



## Anisotropy of Cr-like anomaly in $U_{1-x}Ce_xRu_2Si_2$

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### Abstract

We measured electrical resistivity in  $U_{1-x}Ce_xRu_2Si_2$  single crystals ( $0.00 \leq x \leq 0.050$ ) at ambient pressure and at hydrostatic pressures up to 9 kbar. Our measurements show strong anisotropy, with the in-plane resistivity  $\rho_a$  much higher than  $\rho_c$  parallel to the  $c$ -axis. The substitution of U with Ce in  $URu_2Si_2$  reduces the transition to a Kondo coherent state and the Néel temperature. The minimum in the Cr-like anomaly becomes more prominent but the characteristic shape of the anomaly vanishes. Low temperature characteristic features of  $URu_2Si_2$ , superconductivity and Fermi-liquid behaviour, vanish very rapidly with Ce substitution. The Néel temperature  $T_N$  increases by applying hydrostatic pressure with pressure coefficients  $\partial T_N / \partial p = 0.08 \pm 0.02$  K kbar<sup>-1</sup> for  $x=0.00$  and  $\partial T_N / \partial p = 0.13 \pm 0.02$  K kbar<sup>-1</sup> for  $x=0.025$ . © 1998 Elsevier Science S.A.

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### 1. Introduction

The heavy fermion compounds based on Ce or U elements are very close to the critical point of magnetic instability and their properties are very sensitive to changes of external parameters such as pressure, alloying or external field. Applying hydrostatic pressure leads to volume changes as well as to a change of strength of hybridisation between conduction and f-electrons. This effect is often reflected by a change of the Kondo temperature  $T_K$  or, in the case of concentrated Kondo systems, by a characteristic electronic temperature  $T^*$  [1,2].

$URu_2Si_2$  belongs to a group of heavy fermion materials which display an unusual coexistence of magnetic and superconducting properties. The weak antiferromagnetic magnetic order with an extremely small ordered moment ( $\sim 0.02 \mu_B$ ) below  $T_N = 17.5$  K is considered to arise from the formation of a spin density wave in the quasiparticle bands of the coherent Kondo state and the opening of an energy gap at  $T_N$ . At  $T_N$ , there is a large specific heat jump and Cr-like anomaly in resistivity.  $URu_2Si_2$  exhibits a superconducting transition at  $T_c \sim 1.2$  K. Strong anisotropy of magnetic susceptibility and resistivity is another characteristic feature of  $URu_2Si_2$  [3,4].

Our recent magnetic susceptibility, far-infrared [5] and neutron diffraction measurements [6] support the assumption that the main characteristic features of  $URu_2Si_2$ , a spin-density-wave gap and an antiferromagnetic order, still persist in  $U_{1-x}Ce_xRu_2Si_2$  ( $x \leq 0.05$ ). The size of the magnetic moment and the transition temperature to the ordered state are reduced with Ce substitution of U. In this work we investigate the anisotropy of electrical resistivity in  $U_{1-x}Ce_xRu_2Si_2$  single crystals ( $0.00 \leq x \leq 0.050$ ) at ambient pressure and at hydrostatic pressures up to 9 kbar. We pay special attention to the Cr-like anomaly. We associate the minimum in the anomaly with the Néel temperature.

### 2. Experimental

Single crystals  $U_{1-x}Ce_xRu_2Si_2$  ( $x=0, 0.01, 0.025, 0.05$ ), were grown by a modified tri-arc Czochralski method [7]. The single crystals were grown using the same seed and the same crystal growth procedure. X-ray Laue technique and neutron diffraction confirmed the single-crystalline state of all crystals. Electron-probe microanalyses revealed that all samples are homogenous without any trace of parasitic phases. Samples for electrical resistivity measurements were shaped to blocks with typical dimensions of  $5 \times 1 \times 1$  mm by using a wire saw. Electrical resistivity was

