussed above. Starting at room temperature the resistivity in the tetragonal plane slowly increases with decreasing temperature and develops a broad maximum around 75 K. At lower temperatures a pronounced anomaly develops near

17.5 K, at the same temperature where the specific heat shows a sharp peak. Concerning the origin of this anomaly, various interpretations have been suggested: an itinerant type of antiferromagnetic order [4, 5] or the opening of an energy gap over a portion of the Fermi surface, associated with a spin or charge density wave [8].

## 2. Experimental methods

In this contribution we report high-field magnetisation studies along different crystallographic directions at 1.4 and 4.2 K on annealed monocrystalline as well as on polycrystalline samples. In addition, the pressure dependence for the resistivity versus temperature curve is presented for a monocrystalline sample with the current along the *a*-axis. The aim of the present investigation is to further elucidate the nature of the transition at 17.5 K. According to our experience with spin- and valence-fluctuation compounds [9, 10] we expect the anomalies in the susceptibility versus temperature curves to be reflected in the low-temperature high-field magnetisation curves. For example, the maximum in the dc susceptibility for the hexagonal plane of UPt<sub>3</sub> at 16 K is related to a maximum in the differential susceptibility at 1.5 K around 20 T [11]. Similar relationships have been found for other compounds like UA1<sub>2</sub>, TiBe<sub>2</sub>, UCo<sub>2</sub>, YbCuAl, etc. [9, 10].

High-pressure studies shed light on the physics behind a characteristic temperature in resistivity, susceptibility, specific heat, etc. In the case that the characteristic temperature announces the start of magnetic order, frequently a negative pressure effect on the characteristic temperature is observed. For itinerant systems this is rather the rule than the exception. But also for a system of local magnetic moments that interact by means of polarised conduction electrons, this negative pressure effect on the magnetic ordering temperature can be expected. Positive pressure effects on the characteristic temperature in metallic magnetism occur where spin fluctuations or Kondo scattering are involved. The charac-

teristic temperature for spin fluctuations, just as the Kondo temperature, increases with the band broadening that is induced by hydrostatic pressures. The compensation of a local magnetic moment by Kondo shielding, however, decreases with the pressure-induced decrease in the density of conduction-electron states, eventually leading to an increased interaction between the unshielded part of the local moments and to a positive pressure effects on the magnetic ordering temperature.

## 3. Experimental results

### 3.1. High-field magnetisation

In fig. 1 we present the magnetisation curves for the tetragonal compound URu<sub>2</sub>Si<sub>2</sub> at 1.4 K for directions of the field parallel to the *c* and *a*-axes. The experiments were performed on cylindrical samples (3 mm diameter, 5 to 10 mm length, approximately) that were machined by spark erosion from a Czochralski-grown bulk piece of single-crystalline material. The *c*-axis magnetisation curve has been reproduced for a second monocrystalline sample of the same melt. In the Amsterdam high-field installation [12] magnetic fields can be produced by step-wise pulses with field intervals that can be program-

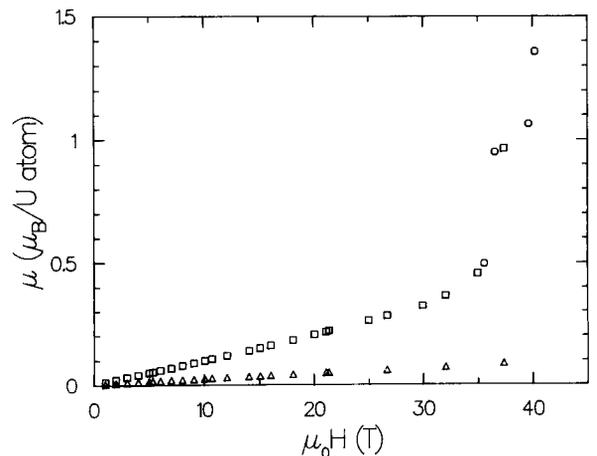


Fig. 1. Magnetisation curves at 1.4 K for monocrystalline samples of tetragonal URu<sub>2</sub>Si<sub>2</sub> along the *c*-axis (□, ○) and along the *a*-axis (△).

