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Specific heat and magnetic behavior of UTGe compounds

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The temperature dependence of the magnetic susceptibility, electrical resistivity, and specific heat, as well as the magnetization in fields up to 35 T, of polycrystalline samples of UTGe (T = transition metal) compounds has been studied. The development from nonmagnetic UCoGe and URuGe through the weak ferromagnet URhGe, which displays the highest γ value, to complicated types of the local $5f$ -moment ordering for T = Pd, Ni, and Pt is in agreement with the expected trends of gradually reduced $5f$ - d hybridization.

I. INTRODUCTION

Most of the ternary intermetallics of composition UTX, where T represents a late transition metal and X a p -type element, crystallize either in the hexagonal structure (Fe_2P type) or in the orthorhombic one (CeCu_2 type). The choice between them seems to be influenced by the atomic size of the element X. For Si and Ge the CeCu_2 structure is obtained, whereas most of the compounds with larger X atoms, like Al, Ga, Sn, and In, crystallize in the ZrNiAl-type structure, the ordered ternary form of the Fe_2P structure.

The basic magnetic properties of UTGe (and also UTSi) compounds have been studied first by Troc and Tran.¹ Apart from UCoGe and URuGe, in all the remaining Ge compounds a magnetically ordered ground state has been found, in particular ferromagnetic in URhGe and anti-ferromagnetic in UNiGe, UIrGe, and UPtGe. For UPdGe, antiferromagnetic ordering below 50 K followed by a transition to the ferromagnetic state at 32 K has been observed. Ramirez, Batlogg, and Bucher² classified UIrGe as a heavy-fermion compound, since $\gamma = 145 \text{ mJ/K}^2 \text{ mol U}$ was obtained by extrapolation of $C(T)/T$ from the high-temperature region.

The crystal structure of the CeCu_2 type (space group *Imma*) consists of U zig-zag chains, so that each U atom has only two nearest U neighbors at a distance to the order of 3.50 Å. The separation of the chains gives the (somewhat larger) distance to the other two U neighbors. The transition-metal and Si or Ge atoms have been supposed to be randomly distributed over the Cu position¹ or to form an ordered ternary compound (TiNiSi-structure type).^{3,4}

In the present paper we report the specific heat and the magnetic properties of UTGe compounds, in some cases also the electrical resistivity. The results of high-field magnetization measurements have been presented elsewhere.⁵ Here we present some new high-field results on UPtGe.

II. EXPERIMENTAL RESULTS AND DISCUSSION

Polycrystalline UTGe samples were prepared by arc melting stoichiometric amounts of the constituents. The proper CeCu_2 structure was verified by x-ray diffraction. In view of the magnetic anisotropy and possible texture of cast specimens, all magnetic measurements were performed on powders obtained by ball-milling under argon atmosphere. The temperature dependence of the magnetic susceptibility in the range 10–300 K was measured by means of a Faraday balance on powders mixed with glue in order to achieve a random orientation of the powder particles and to prevent orientation in the applied field. The temperature dependencies of the reciprocal susceptibilities of the compounds invariably exhibit deviations from the linear Curie–Weiss behavior, which can be accounted for by fitting the data to the modified Curie–Weiss (MCW) law $\chi(T) = C/(T - \theta) + \chi_0$, where χ_0 represents a temperature-independent contribution. However, in U compounds with strongly anisotropy magnetic properties, it is the anisotropy that, for polycrystalline specimens, gives rise to deviations of the temperature dependence of the magnetic susceptibility from the ideal Curie–Weiss behavior. Therefore, from a physical point of view, in these cases a description in terms of the MCW law is not proper and even misleading, so that we have abstained from performing fits to the MCW law. The high-field magnetization of the polycrystalline materials was studied on samples both in the form of free powder oriented by the applied field and randomly oriented powder fixed by frozen alcohol. Magnetization measurements at 4.2 K were carried out in semicontinuous fields up to 38 T in the High-Field Installation at the University of Amsterdam and in pulsed fields up to 50 T with a total pulse duration of about 400 μs in the High-Field Facility at Osaka University.

