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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 \( \text{U(Cu, Al)}_{x} \) compounds in the composition range between \( x = 2.9 \) and \( x = 3.5 \), which were found to form in crystal structures related to the \( \text{CaCu}_5 \) 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 \( \text{U(Cu)}_{2.9} \text{Al}_{2.1} \).

The discovery of the heavy-fermion superconductors \( \text{UNi}_{2.1} \text{Al}_3 \) [1] and \( \text{UPd}_2 \text{Al}_3 \) [2] which crystallize in the \( \text{CaCu}_5 \)-derived \( \text{PrNi}_{2.1} \text{Al}_3 \) structure, has turned the attention to other compounds formed in the \( \text{CaCu}_5 \) structure. Among U-compounds with Cu and Al, only \( \text{U(Cu)}_{3.2} \text{Al}_{1.5} \) has been reported up to now to crystallize in this structure [3]. Recently, we have reported on the structural and magnetic properties of \( \text{U(Cu)}_{3.2} \text{Al}_{1.5} \) [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 \( 3g \) 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 \( \text{U(Cu, Al)}_{x} \) 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 \( \text{CaCu}_5 \) structure (Fig. 1). As the crystallographic ordered version of the \( \text{CaCu}_5 \) structure has been found for \( \text{U(Cu)}_3 \text{Al}_1 \), 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 \( \text{UAl}_2 \) for \( \text{U(Cu)}_{2.9} \text{Al}_{2.1} \) and some composition fluctuations for \( \text{U(Cu)}_{3.2} \text{Al}_{1.5} \).

We performed measurements of the specific heat, the magnetic susceptibility and the high-field magnetization on \( \text{U(Cu, Al)}_{x} \) 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 \( \text{U(Cu)}_{3.1} \text{Al}_{1.7} \). This maximum is shifted to about 8 K for \( \text{U(Cu)}_{3.2} \text{Al}_2 \) and appears as a shoulder at about 4 K in \( \text{U(Cu)}_{2.9} \text{Al}_{2.1} \) (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 \( \text{U(Cu)}_{3.4} \text{Al}_{1.8} \) and \( \text{U(Cu)}_{3.5} \text{Al}_{1.5} \). 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 \( \text{U(Cu)}_{2.9} \text{Al}_{2.1} \). For the Cu-rich compounds...
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 UCuxAl\(_y\) 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 \([8]\). 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 U(Cu, Al)\(_3\) compounds discussed here as well as in other U ternaries containing Cu, e.g., U(Cu, Ga)\(_3\) \([8]\) and U(Cu, Al)\(_{12}\) \([9]\), may indicate that some other mechanism is responsible for the common enhancement of \( c_p/T \) 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 \( \theta_p \) between \(-100 \) K (for UCux2.9Al\(_{1.1}\)) and \(-150 \) K (for UCux3.5Al\(_{1.3}\)) and effective moments \( \mu_{\text{eff}} \) 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 relatively broad metamagnetic transitions (similar to the one shown in Ref. \([4]\)) starting at fields of about 8, 15 and 25 T for UCux2.9Al\(_{1.1}\), UCux3Al\(_2\), and UCux3.5Al\(_{1.3}\), respectively. All U(Cu, Al)\(_3\) compounds investigated display a lack of saturation in magnetic fields up to 35 T. Therefore, the magnetization measurements on UCux3Al\(_2\) and UCux3.5Al\(_{1.3}\) 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 U(Cu, Al)\(_3\), which is indicated by the ratio of \( M_{\text{ix}}/M_{\text{fac}} \). In going from UCu3Al\(_2\) to UCu3.5Al1.9 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)\(_3\) compounds. Anomalies in the bulk measurements presented indicate a possible antiferromagnetic ground state for the crystallographically ordered compounds. For all U(Cu, Al)\(_3\) 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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