med. In this way magnetisation data were taken up to 37.4 T at fields that were constant for 50 ms. Magnetisation data were taken with decreasing fields. By means of a special pulse, in which the field did not reach a really constant value, the behaviour between 37.4 and 40.2 T was determined. The magnetisation curve along the *a*-axis (fig. 1) in the tetragonal plane is approximately linear up to 40 T, with a mean value for the susceptibility of  $(15 \pm 2) \times 10^{-9} \text{ m}^3/\text{mol f.u.}$ . However, along the *c*-axis the magnetisation starts to increase faster than linearly between 15 and 20 T, the susceptibility below these fields being equal to  $(69 \pm 2) \times 10^{-9} \text{ m}^3/\text{mol f.u.}$ , in satisfying agreement with the data of ref. 5. These deviations from linearity grow rapidly with increasing field and result in a value of the measured moment per uranium atom at 35 T of  $0.46 \mu_B$ , which is  $0.11 \mu_B$  larger than expected by a linear extrapolation from the magnetisation data below 15 T. At about 36 T, however, there is a stepwise increase of  $0.45 \mu_B$  in the magnetisation. A second, possibly incomplete, transition occurs at about 40 T, with a jump in the measured moment of  $0.29 \mu_B$ . In both cases the magnetic transition is preceded by increased values for the differential susceptibility just below the transition fields. In order to observe the apparently first-order nature of the magnetic transitions around 36 and 40 T, we followed the magnetisation at a continuous variation of the magnetic field for increasing and decreasing field strengths. A fast recording of the magnetic-field and magnetisation values during this experiment indicates that the hysteresis in the magnetisation curves at the two transitions is certainly less than one tesla (see fig. 2). It should be noted that the magnetisation values obtained from this pulse (denoted by open circles in fig. 1) are less accurate than those obtained at constant fields. High-field magnetisation data have also been obtained on a polycrystalline URu<sub>2</sub>Si<sub>2</sub> sample. In the polycrystalline magnetisation curves no anomalies appear in fields up to 32 T. However, at higher fields a sudden increase in the differential susceptibility is signalled.

In a second series of experiments the magnetisation curves were measured at 4.2 K on the

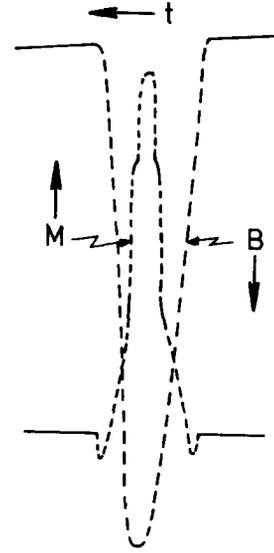


Fig. 2. Magnetisation and field versus time in a high-field pulse experiment on URu<sub>2</sub>Si<sub>2</sub> at 1.4 K, in which a maximum field of 40.2 T was generated. The total pulse length is about 0.6 s. The magnetic transitions at 36 and 40 T can clearly be distinguished.

monocrystalline sample along the different crystallographic directions as well as on the polycrystalline sample. Except for the magnetisation data along the tetragonal axis above 35 T, the 4.2 K curves almost coincide with the 1.4 K curves. Between 35 and 40 T, however, the moments measured at 4.2 K with *B* parallel to the *c*-axis show all the characteristics of a large magnetocaloric effect, i.e. at constant fields the magnetisation does not reach constant values due to a too low cooling rate of the sample immersed in normal liquid helium. This heat exchange between the sample and the surrounding liquid proceeds much faster in superfluid helium. The high-field differential susceptibility follows the temperature dependence of the susceptibility in so far that no anomalies occur perpendicular to the tetragonal axis, whereas along this axis structure in the field and temperature dependence of the susceptibility is observed [5].

### 3.2. High-pressure resistivity

High-pressure resistivity data were taken on the same sample on which the magnetisation

curves along the  $a$ -axis were measured: a cylindrical sample, diameter 3 mm, length about 10 mm. The overall picture of the temperature dependence of the resistivity along this axis is shown in fig. 3. The room-temperature value of the resistivity is  $305 \mu\Omega$  cm, whereas the  $\rho_0$ -value of this annealed sample equals  $20 \mu\Omega$  cm, resulting in a residual resistance ratio of 15. The resistivity curve reveals several anomalies: a relatively weak temperature dependence below 10 K; a sharp anomaly around 17 K followed by a steep increase towards higher temperatures with a maximum around 75 K and a negative temperature derivative above 75 K. Our observations closely agree with the results previously obtained by Palstra et al. [7] on monocrystalline samples prepared from another melt. However, they deviate substantially as far as the absolute values are concerned from recent data by Maple et al. [8] on polycrystalline  $URu_2Si_2$ . These latter authors report room-temperature values that exceed ours by almost one order of magnitude.