A. UCoGe

The low-field part (< 5 T) of the low-temperature magnetization of UCoGe displayed in Ref. 5 exhibits strong curvature, which very likely has to be attributed to the saturation of an impurity contribution, which is also found at higher temperatures in the susceptibility measurements. In the field region between 5 and 10 T the magnetization varies linearly with the field with values for the differential susceptibility as high as $140 \times 10^{-9} \text{ m}^3/\text{mol U}$ for the free powder and $80 \times 10^{-9} \text{ m}^3/\text{mol U}$ for the fixed powder, which should be considered as representative for the intrinsic susceptibility of the compound. At higher fields a slight curvature of the magnetization in the direction of saturation is observed. The high susceptibility gives rise to a field-induced magnetic moment as large as $0.58 \mu_B/\text{f.u.}$ at 35 T, which indicates that UCoGe may be close to ferromagnetic order. The absence of magnetic ordering down to 1.2 K was established by specific-heat measurement. The $C(T)/T$ data show a weak upturn developing below 4 K, which is suppressed by an applied magnetic field of 5 T, yielding $\gamma = 65 \text{ mJ/K}^2 \text{ mol U}$.

B. UNiGe

Similar to the results in Ref. 1 we found a positive paramagnetic Curie temperature θ for UNiGe, which is somewhat puzzling in view of the antiferromagnetic ordering below 40 K, indicated by a sharp maximum in $\chi(T)$. The onset of the magnetic ordering is also manifest in the $R(T)$ dependence, where the negative dR/dT below 100 K leads to a well-resolved maximum at 40 K (Fig. 1). At high temperatures, $R(T)$ displays a very small but positive slope. A complex nature of magnetism in this compound is documented by two metamagnetic-type transitions found in the magnetization curve⁵ with midpoints at about 2 and 10 T (when decreasing the field). A relatively low γ value of $27 \text{ mJ/K}^2 \text{ mol U}$ was derived from the low-temperature specific-heat measurement.

C. URuGe

This compound has been reported in Ref. 1 as a weakly temperature-dependent paramagnet, which was confirmed

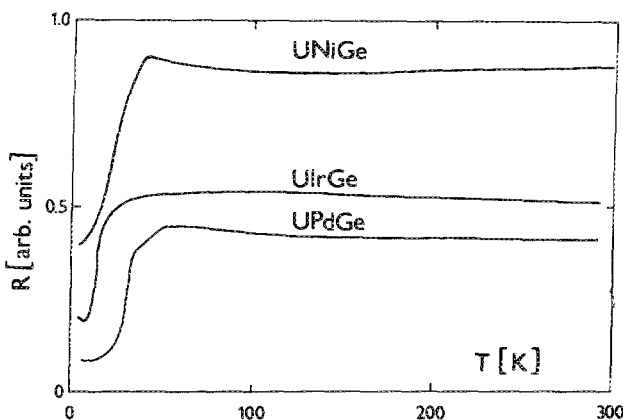


FIG. 1. Temperature dependence of the electrical resistivity of UNiGe, UPdGe, and UIrGe. Absolute values are not given due to the large error introduced by internal cracks.

in our susceptibility measurements. The absence of any magnetic ordering down to 1.2 K is shown by specific-heat measurements, which yield a moderate γ value of $35 \text{ mJ/K}^2 \text{ mol U}$. The high-field magnetization at 4.2 K (Ref. 5) exhibits magnetically isotropic behavior and depends linearly on the field with a susceptibility value of $30 \times 10^{-9} \text{ m}^3/\text{mol U}$. This value can be compared with $18 \times 10^{-9} \text{ m}^3/\text{mol U}$ observed for the susceptibility at room temperature.

