Development of the ground state within the UTSi system

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Magnetic properties and specific heat are reported on compounds of the type UTSi, where T is a transition metal. A tendency towards magnetism is found for the compounds with the late transition metals: UCoSi, URuSi, and UIrSi are nonmagnetic, whereas UNiSi, URhSi, UPdSi, and UPtSi are magnetically ordered. This observation agrees well with the expected behavior on the basis of 5*f*-*d* hybridization.

I. INTRODUCTION

In recent years, the electronic properties of the group of UTX compounds, where T represents a late transition metal and X stands for a *p*-type element, have been the subject of many investigations.[1] The compounds with X = Si are the subject of the present investigation. Their basic magnetic properties, together with those of the compounds with X = Ge, have been investigated first by Troc and Tran.[2] These compounds crystallize in the orthorhombic $CeCu_2$ structure, which consists of zigzag U chains, so that each U ion has two U neighbors. The distance between these nearest U neighbors is of the order of 3.50 Å. The separation of the chains gives the somewhat larger distance to the other two U neighbors. In the literature, the transition-metal and Si or Ge atoms have been proposed to be either randomly distributed over the Cu sites or to form the ordered TiNiSi-type structure.

Earlier, we reported high-field magnetization results on UTSi and UTGe compounds[3] and specific-heat, magnetic, and transport properties of UTGe compounds.[4] In this paper, we will present specific-heat and magnetic properties of UTSi compounds.

II. EXPERIMENTAL RESULTS AND DISCUSSION

UTSi samples were prepared in polycrystalline form by arc-melting of stoichiometric amounts of the constituents. X-ray diffraction showed the orthorhombic $CeCu_2$-type structure for most of the as-cast ingots. The x-ray diagrams of the as-cast compounds with Ni, Pd, and Ir could not be indexed on the basis of the $CeCu_2$ type. These compounds were annealed for 4 weeks at 650 °C. After this heat treatment, the compounds with Pd and Ir showed the $CeCu_2$-type structure. UNiSi was found to possess the AlB_2-type structure. The lattice constants of all compounds are in good agreement with the values found in Refs. 2 and 5.

The temperature dependence of the magnetic susceptibility $\chi(T)$, was measured in fields below 0.6 T in the temperature range 10–300 K in a Faraday balance on powder particles fixed by glue. The shapes of the $\chi(T)$ dependencies, which are generally similar to the results given in Ref. 2, were used primarily for indication of magnetic phase transitions. Although the $\chi(T)$ dependencies in the paramagnetic range can be approximated by a modified Curie–Weiss law in most cases, we are aware that parameters obtained on polycrystalline material with a high magnetic anisotropy can be misleading. The magnetization at 4.2 K was measured in the High-Field Installation at the University of Amsterdam in fields up to 35 T and in the High-Field Facility at Osaka University in fields up to 50 T. In order to investigate the anisotropy of the magnetic properties, high-field magnetization measurements were performed on free powder particles oriented by the applied field and on randomly oriented powder fixed by frozen alcohol. The specific heat was measured between 1.3 and 40 K by a standard adiabatic method in magnetic fields up to 5 T.

A. UCoSi

UCoSi is reported to be paramagnetic down to 4.2 K,[2] which is confirmed in the present study. The high-field magnetization at 4.2 K is practically linear up to 35 T [$M(35) = 0.12\ \mu_B$/f.u.]. Within the accuracy of the measurement, there is no difference between the magnetizations of free powder particles and of powder fixed in alcohol, which might suggest a small magnetocrystalline anisotropy. An alternative explanation may be a poor orientation of the powder particles due to the weak magnetic forces. A small amount of ferromagnetic impurity somewhat obscures the susceptibility data. Therefore, the most reliable parameter is the high-field differential susceptibility which equals 21×10^{-9} m^3/mol U. This is very similar to the value of 18×10^{-9} for χ_0 obtained in Ref. 2 in a modified Curie–Weiss-law analysis. No sign of magnetic

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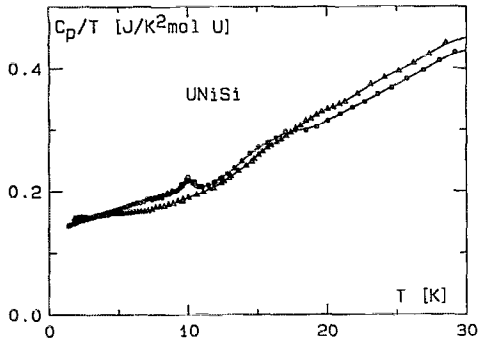

FIG. 1. Temperature dependence of C_p/T for UNiSi at 0 T (○) and 5 T (△).

