acta physica slovaca vol. 46 No. 2, 197 – 201 April 1996

STRUCTURE, MAGNETIC AND ELECTRONIC PROPERTIES OF (U,Ce)Ru_2Si_2[1]

S. Maťaš, M. Mihalik, V. Kavečanský, J. Kováč
Institute of Experimental Physics, SAS, Watsonova 47, 043 53 Košice, Slovak Republic

A.A. Menovský
Van der Waals-Zeman Lab., University of Amsterdam, Valckenierstraat 65, 1018 XE Amsterdam

Received 31 July 1995, accepted 8 February 1996

X-ray, magnetisation, susceptibility and heat capacity measurements were used to study the pseudoternary system $U_xCe_{1-x}Ru_2Si_2$ in wide concentration range between $CeRu_2Si_2$ and URu_2Si_2. The Rietveld analysis of X-ray data revealed good solubility of U in $CeRu_2Si_2$ and Ce in URu_2Si_2. The absolute value of susceptibility and the electronic coefficient γ increase with Ce alloying. An anomalous behaviour of the magnetisation and the susceptibility was found for x=0.4 in the magnetic field H < 0.5 T .

1. Introduction

The compounds which contain unstable f-electron shells due to e.g. cerium or uranium elements, are characterized by rich variety of low temperature behaviour. Some of Ce- or U-based intermetalic materials form at low temperatures a highly correlated electron band at the Fermi level and are characterised by the enhanced low temperature electronic coefficient γ and the enhanced magnetic susceptibility χ. Some of these compounds exhibit Pauli paramagnetism (e.g. $CeRu_2Si_2$), ferromagnetism ($URu_{1.2}Re_{0.8}Si_2$ [1]), antiferromagnetism and superconductivity (URu_2Si_2 [2, 3]). The hybridisation between f-electron states and the p- or d-orbitals of ligand atoms in the heavy fermion systems is extremly sensitive to volume effects induced by applied hydrostatic pressure or by alloying (chemical pressure).

2. Results and Discussion

Our measurements [4, 5] taken on single crystals $U_xCe_{1-x}Ru_2Si_2$ (x = 0.99, 0.975, 0.95) showed that Ce substitution strongly affects anomalies in resistivity, susceptibility and heat capacity data which are related to the magnetic phase transition in URu_2Si_2. The transition is associated with weak antiferromagnetic ordering characterised by small

[1] Presented at 9th Czech and Slovak conference on magnetism, Košice, Slovakia, August 28-30 1995

0323-0465/96

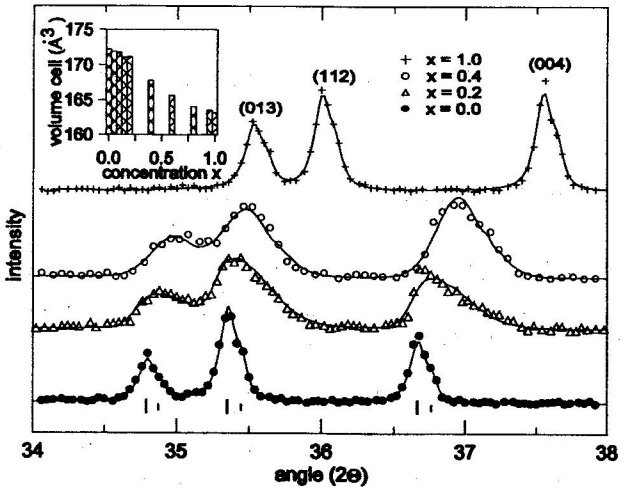

Fig. 1. X-ray results . The inset shows the volume of elementary cell for several x of $U_xCe_{1-x}Ru_2Si_2$.

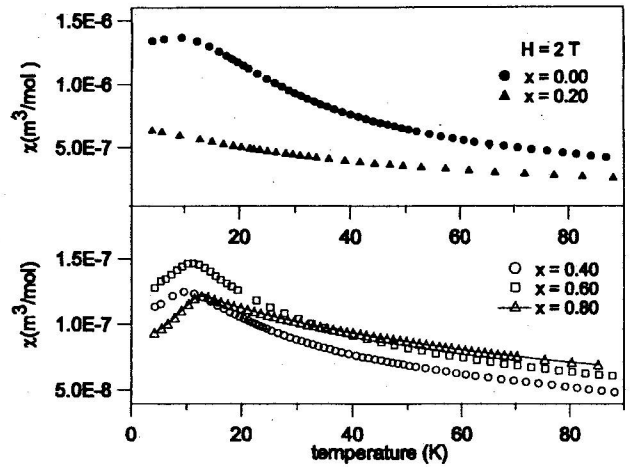

Fig. 2. Susceptibility (χ) as a function of temperature (T) for $U_xCe_{1-x}Ru_2Si_2$, (x=0.0, 0.2, 0.4, 0.6, 0.8).