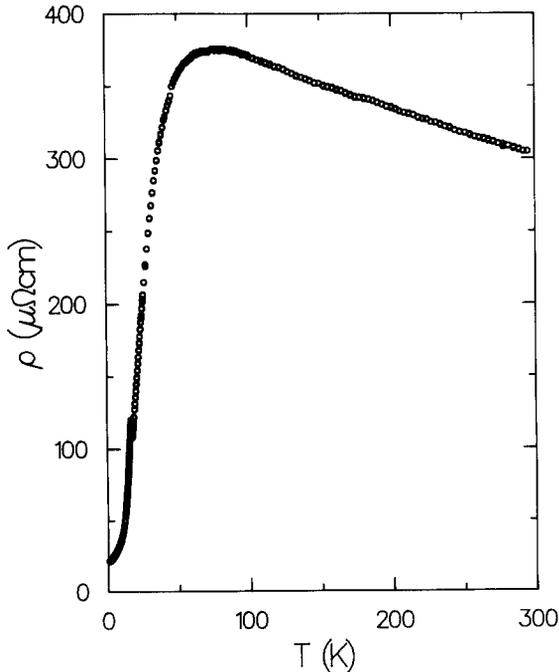


Fig. 3. Resistivity versus temperature curve of the  $a$ -axis sample of  $URu_2Si_2$  between 1.4 and 300 K; the residual resistivity, extrapolated to zero temperature from temperatures above 1.4 K amounts to  $20 \mu\Omega$  cm.

The effect of pressure on the resistivity was studied by a four-terminal method in the same way as reported before by De Visser et al. [2] in their investigation of the compound  $UPt_3$ . Helium served as the pressure transmitting medium and special care was taken to freeze the helium gas under constant pressure in the lower temperature region. Pressures were read in the solid helium pressure range by strain gauges on the beryllium-copper pressure vessel with an accuracy better than 10 bar. At room temperature and at liquid-nitrogen temperature no dramatic changes in resistivity could be observed with increasing pressure: pressure effects were slightly negative but did not exceed 1 percent over 5 kbar. Similar observations hold for the temperature interval between 77 and 290 K. However, in the low-temperature region large and negative pressure effects of 10 percent or more were observed at pressures of 4 kbar. The detailed temperature dependence of the resistivity between 10 and 20 K is shown in fig. 4 at pressures between 1 bar and 3.90 kbar. The residual resistivity turns out to be pressure independent; the increase in the resistivity between 1.5 and 15 K becomes weaker with increasing pressure; the temperatures at which the rounded maximum and the sharp minimum around 17 K occur both increase with pressure with a rate of  $(118 \pm 2)$  mK/kbar, close to the value of 125 mK/kbar reported by Maple et al. [8] for a polycrystalline sample. Above 18 K the resistivity curves at different pressures remain almost parallel, resulting in a pressure-induced shift of the maximum in the resistivity curve to higher temperatures with a rate of about 0.4 K/kbar. Due to the weak temperature dependence of the resistivity above 50 K, it was difficult to extract this value directly from a study of the pressure dependence of the resistivity in the direct neighbourhood of this maximum. The concave temperature dependence of the resistivity between 75 and 290 K is compatible with a Kondo-type of mechanism at higher temperatures. The drop in resistivity below 50 K is similar to those observed in other heavy-fermion systems like  $UBe_{13}$  [1] and  $CeCu_2Si_2$  [13], and could also in this case be

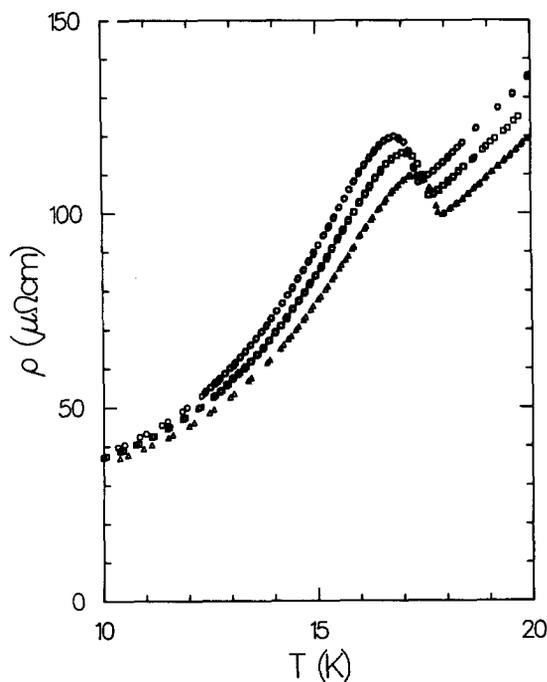


Fig. 4. Resistivity versus temperature curves of an *a*-axis sample of URu<sub>2</sub>Si<sub>2</sub> between 10 and 20 K at different pressures: 1 bar (O); 1.90 kbar (□); 3.91 kbar (Δ).

