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Magnetic and transport properties of $U_{1-x}Ce_xRu_2Si_2$

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Pseudoternary compounds of the heavy fermion series $U_{1-x}Ce_xRu_2Si_2$ ($x \leq 0.1$ and $x \geq 0.9$) have been characterized by means of electrical resistivity and magnetic susceptibility measurements. High field magnetization measurements revealed an increase of the metamagnetic-like transition field of $CeRu_2Si_2$ by alloying with U, and a suppression of the three-step magnetization process in URu_2Si_2 by alloying with Ce. The influence of chemical pressure is investigated.

1. Introduction

The ternary heavy-fermion compounds URu_2Si_2 and $CeRu_2Si_2$ attract much attention, because of their unusual low-temperature properties [1]. The moderately heavy fermion system URu_2Si_2 exhibits a superconducting transition at $T_c \simeq 1.3$ K and antiferromagnetic order below $T_N = 17.5$ K. The ordered moment is extremely small ($0.03\mu_B$/U atom) [2], but a large moment can be induced in very strong magnetic fields in a three-step magnetization process [3]. The Pauli paramagnet $CeRu_2Si_2$ [4] exhibits neither magnetic nor superconducting order, but pronounced antiferromagnetic intersite correlations were found at low temperatures. The well-known metamagnetic-like transition observed at liquid helium temperatures at a field $B^* \simeq 8$ T (along the tetragonal axis), is interpreted as a suppression of these intersite correlations [5].

Physical properties of heavy fermion materials are very sensitive to the interatomic distance because of the strong hybridization. Hydrostatic pressure is often used as an external parameter in order to produce a controlled variation of the relevant physical quantities. On the other hand, chemical pressure, induced by alloying, has also elucidated the physical properties of many heavy-fermion systems. In particular, alloying revealed that most of the heavy fermion compounds are close to an antiferromagnetic instability [6]. In this paper, we report on an investigation of the pseudoternary series $U_{1-x}Ce_xRu_2Si_2$. We have studied the magnetic and transport properties of URu_2Si_2 doped with small amounts of Ce, and $CeRu_2Si_2$ doped with small amounts of U. As the electronic and magnetic properties of these compounds are strongly anisotropic, we have prepared a number of single-crystalline samples.

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2. Experimental

URu_2Si_2 and $CeRu_2Si_2$ both crystallize in the tetragonal $ThCr_2Si_2$-type structure. Polycrystalline $U_{1-x}Ce_xRu_2Si_2$ samples were prepared by arc melting. For several compositions single-crystalline samples were prepared by the Czochralski technique. The microprobe analysis on the single-crystalline samples revealed formation of 'stripes' with a compositional gradient in the Ce–U ratio. The Debye–Scherrer

Table 1
Lattice parameters and molar volume of tetragonal $U_{1-x}Ce_xRu_2Si_2$ compounds.

x	a (Å)	c (Å)	V (Å^3)
0.05	4.13	9.587	163.56
0.85	4.18	9.754	171.11
0.90	4.19	9.784	171.87
0.95	4.19	9.779	171.94
1.00	4.19	9.794	172.25

method was used to determine the lattice parameters of the powdered single-crystalline samples. The results are presented in table 1. A more detailed account of the metallurgical aspects of the $U_{1-x}Ce_x$ Ru_2Si_2 compounds will be published elsewhere [7].

3. Results

The electrical resistivity, $\rho(T)$, was measured for a number of polycrystalline and single-crystalline $U_{1-x}Ce_xRu_2Si_2$ samples with a standard four-point AC technique. In figs. 1 and 2, we show a few selected results of the $\rho(T)$ data taken on polycrystalline samples and single crystals. On the U-rich side, two regions with a distinctly different resistivity behaviour are observed. For $x \leq 0.05$, the ρ versus T curve is very similar to that of URu_2Si_2, but the Cr-like anomaly at T_N, becomes more prominent. For $x > 0.05$ the shape of the anomaly at T_N is quite different. On alloying with Ce, T_N initially decreases and

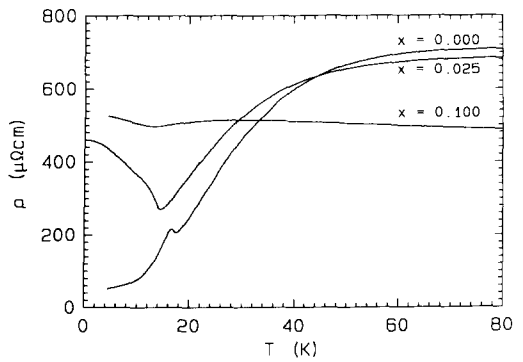

Fig. 1. Temperature dependence of the electrical resistivity of polycrystalline $U_{1-x}Ce_xRu_2Si_2$ for $x = 0.00$, 0.025 and 0.10.