ordered moment 0.04 μ_B below $T_N = 17.5$ K [2]. The ordering arises from the formation of an energy gap about 11 meV (see references in [5]). The origin of the magnetic transition in $U_xCe_{1-x}Ru_2Si_2$ ($0.99 = x > 0.95$) is probably the same as in URu_2Si_2. The far-infrared measurements of $U_{0.99}Ce_{0.01}Ru_2Si_2$ [6] revealed an energy gap of the same size as in URu_2Si_2. On $U_xCe_{1-x}Ru_2Si_2$ single crystals (x = 0.05, 0.10) we did not find any evidence of long-range antiferromagnetic ordering. The changes of the

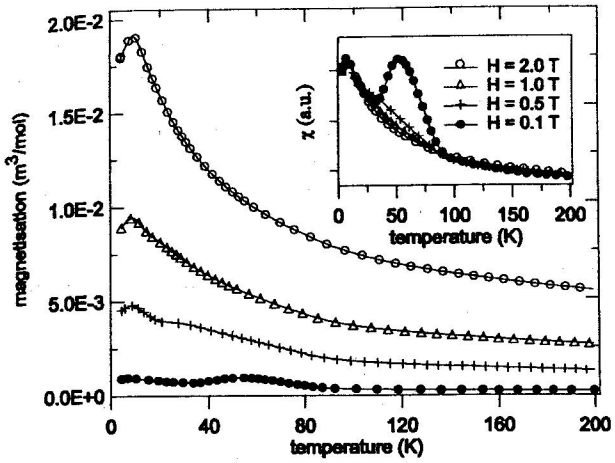

Fig. 3. Magnetisation and susceptibility data of $U_{0.4}Ce_{0.6}Ru_2Si_2$ for different magnetic fields (0.1 T, 0.5 T, 1 T, 2 T). The inset shows susceptibility data.

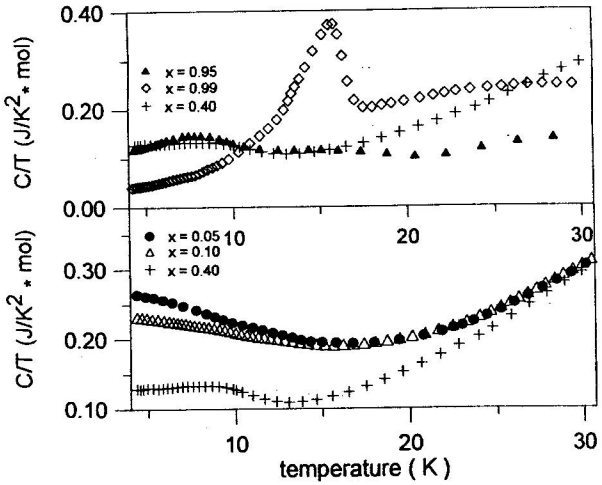

Fig. 4. Heat capacity data of $U_{0.4}Ce_{0.6}Ru_2Si_2$ are compared with data taken on single crystals of $U_xCe_{1-x}Ru_2Si_2$.

susceptibility and the heat capacity were discussed in the frame of volume changes due to the chemical pressure [4, 7].

In this paper we report on x-ray, susceptibility, magnetisation and heat capacity measurements of $U_xCe_{1-x}Ru_2Si_2$ (x = 1, 0.8, 0.6, 0.4, 0.2, 0.0) polycrystalline samples. Several data in the inset of the Fig.1 are taken from [4] and all heat-capacity results, except of the data for $U_{0.4}Ce_{0.6}Ru_2Si_2$, are taken from [5] and [7]. Our samples were prepared by arc-melting stochiometric amounts of the constituent elements under

continuously Ti-gettered argon atmosphere. Fig. 1 shows the strongest X-ray reflections on powdered samples. Symbols represent experimental data and solid line represents a fit after the Rietveld refinement. The Rietveld analysis revealed that each sample adopts the tetragonal $ThCr_2Si_2$ - type structure with the space group I4/mmm. We did not find presence of any second phase. Positioning of alloying atoms in the host lattice led to local volume changes which introduced tensions to the material resulting in broadening of Bragg's reflections. The volume of the elementary cell decreases almost linearly from 172.25 Å for $CeRu_2Si_2$ to 163.15 Å for URu_2Si_2 (see the inset of the Fig. 1). Good solubility of U in $CeRu_2Si_2$ and Ce in URu_2Si_2 was confirmed in whole concentration range in contrast with result presented in [8] where only 45 % solubility of Ce in URu_2Si_2 was found.