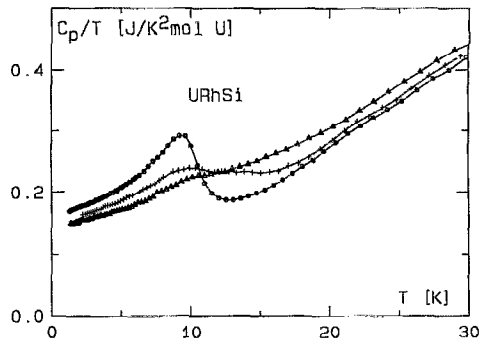

FIG. 2. Temperature dependence of C_p/T for URhSi at 0 T (○), 1 T (+), and 5 T (△).

ordering is observed in the specific heat which was measured down to 1.3 K. A relatively low γ value of 18 mJ/K^2 mol U is found.

B. UNiSi

UNiSi has been reported in Ref. 2 to undergo two antiferromagnetic transitions at 80 and 7.5 K. Moreover, an anomaly in the electrical resistivity was apparent at 18 K. The specific-heat results shown in Fig. 1 exhibit a cusp at 10 K. At higher temperatures, around 17 K, a weak shoulder can be discerned. Both anomalies are suppressed by a magnetic field of 5 T, which points to their magnetic origin. The magnetization at 4.2 K varies almost linearly with the field between 5 and 35 T, but exhibits a nonzero value for the extrapolated magnetization $M(0)$. With increasing field, the magnetization curves of the free and the fixed powder approach each other and at 35 T the magnetic moments are similar (about 0.5 μ_B/f.u.), indicating an anisotropy field of the order of 50 T. The magnetic isotherms measured in the pendulum magnetometer show that the intercept $M(0)$, which indicates the existence of a ferromagnetic component of the magnetization, starts to develop below 18 K. It reaches a maximum value at about 10 K and decreases upon further lowering of the temperature. The observed, relatively high, γ value of 134 mJ/K^2 mol U at 1.4 K may be overestimated due to the magnetic order at 10 K.

C. URuSi

URuSi is reported in Ref. 2 as a weakly temperature-dependent paramagnet. The paramagnetic ground state of this compound has been confirmed by the absence of any anomaly in our specific-heat measurements down to 1.3 K. The low-temperature γ value amounts to 58 mJ/K^2 mol U. At 4.2 K, the magnetization varies proportionally with the field up to the highest field of 35 T with a susceptibilty of 30×10^{-9} m^3/mol U. The magnetization is isotropic within the accuracy of the measurement, i.e., no difference is found between the free and fixed powder magnetizations.

D. URhSi

The ferromagnetic order reported in Ref. 2 to occur in URhSi below 9.5 K is confirmed by the specific-heat results shown in Fig. 2. The somewhat rounded maximum at about 9 K is suppressed and shifted to higher temperatures by an applied external field, as expected for a ferromagnet. The relatively high γ of 133 mJ/K^2 mol U may reflect some magnetic contribution to C even at 1.3 K. The slightly curved high-field magnetic isotherms at 4.2 K measured on free and fixed powder samples, have extrapolated $M(0)$ values of 0.31 and 0.23 μ_B/f.u., respectively, thus exhibiting appreciable anisotropy. At 35 T, the respective magnetizations reach values of 0.67 and 0.52 μ_B/f.u.

E. UPdSi

In Ref. 2 the susceptibility of UPdSi is reported to show two anomalies corresponding to antiferromagnetic transitions: a shoulder at 10 K and a maximum at 29 K, which in the electrical resistivity show up at 7 and 27 K, respectively. Our specific-heat measurement, shown in Fig. 3, reveals only the transition at the higher temperature, in the present case at 26 K. The peak is strongly suppressed by an external field of 5 T and shifted to lower temperatures as expected for a transition to the antiferromagnetic state. The magnetization at 4.2 K shows metamagnetic transitions at 4 and 7 T (Fig. 4). The magnetic moments observed for the free and fixed powders at 35 T correspond

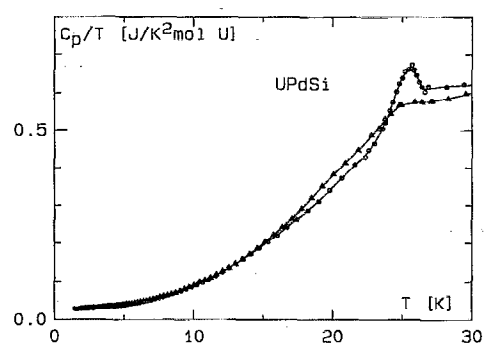

FIG. 3. Temperature dependence of C_p/T for UPdSi at 0 T (○) and 5 T (△).

