Possible heavy-fermion behaviour of new U(Cu,Al)5 compounds

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

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Abstract
We have synthesized several new \( \text{U(Cu, Al)}_5 \) compounds in the composition range between \( x = 2.9 \) and \( x = 3.5 \), which were found to form in crystal structures related to the \( \text{CaCu}_5 \) structure. Specific-heat measurements reveal a considerable enhancement of the low-temperature specific-heat coefficient \( \gamma \) for all \( \text{U(Cu, Al)}_5 \) compounds investigated, with a maximum value of 450 mJ/molK² at 1.2 K for \( \text{UCu}_{2.9}\text{Al}_{2.1} \).

The discovery of the heavy-fermion superconductors \( \text{UNi}_2\text{Al}_3 \) [1] and \( \text{UPd}_2\text{Al}_3 \) [2] which crystallize in the \( \text{CaCu}_5 \)-derived \( \text{PrNi}_2\text{Al}_3 \) structure, has turned the attention to other compounds formed in the \( \text{CaCu}_5 \) structure. Among U-compounds with Cu and Al, only \( \text{UCu}_{3.2}\text{Al}_{1.5} \) has been reported up to now to crystallize in this structure [3]. Recently, we have reported on the structural and magnetic properties of \( \text{UCu}_{3.3}\text{Al}_{1.7} \) [4]. Using neutron diffraction, this compound was found to form in an ordered variant of the \( \text{CaCu}_5 \) structure, where \( \text{U-Cu}_2 \) layers are separated by layers of Al and the remaining Cu atoms, which are randomly distributed over the \( \text{3g} \) 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 \( \text{U(Cu, Al)}_5 \) 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 \( x = 3.5 \) were found to form in the proper \( \text{CaCu}_5 \) structure (Fig. 1). As the crystallographic ordered version of the \( \text{CaCu}_5 \) structure has been found for \( \text{UCu}_3\text{Al}_2 \), a possible random distribution of the Cu and Al atoms over the Cu sites of the \( \text{CaCu}_5 \) 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 UAl₂ 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 \( \text{UCu}_3\text{Al}_2 \) 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 \( \text{UCH}_3.4\text{Al}_{1.6} \) and \( \text{UCH}_3.5\text{Al}_{1.5} \). 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 \( \gamma \)-values as large as 450 mJ/molK² at 1.2 K for \( \text{UCu}_{2.9}\text{Al}_{2.1} \). For the Cu-rich compositions, the Cu content may affect the properties of these compounds.

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Fig. 1. Variation of lattice parameters \( a \) (circles) and \( c \) (triangles) versus copper content \( x \). Note, two sets of lattice parameters were found for \( \text{UCu}_{3.1}\text{Al}_{1.7} \) (open and closed symbols), indicating a change from ordered type of structure to a disordered one.
compositions ($x \geq 3.4$), we observe smaller $\gamma$-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 UCu$_{1.4}$Al$_{1.6}$ 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$_3$Al$_2$ [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 $\gamma$-values in all UCu$_x$Al$_{3-x}$ compounds discussed here as well as in other U ternaries containing Cu, e.g., UCu(AI)$_{2-3}$ [8] and UCu(Cu, AI)$_2$ [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 $\Theta$, between $-100$ K (for UCu$_{2.9}$Al$_{1.1}$) and $-150$ K (for UCu$_{3.5}$Al$_{1.5}$) and effective moments $\mu_{\text{eff}}$ around 3.5 $\mu_B$/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$_{1.1}$, UCu$_{1.4}$Al$_2$ and UCu$_{3.5}$Al$_{1.5}$, respectively. All UCu$_x$Al$_y$ compounds investigated display a lack of saturation in magnetic fields up to 35 T. Therefore, the magnetization measurements on 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 0.95$\mu_B$ 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 UCu$_3$Al$_2$, which is indicated by the ratio of $M_{\text{ix}}/M_{\text{rec}}$. In going from UCu$_{3.6}$Al$_{1.9}$ 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 UCu$_x$Al$_{3-x}$ compounds. Anomalies in the bulk measurements presented indicate a possible antiferromagnetic ground state for the crystallographically ordered compounds. For all UCu$_x$Al$_y$ 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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References