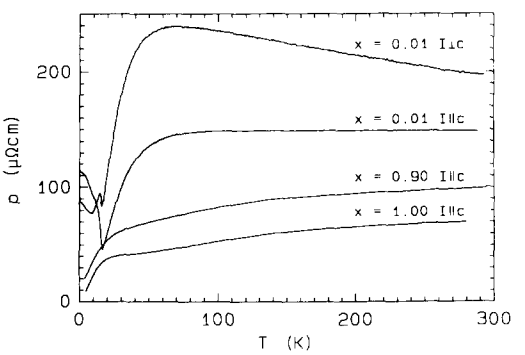

Fig. 2. Temperature dependence of the electrical resistivity of single crystalline $U_{1-x}Ce_xRu_2Si_2$ for $x = 0.01$, 0.90 and 1.00 and for current directions as indicated.

amounts to a value of 14 K for $x = 0.025$. The origin of the faint anomaly at 13 K for $x = 0.1$ is not clear, but possibly indicates a leveling off of the Néel temperature for higher Ce contents. Recently, Roy and Coles [8] reported similar results. Superconductivity is suppressed rapidly by alloying with Ce, as was evidenced by $\rho(T)$ measurements down to 300 mK for $x = 0.01$. On the Ce-rich side, substitution of U leads to an increase in the absolute value of the resistivity, but the overall $\rho(T)$ behaviour remains quite similar to that of pure $CeRu_2Si_2$. No evidence for a magnetic transition was found.

In fig. 3 we show DC susceptibility ($\chi(T)$) data on pure $CeRu_2Si_2$ and $Ce_{0.95}U_{0.05}Ru_2Si_2$, obtained with a commercial Faraday susceptometer (Oxford Instruments) in a field of 0.5 T ($B \parallel c$). The absolute value of χ at low temperatures decreases by alloying and the maximum in $\chi(T)$, at $T^* = 10$ K for $CeRu_2Si_2$, shifts up to 13.8 K for 5 at% U.

In fig. 4 we show the results of the high field magnetization, $M(B)$, measurements performed in pulsed magnetic fields up to 40.5 T produced by the High Magnetic Field Installation of the University of Amsterdam. The metamagnetic-like transition, observed at $B^* = 8.7$ T ($T = 4.2$ K) for $B \parallel c$ in pure $CeRu_2Si_2$, shifts towards higher fields by alloying with U. In contrast to this, the three-step metamagnetic transition, observed at fields of 35.8, 37.3 and 39.4 T in pure URu_2Si_2 [3], is rapidly suppressed by alloying with Ce. For $U_{0.99}Ce_{0.01}Ru_2Si_2$, the data, taken at $T = 1.5$ K, show that the upper and lower transitions are shifted to 34.5 and 38.2 T, while only a faint indication of the middle transition remains visible at 35.5 T. For a 5 at% Ce compound, a considerable increase in $M(B)$ is still observed in the high field range, but no clear metamagnetic behaviour results, indicating a strong broadening of these phenomena by alloying.

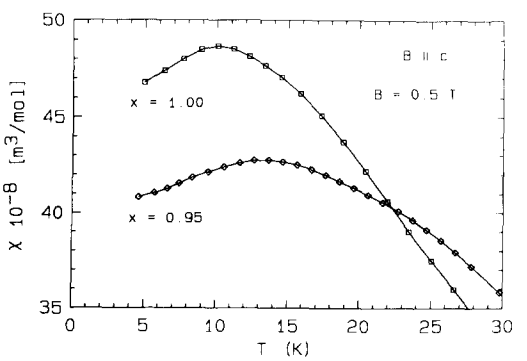

Fig. 3. Magnetic susceptibility ($B = 0.5$ T, $B \parallel c$) of single crystalline $U_{1-x}Ce_xRu_2Si_2$ for $x = 1.00$ and $x = 0.95$.