D. URhGe

This compound has been described¹ as an itinerant-electron ferromagnet due to the low value of its magnetic moment and the lack of magnetic saturation. This conclusion is supported by the high-field magnetization measurements,⁵ which show absence of saturation up to 35 T. The magnetization curves for the free and fixed powders are almost parallel with a difference of about $0.16 \mu_B/\text{f.u.}$ The specific-heat data (Fig. 2) display a pronounced anomaly with a maximum at 8.7 K, corresponding to the transition to the ferromagnetic state. This sharp feature is suppressed by a field of 5 T. The γ coefficient cannot be unequivocally specified, as the magnetic contribution to the specific heat does not vanish even at lowest temperatures. A rough estimate leads to a γ value between 100 and $150 \text{ mJ/K}^2 \text{ mol U}$.

E. UPdGe

Our susceptibility measurements confirm the existence of the paramagnetic-antiferromagnetic transition at about 50 K and the antiferromagnetic transition at 32 K, which have been reported in Ref. 1. The anomaly in $C(T)$ at the lower transition is strongly suppressed by a magnetic field, but the γ value of $22 \text{ mJ/K}^2 \text{ mol U}$ remains unaffected. Both magnetic transitions can also be seen in $R(T)$ (Fig. 1). Above 60 K a negative dR/dT is observed. Similar to URhGe the magnetization at 4.2 K (Ref. 5) does not saturate even in the highest fields.

F. UIrGe

A very pronounced peak in $\chi(T)$ at 18 K, pointing to an antiferromagnetic transition, correlates well with the results

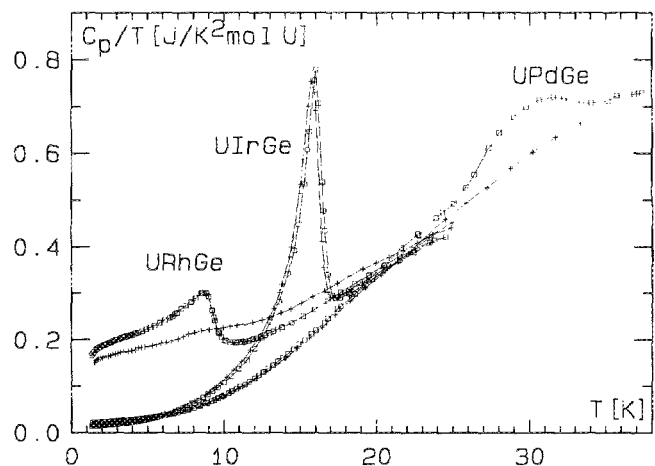


FIG. 2. Temperature dependence of C/T for URhGe, UPdGe, and UIrGe in 0 T (\square) and 5 T ($+$).

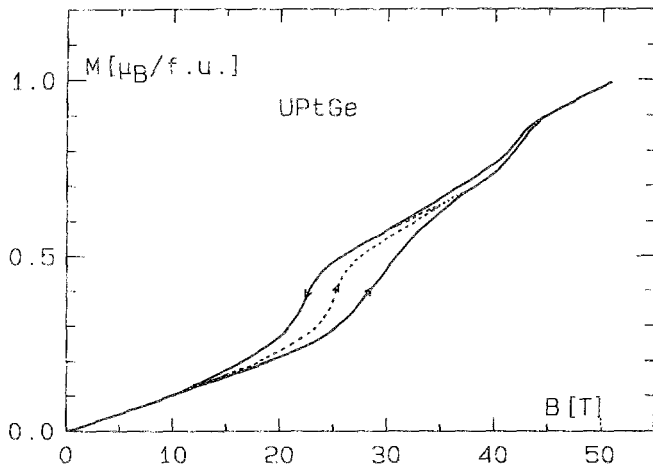


FIG. 3. Magnetization curves of free UPtGe powder at 4.2 K. The solid line corresponds to a 50-T pulse, the dashed curve to a 40-T pulse.