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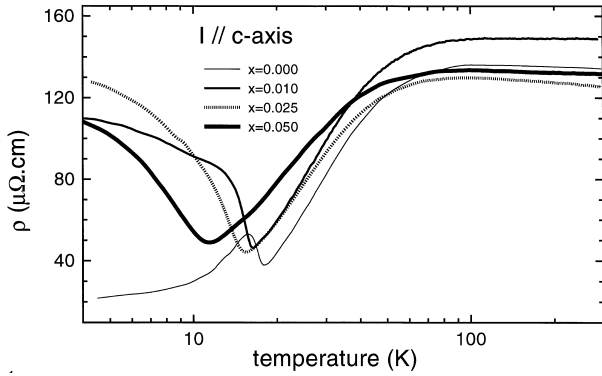


Fig. 1. Typical curves of electrical resistivity along the  $c$ -axis for  $U_{1-x}Ce_xRu_2Si_2$ .

measured by means of a standard DC four-probe method in the temperature range from 350 mK to room temperature at ambient pressure and from 12 K to room temperature at hydrostatic pressures up to 9 kbar. The hydrostatic pressure was generated by using a standard Cu-Be piston cylinder device filled with a mixture of mineral oils as the pressure transmitter. The pressure inside the chamber was measured using a manganin sensor.

### 3. Results and discussion

The electrical resistivity  $\rho_c$  measured along the  $c$ -axis is shown in Fig. 1.  $\rho_c$  is nearly temperature independent down to 100 K, a sharp decrease follows and a minimum occurs at the Néel temperature. The Ce substitution of U in  $URu_2Si_2$  affects only the Cr-like anomaly in a dramatic way. The characteristic shape of the anomaly, the minimum followed by a maximum, vanishes. The resistivity minimum associated with the magnetic transition shifts from  $T_N=17.8$  K for  $x=0$  to temperatures 16.4 K, 15.5 K, and 11.4 K with  $x$  increasing from  $x=0.01$  to 0.025 and 0.05, respectively. The electrical resistivity  $\rho_a$ , measured along the  $a$ -axis, (Fig. 2) exhibits a Kondo-like behaviour

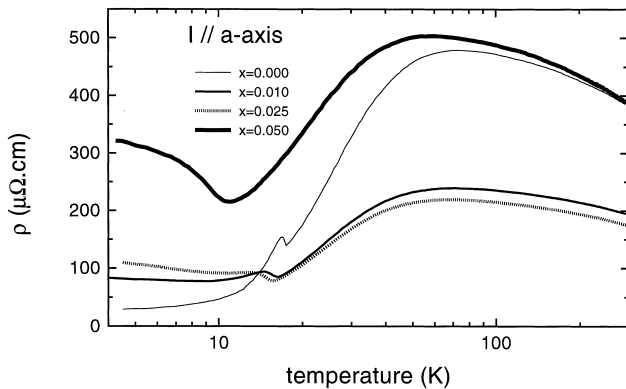


Fig. 2. Typical curves of electrical resistivity along the  $a$ -axis for  $U_{1-x}Ce_xRu_2Si_2$ .

with the negative temperature coefficient down to 90 K followed by a broad maximum. The temperature of the maximum  $T^*=72.5$  K for  $x=0.00$  decreases with Ce substitution to temperatures 70 K, 68 K and 58 K for  $x=0.01$  to 0.025 and 0.05. The maximum at  $T^*$  is followed by a rapid decrease of  $\rho_a$  and an anomaly at the magnetic transition. The small maximum in the Cr-like anomaly similar to  $URu_2Si_2$  was found only for concentrations  $x\leq 0.025$ . At higher concentration,  $\rho_a$  shows only a minimum at the transition temperature. The transition looks like a metal-insulator transition. Reduction of the Néel temperature with Ce substitution is not dependent on crystallographic orientation. The characteristic feature of  $URu_2Si_2$ , i.e. strong anisotropy in resistivity, with  $\rho_a$  several times higher than  $\rho_c$  is reduced at least for  $x\leq 0.025$ ; the residual resistivity  $\rho_a(300\text{ K})/\rho_a(4.2\text{ K})=13.3$ , 2.3 and 1.6 for  $x=0.0$ , 0.010 and 0.025. This feature is strongly sample dependent and usually reflects the quality of a single crystal. We believe that our crystals are of the same quality because the crystals were grown under identical conditions. The reduction of  $\rho_a(300\text{ K})/\rho_a(4.2\text{ K})$  can be attributed to Ce substitution.

