Magnetic order in $UCu_{4+x}Al_{8-x}$

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A neutron diffraction study has been performed on $UCu_{4+x}Al_{8-x}$. The compound was chosen as an example of a uranium-based system, which goes from a magnetically ordered state to a pure heavy-fermion state. In the range $x = 0.25$–1, $UCu_{4+x}Al_{8-x}$ orders in a simple collinear antiferromagnetic structure. With increasing concentration of Cu, the ordering temperature decreases and moment compensation develops due to the increasing hybridization of the 5f electrons.

$UCu_{4+x}Al_{8-x}$ crystallizes in the $ThMn_{12}$ structure [1–3] with a homogeneity range from $0.1 \leq x \leq 1.95$ [3]. Magnetic susceptibility, heat capacity and resistivity measurements have been reported previously [3,4] and revealed antiferromagnetic (AF) order for $x \leq 1.25$ and a Fermi liquid state for $x \geq 1.5$. This alloying-induced transition is nicely reflected for example in the heat capacity coefficient γ, going from 100 to 800 mJ/K^2 mol for concentrations $x = 0.1$ and 1.95 respectively.

The samples were arc-melted and subsequently annealed for five days at 750°C. The polycrystalline powder samples, each with a mass of about 20 g, covered the whole rage of composition, i.e. concentrations $x = 0.25$, 0.5, 0.75, 1, 1.25 and 1.9. Preliminary X-ray measurements did not reveal any parasitic phases in the samples. The neutron diffraction experiments were carried out on the multidetector diffractometer D1B at the ILL Grenolble. The incident wavelength, selected by a pyrolytic graphite monochromator, was 2.52 Å. Fig. 1a shows the recorded diffraction pattern for $x = 1$ at $T = 1.6$ K. Fig. 1b displays the magnetic intensities only. They were separated from the nuclear reflections by subtracting the diffraction pattern at $T > T_N$ in the paramagnetic phase from the one recorded at $T < T_N$ in the magnetically ordered phase. Well defined magnetic peaks in between the nuclear reflections pointed towards an AF structure. To analyse the neutron diffraction data in detail, standard Rietveld analyses have been performed for both the nuclear as well as the magnetic structure. The fitted parameters include the lattice constants, temperature factors, atomic coordinates and magnetic moment. All samples investigated revealed the tetragonal $ThMn_{12}$ structure (space group 14/mmm) with two inequivalent Al sites, Wyckoff indices *i* and *j* respectively. As an important structural result, our neutron diffraction data reveal unambiguously, that the excess of Cu is randomly substituted on the *j* sites only. This property is probably just a result of steric constraints. For all compounds with $x \leq 1$, the analyses of the magnetic diffraction pattern results in a simple AF-1 structure with magnetic moments lying along the tetragonal *c* axis. For $x = 1.25$, no magnetic order could be detected in contrast to the transport measurements [3,4]. This

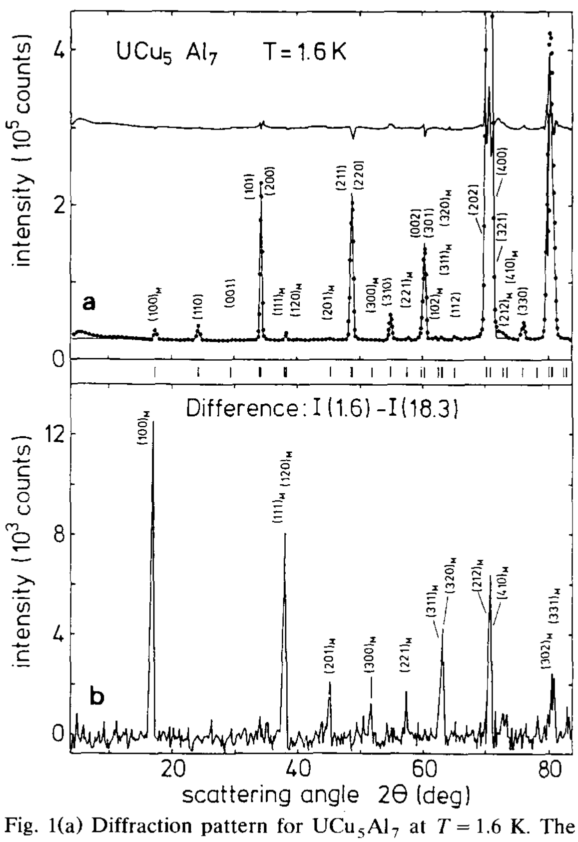

Fig. 1(a) Diffraction pattern for UCu_5Al_7 at $T = 1.6$ K. The solid line through the experimental points is the result of a Rietveld fit to the nuclear and magnetic Bragg reflections. As a measure of the quality of the fit, the difference between calculated and observed intensity is displayed as well (upper line). (b) The magnetic intensities for UCu_5Al_7 resulting from the difference I(1.6 K) − I(18.3 K).

Table 1
Lattice constants a and c, ordered moment μ_S at $T = 1.6$ K and AF ordering temperature T_N

x	a [Å]	b [Å]	μ_S [μ_B]	T_N [K]
0.25	8.7478	5.0955	1.6(1)	37(1)
0.5	8.7290	5.0904	1.65(1)	35(1)
0.75	8.7100	5.0833	1.6(1)	27(1)
1	8.6943	5.0747	1.2(1)	18(2)
1.25	8.6807	5.0595	< 0.25	10(3)
1.5	8.6764	5.0595	–	–
1.9	8.6680	5.0525	–	–

fact can be explained quite easily due to the limited sensitivity of the instrument used. The integrated back-ground intensity across the strongest magnetic Bragg peak has a certain standard deviation due to the statistics. Any magnetic Bragg peak with a lower integrated intensity than this standard deviation of the background cannot be detected. In this way, we get an upper limit of the ordered moment for the $x = 1.25$ compound of $\mu \leq 0.23\mu_B$. The main results are summarised in table 1.

The properties of the heavy-fermion system $UCu_{4+x}Al_{8-x}$ can be interpreted in the framework of Doniach's phase diagram [5]. There is a competition between single-site Kondo-type interaction and inter-site RKKY interaction. Substituting Al by Cu decreases the unit cell resulting in an increase of the 5f conduction electron hybridization, i.e. increase of the Kondo-type interaction. One feature that comfirms this behaviour is a gradual decrease of the transition temperature. For $x > 0.75$, a significant moment compensation sets in, and at about $x = 1.25$ the long-range magnetic order has disappeared. Meanwhile, the specific-heat coefficient γ is raised from 100 mJ/K^2 mol for $x = 0.1$ to 400 mJ/K^2 mol at $x = 1.25$, going further up to 800 mJ/K^2 mol for $x = 1.95$ [3,4]. This transition from a magnetically ordered to a Fermi liquid groundstate is very rare for an actinide-based heavy-fermion system. However, there are some facts that remain to be explained; the ordered moment is always smaller than the effective moment, derived by high-temperature susceptibility measurements [3]. To clarify this behaviour, it is necessary to determine the crystal field level scheme. Therefore, a neutron scattering study employing the time-of-flight technique is now in progress. Additionally, the lattice constants show a significant deviation from linear concentration dependence due to hybridization effects [3,4]. This may find an explanation in the change of the electronic structure of the conduction band due to the replacement of Al by Cu atoms.

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