of others.^{1,2} A complicated character of the magnetic structure can be anticipated because of the two metamagnetic-type transitions observed at 4.2 K.⁵ A broad maximum in $R(T)$, found around 70 K may be indicative of spin-fluctuation effects. In the specific heat the antiferromagnetic transition is manifest by a sharp maximum located at 16 K (Fig. 2). The shape and position of the anomaly are only slightly influenced by the applied field of 5 T: T_{\max} shifts to lower temperatures by about 0.5 K. The γ value of 14 mJ/K² mol U is curiously low among uranium transition-metal intermetallics.

G. UPtGe

In accordance with Ref. 1 pronounced susceptibility maximum was observed at 51 K, indicating a transition to the antiferromagnetic state. The magnetic isotherms at 4.2 K taken on free powder in short-pulse (400 μ s) fields (Fig. 3) at Osaka University show two broad field-induced transitions around 23 and 42 T (with decreasing field). The transition at the lower field exhibits very strong hysteresis and a strong dependence of the transition field upon the rate of increase of the field with time is observed during the up-sweep of the field: In the 40-T pulse the transition occurs at 25 T, whereas in the 50-T pulse the transition is much broader and takes place around 28 T. In the highest fields the magnetization increases still strongly and other transition(s) above 50 T cannot be excluded. These results obtained by means of short-pulse fields can be compared with the results in Fig. 4 obtained at Amsterdam University in semicontinuous fields (triangles for the fixed powder and circles for the free powder) and in pulsed fields with a much larger pulse duration of the order of 1 s. It can be seen that the agreement is quite satisfactory. The dependence of the hysteretic behavior on the rate of change of the field is further illustrated by a reduction of the width of the hysteresis loop by a factor of 2 with respect to the 40-T result in Fig. 3.

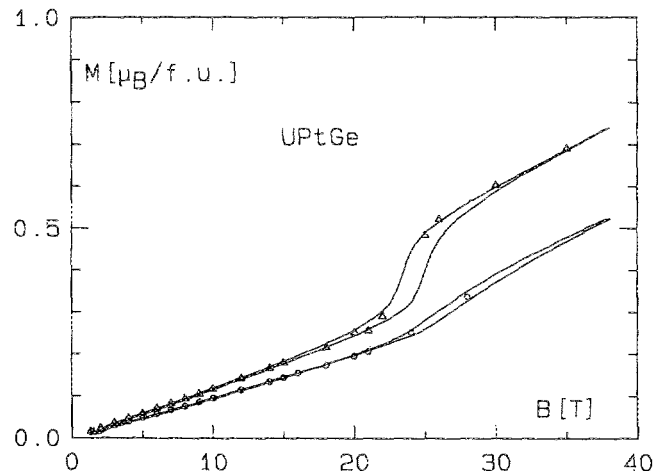


FIG. 4. Magnetization curves of free (Δ) and randomly oriented (O) UPtGe powder at 4.2 K. The solid curves correspond to data taken during an uncontrolled increase of the field up to 38 T in about 0.2 s, followed by a decrease linearly in time at about 55 T/s.

III. CONCLUSIONS

The development from nonmagnetic to magnetically ordered compounds when proceeding towards the end of the transition-metal series is a quite common feature of U compounds with transition metals ascribed to the reduction of $5f$ - d hybridization.⁶ Besides the general similarity, there are features specific for UTGe compounds when compared, e.g., to UTX compounds with the ZrNiAl hexagonal structure. Most of the latter compounds are magnetically uniaxial with the easy axis perpendicular to high-U-coordination planes. The symmetry in the UTGe compounds is probably more complicated. Magnetization data point either to an itinerant character of all compounds, or to an incomplete field alignment of the $5f$ moments of high fields for T = Ni, Pd, Ir, and Pt. The existence of local $5f$ moments is indicated by the low γ values in these compounds.

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