Our  $URu_2Si_2$  sample appears superconducting (Fig. 3) with the midpoint of the resistivity transition at  $T_C=1.1$  K. The transition is quite narrow, with a width between 10% and 90% of  $\Delta T_C=0.2$  K. The temperature dependence between 1 K and 17 K can be accurately described by using the formula appropriate for an energy gap  $\Delta$  anti-ferromagnet with an additional  $T^2$ -term describing the Fermi-liquid behaviour [3].

$$\rho = \rho_0 + bT[1 + 2T/\Delta] \exp[-\Delta/T] + cT^2 \quad (1)$$

Best fitting of  $\rho_a(T)$  yields  $\Delta=64.6$  K,  $b=159$   $\mu\Omega\text{cm}/\text{K}$ ,  $c=0.18$   $\mu\Omega\text{cm}/\text{K}^2$  and  $\rho_0=25.5$   $\mu\Omega\text{cm}$  parallel to the  $a$ -axis. Fermi-liquid behaviour is rapidly suppressed by Ce substitution as it is evidenced from the  $\rho_a$  low temperature curve for  $U_{0.99}Ce_{0.01}Ru_2Si_2$ . The inset of Fig. 3 shows anisotropy between  $\rho_a$  and  $\rho_c$  in the low temperature range. The quadratic and linear formula ( $\rho_a=87.1+0.15T-0.3T^2$  and  $\rho_c=113.5-0.7T$ ) give a reasonably good description of the low temperature resistivity data measured in the temperature range from 0.5 K to 3 K, although the physical interpretation is unclear now.

Typical  $\rho_a(T)$  and  $\rho_c(T)$  curves measured on single crystals with  $x=0.0$  and 0.025 between 12 K and 22 K at different hydrostatic pressure up to 6.84 kbar are shown in Fig. 4. To our knowledge, a resistivity measurement at high hydrostatic pressure was for the first time performed on a  $URu_2Si_2$  single crystal. The Néel temperature  $T_N$  increases with applied hydrostatic pressure. We did not find a remarkable difference between pressure coefficients determined from  $\rho_a(T)$  and  $\rho_c(T)$  measurements. From  $\rho_a$  measurements we determined  $\partial T_N(\rho_a)/\partial p=0.09$   $\text{K kbar}^{-1}$  and  $0.12\pm 0.02$   $\text{K kbar}^{-1}$  for  $x=0.0$  and 0.025. From resistivity measurements along the  $c$ -axis, we ob-

