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

Nakotte, H.; Buschow, K.H.J.; Klaasse, J.C.P.; Prokes, K.; de Boer, F.R.; Andreev, A.V.; Sugiyama, K.; Kuroda, T.; Date, M.

Published in:
Journal of Magnetism and Magnetic Materials

DOI:
10.1016/0304-8853(94)00988-0

Citation for published version (APA):

General rights
It is not permitted to download or to forward/distribute the text or part of it without the consent of the author(s) and/or copyright holder(s), other than for strictly personal, individual use, unless the work is under an open content license (like Creative Commons).

Disclaimer/Complaints regulations
If you believe that digital publication of certain material infringes any of your rights or (privacy) interests, please let the Library know, stating your reasons. In case of a legitimate complaint, the Library will make the material inaccessible and/or remove it from the website. Please Ask the Library: http://uba.uva.nl/en/contact, or a letter to: Library of the University of Amsterdam, Secretariat, Singel 425, 1012 WP Amsterdam, The Netherlands. You will be contacted as soon as possible.
Possible heavy-fermion behaviour of new U(Cu, Al)\textsubscript{5} compounds

H. Nakotte\textsuperscript{a,1}, K.H.J. Buschow\textsuperscript{a}, J.C.P. Klaasse\textsuperscript{a}, K. Prokeš\textsuperscript{a,*}, F.R. de Boer\textsuperscript{a}, A.V. Andreev\textsuperscript{b}, K. Sugiyama\textsuperscript{c}, T. Kuroda\textsuperscript{c}, M. Date\textsuperscript{c}

\textsuperscript{a} Van der Waals–Zeeman Laboratory, University of Amsterdam, 1018 XE Amsterdam, Netherlands
\textsuperscript{b} Ural State University, 620083 Ekaterinburg, Russia
\textsuperscript{c} Department of Physics, Osaka University, Osaka 560, Japan

Abstract

We have synthesized several new U(Cu, Al)\textsubscript{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 CaCu\textsubscript{5} structure. Specific-heat measurements reveal a considerable enhancement of the low-temperature specific-heat coefficient \(\gamma\) for all U(Cu, Al)\textsubscript{5} compounds investigated, with a maximum value of 450 mJ/molK\textsuperscript{2} at 1.2 K for UCu\textsubscript{2.9}Al\textsubscript{2.1}.

The discovery of the heavy-fermion superconductors UN\textsubscript{1.5}Al\textsubscript{3.5} [1] and UPd\textsubscript{2}Al\textsubscript{3} [2] which crystallize in the CaCu\textsubscript{5}-derived PrNi\textsubscript{2}Al\textsubscript{3} structure, has turned the attention to other compounds formed in the CaCu\textsubscript{5} structure. Among U-compounds with Cu and Al, only UCu\textsubscript{3.2}Al\textsubscript{1.8} has been reported up to now to crystallize in this structure [3]. Recently, we have reported on the structural and magnetic properties of UCu\textsubscript{3}Al\textsubscript{2} [4]. Using neutron diffraction, this compound was found to form in an ordered variant of the CaCu\textsubscript{5} structure, where U–Cu\textsubscript{2} layers are separated by layers of Al and the remaining Cu atoms, which are randomly distributed over the 3\textsubscript{g} 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)\textsubscript{5} compounds over a more extended composition range.

Various UCu\textsubscript{x}Al\textsubscript{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\textsubscript{5} structure (Fig. 1). As the crystallographic ordered version of the CaCu\textsubscript{5} structure has been found for UCu\textsubscript{3}Al\textsubscript{2}, a possible random distribution of the Cu and Al atoms over the Cu sites of the CaCu\textsubscript{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\textsubscript{2} for UCu\textsubscript{2.9}Al\textsubscript{2.1} and some composition fluctuations for UCu\textsubscript{3.2}Al\textsubscript{1.8}.

We performed measurements of the specific heat, the magnetic susceptibility and the high-field magnetization on UCu\textsubscript{x}Al\textsubscript{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\textsubscript{3}Al\textsubscript{2}. This maximum is shifted to about 8 K for UCu\textsubscript{3.2}Al\textsubscript{1.8} and appears as a shoulder at about 4 K in UCu\textsubscript{2.9}Al\textsubscript{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\textsubscript{3.4}Al\textsubscript{1.6} and UCu\textsubscript{3.5}Al\textsubscript{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\textsuperscript{2} at 1.2 K for UCu\textsubscript{2.9}Al\textsubscript{2.1}. For the Cu-rich

\begin{figure}
\centering
\includegraphics[width=0.5\textwidth]{fig1.png}
\caption{Variation of lattice parameters \(a\) (circles) and \(c\) (triangles) versus copper content \(x\). Note, two sets of lattice parameters were found for UCu\textsubscript{3.1}Al\textsubscript{1.7} (open and closed symbols), indicating a change from ordered type of structure to a disordered one.}
\end{figure}

\textsuperscript{1} Present address: Los Alamos National Laboratory, LANSCE, Los Alamos, New Mexico 87545, USA.

\textsuperscript{*} Corresponding author. Fax: +31-20-525 5788; email: prokes@phys.uva.nl.
compositions \((x \geq 3.4)\), we observe smaller \(\gamma\)-values (about 290 \text{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 \(T^2\) 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 \(\text{UCu}_3\text{Al}_{5}\) 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 studies. For these compounds, 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 \(\text{UCu}_3\text{Al}_{5}\) compounds discussed here as well as in other \(\text{U}\) ternaries containing Cu, e.g., \(\text{UCu}_3\text{Ga}_{5}\) \([8]\) and \(\text{UCu}_3\text{Al}_{12}\) \([9]\), may indicate that some other mechanism is responsible for the common enhancement of \(c_p/T\) in Cu-containing \(\text{U}\) 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 \(\Theta_c\) between \(-100\) K (for \(\text{UCu}_{2.9}\text{Al}_{2.1}\)) and \(-150\) K (for \(\text{UCu}_{3.5}\text{Al}_{1.9}\)) 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 fixed in random orientations by frozen alcohol, we find a change in the type of magnetic anisotropy for the ordered \(\text{U(Cu, Al)}_5\), which is indicated by the ratio of \(M_{\text{sat}}/M_{\text{free}}\). In going from \(\text{UCu}_3\text{Al}_{12}\) 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 \(\text{UCu}_3\text{Al}_5\) compounds. Anomalies in the bulk measurements presented indicate a possible antiferromagnetic ground state for the crystallographically ordered compounds. For all \(\text{UCu}_3\text{Al}_5\) 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.

Acknowledgement: Work was supported by the ‘Stichting voor Fundamenteel Onderzoek der Materie’ (FOM).

References