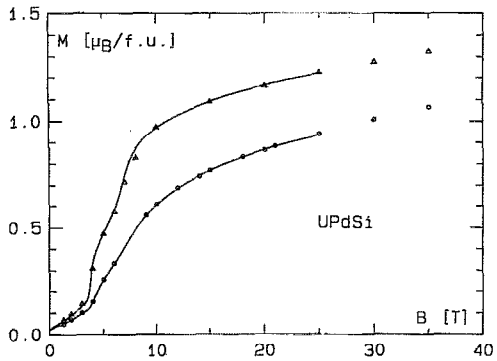

FIG. 4. Magnetization curves for free (△) and fixed (○) UPdSi powder at 4.2 K. The solid curves correspond to data taken during a controlled field pulse in which the field decreased linearly in time at about 55 T/s.

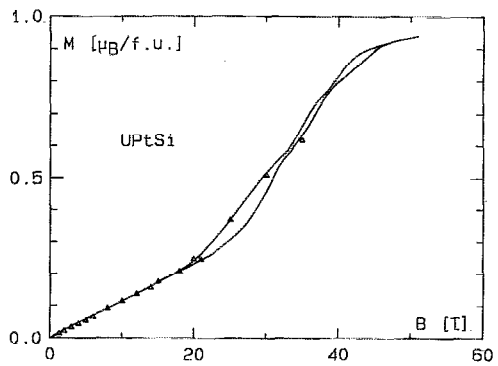

FIG. 5. Magnetization vs field for free UPtSi powder at 4.2 K measured in a 50 T pulse. The ○ symbols correspond to data taken in semicontinuous fields up to 35 T.

to about 1.3 and 1.1 μ_B/f.u., illustrating the strong magnetic anisotropy in UPdSi.

F. UIrSi

The susceptibility of UIrSi has been reported in Refs. 2 and 5 to follow a Curie–Weiss law below room temperature with large negative paramagnetic Curie temperature, followed by a maximum around 30– 40 K. At low temperatures the susceptibility becomes more or less temperature independent. Our specific-heat data confirm the absence of magnetic order down to 1.3 K. The γ value of 11 mJ/K^2 mol U is very low among uranium intermetallics. The magnetization at 4.2 K is linear up to 35 T, yielding the anisotropic susceptibility values of 30×10^{-9} and 39×10^{-9} m^3/mol U for the fixed and free powder samples, respectively.

G. UPtSi

In Ref. 2 it is described that the susceptibility of UPtSi exhibits two peaks at 24 and 50 K, the transition at the latter temperature being confirmed by an anomaly in the electrical resistivity at 47 K and by our susceptibility measurement displaying a maximum around 45 K. The specific heat does not show any transition between 1.3 and 40 K. The low-temperature γ value is 92 mJ/K^2 mol U. The antiferromagnetic ground state is documented by three metamagnetic transitions, accompanied by considerable hysteresis, in the magnetization curve at 4.2 K (see Fig. 5). The magnetization in the highest field of 50 T corresponds to 0.94 μ_B/f.u. (for the free powder), but still increases, so that it cannot be excluded that above 50 T other transitions may occur.

III. CONCLUSIONS

The results presented above show a development from nonmagnetic to magnetically ordered compounds upon proceeding towards the end of each (3*d*, 4*d*, 5*d*) transition-metal series: UCoSi, URuSi, and UIrSi are nonmagnetic, whereas UNiSi, URhSi, UPdSi, and UPtSi are magnetically ordered. This type of development is quite a common feature found already not only for the UTGe system, but also for U–transition-metal compounds in general, and can be attributed to the reduction of 5*f*-*d* hybridization.[1] A comparison with the UTGe system shows, besides a general similarity, rather weaker magnetic properties of the corresponding silicides. As a rule, a lower magnetic susceptibility at 4.2 K is found for the paramagnets and a lower magnetization at 35 T for the magnetically ordered materials. The most significant difference is found between UIrGe with the antiferromagnetic ground state and UIrSi, which is nonmagnetic. The larger atomic volume of Ge with respect to Si, leading to a reduced 5*f*-*p* hybridization in the first case, is a plausible origin of this tendency, which can be traced already in the binaries USi_3 and UGe_3.[6]

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