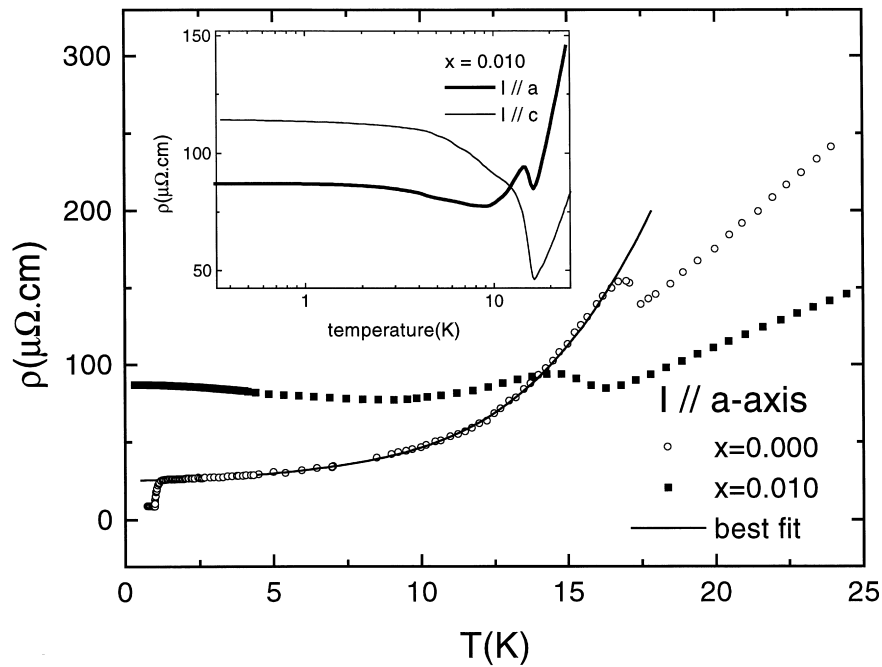


Fig. 3. Electrical resistivity along the  $a$ -axis at low temperatures. The solid line represents the best fit of Eq. (1). The inset shows  $\rho_a$  and  $\rho_c$  for  $U_{0.99}Ce_{0.01}Ru_2Si_2$  on a logarithmic scale.

tained  $\partial T_N(\rho_c)/\partial p = 0.07 \text{ K} \pm 0.02 \text{ K kbar}^{-1}$  and  $0.15 \pm 0.02 \text{ K kbar}^{-1}$  for  $x=0.00$  and  $0.025$ . Our results on  $URu_2Si_2$  are slightly smaller but comparable with results determined on polycrystalline samples [2,8]. The Ce

substitution for U in  $URu_2Si_2$  induces an expansion of elementary cell, inducing a negative chemical pressure [9]. The Vegard's law is roughly fulfilled in the system in the whole concentration range as it follows from our study on

