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

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Abstract

We have synthesized several new $\text{UCu}_x\text{Al}_{5-x}$ 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 $\text{U}(\text{Cu}, \text{Al})_5$ compounds investigated, with a maximum value of 450 mJ/molK^2 at 1.2 K for $\text{UCu}_{2.9}\text{Al}_{2.1}$.

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 $\text{UCu}_{3.5}\text{Al}_{1.5}$ 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–Cu₂ 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 $\text{U}(\text{Cu}, \text{Al})_5$ compounds over a more extended composition range.

Various $\text{UCu}_x\text{Al}_{5-x}$ 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 $\text{UCu}_{2.9}\text{Al}_{2.1}$ and some composition fluctuations for $\text{UCu}_{3.2}\text{Al}_{1.8}$.

We performed measurements of the specific heat, the magnetic susceptibility and the high-field magnetization on $\text{UCu}_x\text{Al}_{5-x}$ 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 $\text{UCu}_{3.1}\text{Al}_{1.9}$. This maximum is shifted to about 8 K for UCu_3Al_2 and appears as a shoulder at about 4 K in $\text{UCu}_{2.9}\text{Al}_{2.1}$ (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 $\text{UCu}_{3.4}\text{Al}_{1.6}$ and $\text{UCu}_{3.5}\text{Al}_{1.5}$. 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 $\text{UCu}_{2.9}\text{Al}_{2.1}$. 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 $\text{UCu}_{3.3}\text{Al}_{1.7}$ (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 $\text{UCu}_x\text{Al}_{5-x}$ 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 $T^3 \ln T$ -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 $\text{UCu}_x\text{Al}_{5-x}$ 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 $\text{U}(\text{Cu}, \text{Al})_5$ compounds discussed here as well as in other U ternaries containing Cu, e.g., $\text{U}(\text{Cu}, \text{Ga})_5$ [8] and $\text{U}(\text{Cu}, \text{Al})_{12}$ [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 $\text{UCu}_{2.9}\text{Al}_{2.1}$) and -150 K (for $\text{UCu}_{3.5}\text{Al}_{1.5}$) and effective moments μ_{eff} around $3.5 \mu_B/\text{f.u.}$ 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 rela-

tively broad metamagnetic transitions (similar to the one shown in Ref. [4]) starting at fields of about 8, 15 and 25 T for $\text{UCu}_{2.9}\text{Al}_{2.1}$, UCu_3Al_2 and $\text{UCu}_{3.1}\text{Al}_{1.9}$, respectively. All $\text{U}(\text{Cu}, \text{Al})_5$ compounds investigated display a lack of saturation in magnetic fields up to 35 T. Therefore, the magnetization measurements on UCu_3Al_2 and $\text{UCu}_{3.5}\text{Al}_{1.5}$ have been extended to 50 T at the Osaka High-Field Facility revealing a magnetic response at 50 T of about $0.95 \mu_B$ and $1.05 \mu_B$, 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 $\text{U}(\text{Cu}, \text{Al})_5$, which is indicated by the ratio of $M_{\text{fix}}/M_{\text{free}}$. In going from UCu_3Al_2 to $\text{UCu}_{3.1}\text{Al}_{1.9}$ 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 $\text{U}(\text{Cu}, \text{Al})_5$ compounds. Anomalies in the bulk measurements presented indicate a possible antiferromagnetic ground state for the crystallographically ordered compounds. For all $\text{U}(\text{Cu}, \text{Al})_5$ 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 compounds cannot be decided at present.

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