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Possible heavy-fermion behaviour of new $U(Cu, Al)_5$ compounds

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Abstract

We have synthesized several new UCu_xAl_{5-x} compounds in the composition range between $x = 2.9$ and $x = 3.5$, which were found to form in crystal structures related to the $CaCu_5$ structure. Specific-heat measurements reveal a considerable enhancement of the low-temperature specific-heat coefficient γ for all $U(Cu, Al)_5$ compounds investigated, with a maximum value of 450 mJ/molK^2 at 1.2 K for $UCu_{2.9}Al_{2.1}$.

The discovery of the heavy-fermion superconductors UNi_2Al_3 [1] and UPd_2Al_3 [2] which crystallize in the $CaCu_5$-derived $PrNi_2Al_3$ structure, has turned the attention to other compounds formed in the $CaCu_5$ structure. Among U-compounds with Cu and Al, only $UCu_{3.5}Al_{1.5}$ has been reported up to now to crystallize in this structure [3]. Recently, we have reported on the structural and magnetic properties of UCu_3Al_2 [4]. Using neutron diffraction, this compound was found to form in an ordered variant of the $CaCu_5$ structure, where U–Cu_2 layers are separated by layers of Al and the remaining Cu atoms, which are randomly distributed over the 3g sites. The high-field-magnetization and magnetic-susceptibility measurements performed on a single crystal were interpreted in terms of an antiferromagnetic ground state [4]. In order to study the influence of 5f-ligand hybridization on the occurrence of magnetic ordering in more detail, we have investigated $U(Cu, Al)_5$ compounds over a more extended composition range.

Various UCu_xAl_{5-x} compounds have been prepared by arc-melting stoichiometric amounts of the elements. After annealing for two months at 600°C, only samples with Cu compositions between $x = 2.9$ and 3.5 were found to form in the proper $CaCu_5$ structure (Fig. 1). As the crystallographic ordered version of the $CaCu_5$ structure has been found for UCu_3Al_2, a possible random distribution of the Cu and Al atoms over the Cu sites of the $CaCu_5$ structure for the Cu-rich compositions may occur. Microprobe analysis reveals the proper composition and the absence of any impurity phase for all compounds reported here, except a small amount of UAl_2 for $UCu_{2.9}Al_{2.1}$ and some composition fluctuations for $UCu_{3.2}Al_{1.8}$.

We performed measurements of the specific heat, the magnetic susceptibility and the high-field magnetization on UCu_xAl_{5-x} compounds with $x = 2.9$, 3.0, 3.1, 3.4 and 3.5. The specific heat shows a broad maximum at about 12 K in $UCu_{3.1}Al_{1.9}$. This maximum is shifted to about 8 K for UCu_3Al_2 and appears as a shoulder at about 4 K in $UCu_{2.9}Al_{2.1}$ (see Fig. 2). Although, these anomalies were found to be hardly affected by an applied field of 5 T, we can speculate about the origin in the long-range order of small U-moments. Such anomalies are not observed for $UCu_{3.4}Al_{1.6}$ and $UCu_{3.5}Al_{1.5}$. Another interesting feature is the strong enhancement of the specific heat at low temperatures, which was found to increase with decreasing Cu content for $x \leq 3.1$ and leads to γ-values as large as 450 mJ/molK^2 at 1.2 K for $UCu_{2.9}Al_{2.1}$. For the Cu-rich

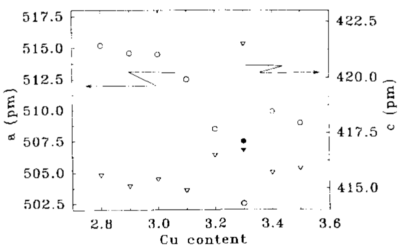

Fig. 1. Variation of lattice parameters *a* (circles) and *c* (triangles) versus copper content *x*. Note, two sets of lattice parameters were found for $UCu_{3.3}Al_{1.7}$ (open and closed symbols), indicating a change from ordered type of structure to a disordered one.