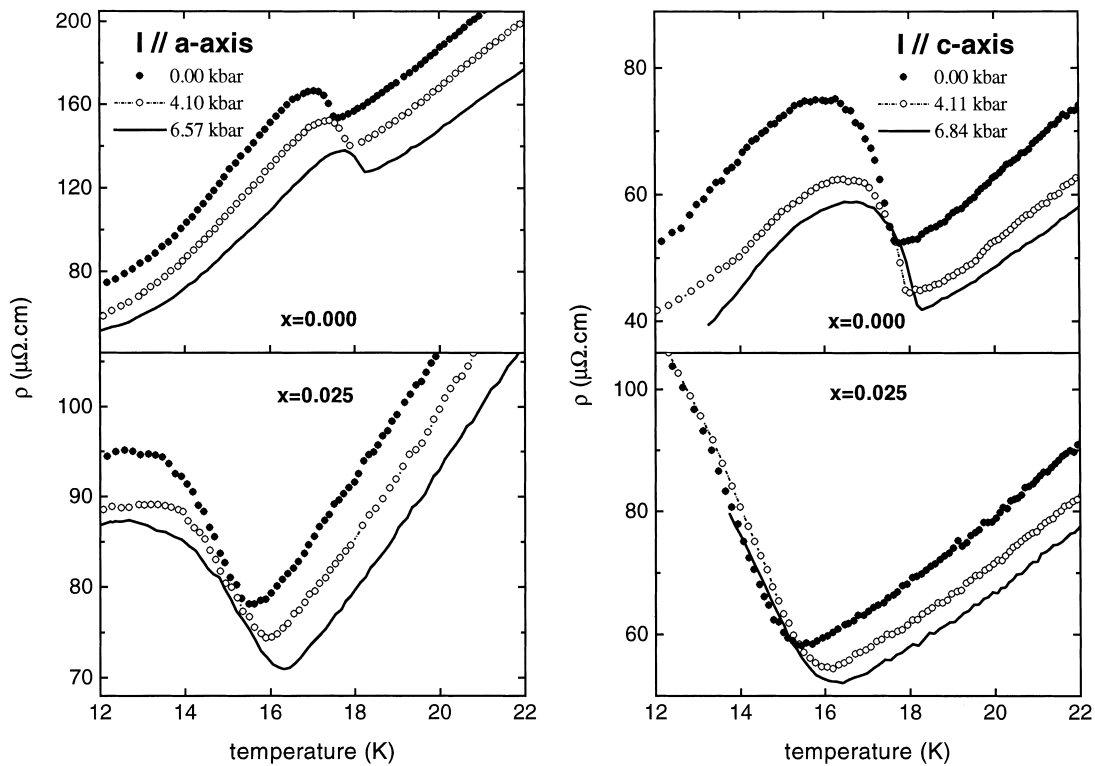


Fig. 4. The temperature dependence of electrical resistivity  $\rho_a$  and  $\rho_c$  for  $U_{1-x}Ce_xRu_2Si_2$  at different hydrostatic pressures.

powdered polycrystalline samples [10]. The volume change due to substitution of 2.5% Ce for U in  $\text{URu}_2\text{Si}_2$  estimated from Vegard's law is  $0.27 \text{ \AA}^3$ . Since the change of the volume with pressure is known for  $\text{URu}_2\text{Si}_2$  [2] we estimate a hydrostatic pressure which induces the same volume change as alloying. In our case, the chemical pressure is negative and equal to  $-4.00 \text{ kbar}$ . An obvious question is now whether the changes in characteristic anomalies like Cr-anomaly can be attributed only to volume changes or not. The change of the Néel temperature induced by chemical pressure  $\Delta_{\text{ch}} = -1.9 \text{ K}$ , reveals a pressure coefficient  $|\partial T_{\text{N}}/\partial p| = 0.48 \text{ K kbar}^{-1}$  which is approximately 4.3 times larger than the value we determined from pressure measurement. Moreover, one can expect that with applying pressure the shape of the anomaly for the sample with  $x=0.025$  should change and should be similar to the Cr-like anomaly in  $\text{URu}_2\text{Si}_2$  if the volume of its elementary cell is approaching the volume of the  $\text{URu}_2\text{Si}_2$  elementary cell. We have found that applied hydrostatic pressure does not change the shape of the anomaly, it merely increases  $T_{\text{N}}$ . Our results support an assumption that volume changes are not only ones responsible for changes in the Cr-anomaly due to U substitution with Ce in  $\text{URu}_2\text{Si}_2$ .

#### 4. Conclusion

Low temperature characteristic features of  $\text{URu}_2\text{Si}_2$  are rapidly suppressed with Ce substitution for U in  $\text{URu}_2\text{Si}_2$ . The resistivity minimum associated with the magnetic transition shifts from  $T_{\text{N}} = 17.5 \text{ K}$  for  $x=0$  to low temperatures  $16.3 \text{ K}$ ,  $15.6 \text{ K}$ , and  $11.0 \text{ K}$ , ( $x=0.01, 0.025, 0.05$ ). The Néel temperature  $T_{\text{N}}$  determined from the temperature dependence of electrical resistivity of  $\text{U}_{1-x}\text{Ce}_x\text{Ru}_2\text{Si}_2$

single crystals, increases with pressure coefficients  $\partial T_{\text{N}}(\rho_{\text{a}})/\partial p = 0.09 \text{ K} \pm 0.02 \text{ K kbar}^{-1}$ ,  $0.12 \pm 0.02 \text{ K kbar}^{-1}$  and  $\partial T_{\text{N}}(\rho_{\text{c}})/\partial p = 0.07 \text{ K} \pm 0.02 \text{ K kbar}^{-1}$  and  $0.15 \pm 0.02 \text{ K kbar}^{-1}$  for  $x=0.00$  and  $0.025$ . We did not find remarkable differences between the pressure coefficient determined from  $\rho_{\text{a}}(T)$  and  $\rho_{\text{c}}(T)$  measurements. Our results support an assumption that not only volume changes are responsible for Ce substitution induced changes of the electrical resistivity in  $\text{URu}_2\text{Si}_2$ .

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