associated with the onset of coherent scattering. With pressure this onset temperature apparently shifts to higher temperatures with the above given rate of about 0.4 K/kbar. The anomalies in the resistivity curve around 17 K can be characterised by the temperature at which the sharp local minimum occurs: 17.4 K. This temperature is almost equal to the temperature at which the peak in the specific heat occurs (17.5 K) [5, 8].

#### 4. Discussion

As we already indicated above, two different explanations have been proposed so far for the transition at 17.5 K. The arguments for an electronic transition as proposed by Maple et al. [8], find their origin in the observed exponential term in the temperature dependence of the specific heat below this temperature. The pronounced temperature dependence of the resistivity below 17 K reminds to that of the specific heat, which is

explained by Maple et al. in terms of an opening of an energy gap over a part of the Fermi level at 17.4 K and which leads to an exponential term of the form  $\exp(-\Delta/T)$ . According to Schlabitz et al. [4] and Palstra et al. [5], evidence for an itinerant antiferromagnetic ordering is given by a kink in the slope of the susceptibility versus temperature, by a bump in the electrical resistivity and by the lambda-like anomaly in the specific heat below which the coefficient of the linear term in the specific heat is reduced at least by a factor of three compared to the values above 18 K. Palstra et al. [5] compare the specific heat data of URu<sub>2</sub>Si<sub>2</sub> with those of the antiferromagnetic compounds U<sub>2</sub>Zn<sub>17</sub> and UCd<sub>11</sub>, and conclude to the same type of magnetic order for URu<sub>2</sub>Si<sub>2</sub>. We remark that the positive pressure dependence of the transition temperature of 17.4 K in URu<sub>2</sub>Si<sub>2</sub> is indeed similar in sign to that observed in these two compounds [14]. However, as we indicated above, this positive dependence is not suggestive for a purely itinerant type of magnetic order.

In conclusion, we note that the magnetic transitions observed in the magnetisation curves of URu<sub>2</sub>Si<sub>2</sub> are different from the high-field behaviour measured for UPt<sub>3</sub> along directions in the hexagonal plane [6]. In this latter compound a broad peak in the differential susceptibility is observed around 20 T without any sharp increase in the magnetisation. An interpretation of the anomalies in URu<sub>2</sub>Si<sub>2</sub> around 17 K in terms of an antiferromagnetic type of order is supported by the first-order transitions in the magnetisation curve around 36 and 40 T. The two step-wise transitions, however, point to a complex magnetic behaviour. Similar complex magnetisation curves do exist for antiferromagnetically ordered uranium monpnictides, for instance for UP [15]. For this compound, contrary to our observations for URu<sub>2</sub>Si<sub>2</sub>, a large hysteresis is found around the magnetic transitions. Neutron-diffraction studies on other UT<sub>2</sub>Si<sub>2</sub> compounds [16, 17], show simple collinear antiferromagnetic type of order for T = Co and Rh; for T = Ni and Pd transitions occur from magnetic order with an oscillatory character to collinear antiferromagnetism, whereas the compound with T = Cu is a

simple collinear ferromagnetic system. The same neutron experiments indicate that the magnetic moments in these systems are directed along the tetragonal axis and localised on the uranium ions with values that range from  $1.6 \mu_B$  for T = Cu to  $2.9 \mu_B$  for T = Ni. The value of  $1.36 \mu_B$  for URu<sub>2</sub>Si<sub>2</sub> at 40.2 T approaches the just mentioned range of magnetic moment values quite closely. Neutron diffraction studies [18] on this latter compound, however, give an upper limit for the moment per uranium atom of  $0.1 \mu_B$ . This discrepancy between the high-field magnetisation studies and the neutron diffraction results points to fascinating electronic properties that lead to field-induced magnetic moments at 36 and 40 T.

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