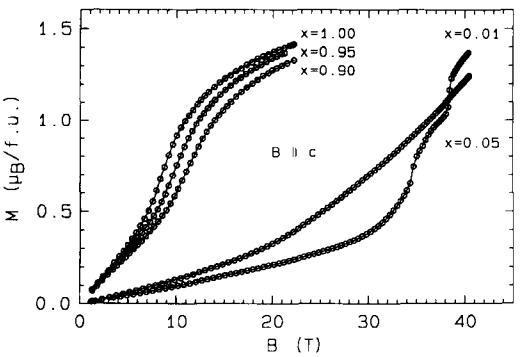

Fig. 4. High field magnetization ($B \parallel c$) of single crystalline $U_{1-x}Ce_xRu_2Si_2$ for $x = 1.00$, 0.95, 0.90 and 0.05 at $T = 4.2$ K and for $x = 0.01$ at $T = 1.5$ K.

4. Discussion

Alloying of heavy-fermion compounds leads in general to a rapid variation of the physical properties because of the strong hybridization. In a few exemplary systems, alloying can be reduced to a chemical pressure effect; in most systems, however, the hybridization is strongly anisotropic and also shape effects (e.g. the change of the c/a ratio) should be taken into account [6]. In (U, Ce)Ru_2Si_2, the situation is even more complicated because of the magnetic character of both the U and Ce ions. However, in a first attempt to explain the data in figs. 1–4, we make a comparison with the effects induced by hydrostatic pressure.

The substitution of Ce in URu_2Si_2 results in an increase of the volume (see table 1), i.e. a negative chemical pressure. As a result T_N initially decreases, in accordance with the resistivity data under hydrostatic pressure that yield an increase of T_N at a rate of 0.4 K/kbar [9]. Unfortunately, a quantitative comparison of the chemical and hydrostatic pressure effects is not possible, because of the spread in the lattice parameters determined so far (the compressibility of URu_2Si_2 is not known). The data in figs. 1, 2 and 4 confirm that very small amounts of impurities strongly effect the antiferromagnetic properties. Similar effects have been observed by substituting Ru by Rh [10]. The present data support the claim that a spin-density wave opens over a part of the Fermi surface [1], as nesting of the Fermi surface is easily destroyed by impurities.

The substitution of U in $CeRu_2Si_2$ results in a decrease of the volume, i.e. a positive chemical pressure. As a consequence, both T^* (fig. 3) and B^* (fig. 4) increase by alloying (at least for low U content, $x > 0.9$). Extensive experimental work performed on $CeRu_2Si_2$ by Mignot et al. [11] revealed that the low temperature properties of $CeRu_2Si_2$ can be described with only one (volume dependent) energy scale, i.e. the thermal and magnetic energy scales are equal: $k_B T^* \approx \mu_B B^*$. This remarkable scaling was in particular demonstrated by the comparison of the thermal and magnetic Grüneisen parameters, $\Gamma_T = -\partial \ln T^* / \partial \ln V$ and $\Gamma_B = -\partial \ln B^* / \partial \ln V$, respectively, as deduced from e.g. susceptibility and magnetization measurements under hydrostatic pressure. The analysis yields an anomalously large value $\Gamma_T \simeq \Gamma_B \simeq 180$. Further support for the scaling law was deduced from alloying studies, e.g. (Ce, La)Ru_2Si_2 and (Ce, Y)Ru_2Si_2. The substitution of small amounts of La and Y on the Ce lattice could be reduced to a negative and positive chemical pressure, respectively. In the following, we investigate the chemical pressure effect induced by alloying with U. For the pure compound, we deduce that $T^* = 10$ K (fig. 3) and $B^* = 8.7$ T (fig. 4). Using the data in table 1, and a value for the Grüneisen parameter of 180, we calculate shifts $\Delta T^* = 3.8$ K and $\Delta B^* = 3.3$ T for 5 at% U, while the shifts deduced from figs. 3 and 4 amount to $\Delta T = 3.8$ K and $\Delta B^* = 1.1$ T, respectively. Hence, a good agreement between the chemical and hydrostatic pressure effect is found from the $\chi(T)$ data, but not from the high field magnetization data.

Specific heat measurements on these pseudo-ternary $U_{1-x}Ce_xRu_2Si_2$ compounds are under way in order to elucidate the present results [7].

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