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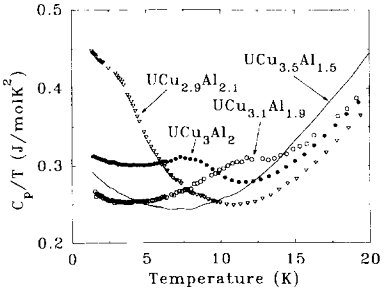

Fig. 2. Temperature dependence of the specific heat of various UCu_xAl_{5-x} compounds in the representation c_p/T vs. T.

compositions ($x \geq 3.4$), we observe smaller γ-values (about 290 $mJ/molK^2$) which do not depend much on the stoichiometry. For these compounds, we find that the upturn in c_p/T vs. T cannot be satisfactorily fitted with an additional T^3lnT-term to the specific heat derived from the paramagnon theory [5]. Much better fits, however, can be achieved with an additional quadratic term, usually attributed to the occurrence of a spin-glass state [6]. The disorder of the non-magnetic atoms, which is indicated for the Cu-rich compounds, indeed promotes some randomness in exchange interactions and may eventually lead to the formation of a spin-glass state. Gschneidner et al. [7] have shown that spin-glass behaviour arising from non-magnetic atomic disorder (NMAD) may cause a large enhancement of the low-temperature specific heat. However, the above mechanism cannot be used in order to explain the enhancement of the low-temperature specific heat in UCu_xAl_{5-x} compounds with $x \leq 3.1$, because complete disorder of Cu and Al atoms was ruled out for these compounds as indicated from neutron-diffraction results on UCu_3Al_2 [4]. For this compound, the strong magnetic anisotropy found in single-crystal studies can be taken as a further argument against a spin-glass state. On the other hand, the observation of high γ-values in all $U(Cu,Al)_5$ compounds discussed here as well as in other U ternaries containing Cu, e.g., $U(Cu,Ga)_5$ [8] and $U(Cu,Al)_{12}$ [9], may indicate that some other mechanism is responsible for the common enhancement of c/T in Cu-containing U ternaries. For $x \leq 3.1$, the onset of magnetic ordering is manifest in the occurrence of maxima in the temperature dependence of the magnetic susceptibility at slightly higher temperatures than those found in the specific-heat measurements. In all cases, we find the Curie–Weiss behaviour obeyed for temperatures above 50 K leading to paramagnetic Curie temperatures Θ_p between −100 K (for $UCu_{2.9}Al_{2.1}$) and −150 K (for $UCu_{3.5}Al_{1.5}$) and effective moments μ_{eff} around 3.5 μ_B/f.u. In the Amsterdam High-Field Installation, magnetization measurements were performed on powder particles free to be oriented in magnetic fields up to 35 T. The results corroborate a possible antiferromagnetic ground state of the compounds with $x \leq 3.1$ as these compounds exhibit a relatively broad metamagnetic transitions (similar to the one shown in Ref. [4]) starting at fields of about 8, 15 and 25 T for $UCu_{2.9}Al_{2.1}$, UCu_3Al_2 and $UCu_{3.1}Al_{1.9}$, respectively. All $U(Cu,Al)_5$ compounds investigated display a lack of saturation in magnetic fields up to 35 T. Therefore, the magnetization measurements on UCu_3Al_2 and $UCu_{3.5}Al_{1.5}$ have been extended to 50 T at the Osaka High-Field Facility revealing a magnetic response at 50 T of about 0.95μ_B and 1.05μ_B, respectively, but still a considerably high-field susceptibility is present for both compounds. This emphasises that for the determination of the ordered moments even higher magnetic fields and/or neutron diffraction results are needed. Furthermore, comparing the magnetization on 'free powders' with measurements performed on powder particles fixed in random orientations by frozen alcohol, we find a change in the type of magnetocrystalline anisotropy for the ordered $U(Cu,Al)_5$, which is indicated by the ratio of M_{fix}/M_{free}. In going from UCu_3Al_2 to $UCu_{3.1}Al_{1.9}$ the type of anisotropy presumably changes from multiaxial to uniaxial. To clarify this behaviour single-crystal results are required.

In conclusion, we have reported on the structural and electronic properties of new $U(Cu,Al)_5$ compounds. Anomalies in the bulk measurements presented indicate a possible antiferromagnetic ground state for the crystallographically ordered compounds. For all $U(Cu,Al)_5$ compounds investigated, we find a considerable enhancement of the specific heat at low temperatures. Whether this enhancement is due to a heavy-fermion state in these compouns cannot be decided at present.

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