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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$_x$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 $\gamma$ for all U(Cu, Al) 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$_2$Al$_3$ [1] and UPd$_2$Al$_3$ [2] which crystallize in the CaCu$_5$-derived PrNi$_2$Al$_3$ structure, has turned the attention to other compounds formed in the CaCu$_5$ structure. Among U-compounds with Cu and Al, only UCu$_{3.3}$Al$_{1.7}$ has been reported up to now to crystallize in this structure [3]. Recently, we have reported on the structural and magnetic properties of UCu$_{3.3}$Al$_2$ [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$_x$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$_{3.3}$Al$_2$, 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$_x$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 UCu$_{3.3}$Al$_{1.7}$. This maximum is shifted to about 8 K for UCu$_{3.3}$Al$_2$ 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$_{1.8}$ 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 $\gamma$-values as large as 450 mJ/molK$^2$ at 1.2 K for UCu$_{2.9}$Al$_{2.1}$. For the Cu-rich compositions, the magnetic susceptibility shows a broad maximum at about 12 K for UCu$_{3.3}$Al$_{1.7}$.

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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Amsterdam High-Field Installation, magnetization measured as a further argument against a spin-glass state. On magnetic anisotropy found in single-crystal studies can be these compounds as indicated from neutron-diffraction results. Complete disorder of Cu and Al atoms was ruled out for ever, the above mechanism cannot be used in order to explain the enhancement of the low-temperature specific heat. How-ever, the above mechanism cannot be used in order to explain the enhancement of the low-temperature specific heat. 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 $M_{ix}/M_{xy}$. In going from UCu$_3$Al$_2$ 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 compounds cannot be decided at present.

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