Magnetisation measurements were fulfilled by sample vibrating magnetometer in magnetic fields with induction 0.1 T, 0.5 T, 1 T and 2 T. The magnetic susceptibility was almost independent on magnetic field apart from the sample with x=0.4. The broad maximum in susceptibility at 50 K for URu_2Si_2 becomes more sharp and shifts to lower temperatures with Ce alloying and the absolute value of susceptibility is higher (Fig. 2). The results for x<0.4 are very similar to those published in [8]. Our susceptibility data obey Curie-Weiss law above 100 K. Curie-Weiss constant theta and effective magnetic moment estimated from high temperature region decrease with Ce-alloying (3.02 μ_B/f.u., 2.66 μ_B/f.u., 2 μ_B/f.u. for x=0.8, 0.6 to 0.4). The value of moment is smaller compared to that found for single crystal. It is probably caused by high magnetic anisotropy of single crystals [5, 7]. The sample with x = 0.4 represents a border between behaviour which is close to URu_2Si_2 and $CeRu_2Si_2$. We have found an anomalous behaviour of magnetisation for $U_{0.4}Ce_{0.6}Ru_2Si_2$ below 90 K (Fig. 3). A small anomaly develops at about 50 K for magnetic fields 0.1 T and 0.5 T. The anomaly is more visible in susceptibility data (inset of Fig. 3) and it is not possible to explain the anomaly only by magnetic impurities. It seems that some kind of magnetic correlation develops there.

The specific heat data were measured between 1.5 K and 30 K using an adiabatic method. Our heat capacity measurements for Ce-alloying (Fig. 4) show rapid destruction of λ anomaly of URu_2Si_2 and development to the behaviour which is typical for $CeRu_2Si_2$. The sharp λ like anomaly broadens for x = 0.99 and T_N decreases down to 15.5 K. The specific-heat anomaly for x = 0.95 exhibits enhanced value of C/T below 20 K with a grab maximum at 16 K, a broad maximum at 8 K. The specific heat for x=0.4 exhibits small maximum at 8 K (similar to that observed for sample x=0.95) while for temperatures larger than 15 K one can observe a rapid increase of C/T values to value of 0.3 J/K^2*mol at 30 K comparable to the specific heat of sample with x=0.05. (lower part in the Fig. 4). The maximum in C/T(T) at 8 K corresponds to the maximum in susceptibility. The electronic coefficient γ increases with Ce alloying.

3. Conclusions

The Rietveld analysis of our X-ray data revealed good solubility of U in $CeRu_2Si_2$ and Ce in URu_2Si_2. The volume of the elementary cell decreases almost linearly from 172.25 Å for $CeRu_2Si_2$ to 163.15 Å for URu_2Si_2. The absolute value of susceptibility

and the electronic coefficient γ of specific heat increase with Ce alloying. We have found an anomalous behaviour of the magnetisation (susceptibility) for concentration x=0.4 at the temperature of 50 K and for magnetic field 0.1 T and 0.5 T. The observed anomaly can not be explain only by the presence of magnetic impurities or crystal deffects. The anomaly can be an intrinsic property of $U_{0.4}Ce_{0.6}Ru_2Si_2$. It seems that this sample represents a border between behaviour which is close to URu_2Si_2 or to $CeRu_2Si_2$ on the other hand.

References

[1] M.S. Torikachvili, L. Rebelsky, K. Motoya, S.M.Shapiro, Y. Dalichaouch: *Physica B* **163** (1990) 117-120

[2] C. Broholm, J.K. Kjems, W.J.L.Buyers, P. Matthews, T.T. Palstra, A.A. Menovsky and J.A. Mydosh: *Phys. Rev. Lett.* **58** (1987) 1467

[3] M.B. Maple, J.W. Chen, Y. Dalichaouch, T. Kohara, C.Rossel, M.S. Torikachvili, M.W. McElfresh, and J.D. Thopmson: *Phys. Rev. Lett.* **56** (1986) 185

[4] M. Mihalik , A. de Visser, K. Bakker, L.T. Tai, A.A. Menovsky, R.W.A. Hendrikx, T.J. Gortenmulder, S. Maťaš, N. Sato: *Physica B* **186-188** (1993) 507.

[5] M. Mihalik, S. Maťaš, K. Prokeš , H. Nakotte, N.H. Kim-Ngan, A.A. Menovsky: *IEEE Trans. Magn.* **30** (1994) 1142.

[6] D. Macko: unpublished.

[7] M. Mihalik, S. Maťaš, M. Hrabčák, N.H. Kim-Ngan, A.A. Menovsky: *IEEE Trans. Magn.* **30** (1994) 1196.

[8] J.-G. Park, S.B. Roy, B.R. Coles: *J. Phys.: Condens. Matter* **6** (1994) 5937.