Neutron diffraction study of the heavy fermion superconductors UM_2Al_3 (M = Pd, Ni)

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An elastic neutron scattering study was performed on the new superconducting heavy fermion systems UPd_2Al_3 and UNi_2Al_3. The neutron diffraction patterns reveal unambiguously long range antiferromagnetic order in UPd_2Al_3 with an ordered magnetic moment $\mu_U = (0.85 \pm 0.03)\,\mu_B$, which coexists with the superconducting state. This is by far the largest μ_U value observed for any heavy fermion superconductor. For UNi_2Al_3, no long-range magnetic order could be observed for temperatures $T \gtrsim 1.5$ K, yielding an upper limit of the ordered moment of $0.2\,\mu_B$.

The discovery of two new heavy fermion superconductors has been reported recently [1, 2]. Bulk measurements revealed transitions into antiferromagnetic states at $T_N = 4.5$ K and $T_N = 14$ K for UNi_2Al_3 and UPd_2Al_3, respectively, followed by transitions to superconducting ground states at $T_c = 1$ K and $T_c = 2$ K, the highest transition temperature among all heavy fermion systems known so far. In this Rapid Note we present the main results of a neutron diffraction study in order to elucidate the magnetic properties of these compounds. Details will be given in a forthcoming publication.

Polycrystalline powder samples with a mass of 20 g each were prepared and characterized as described previously [1, 2]. Part of the UPd_2Al_3 sample was studied by dc-magnetization measurements: The diamagnetic transition was found to be centered at $T_c = 1.8$ K with a width (10–90%) of 0.2 K. The neutron diffraction experiments were carried out on the multidetector powder diffractometer DMC at the 10 MW Saphir reactor of PSI, Villigen, Switzerland, in the high intensity mode using neutrons of wavelength $\lambda = 1.7008$ Å [3]. The diffraction patterns were recorded in the angular range from 0 to 133 degrees with steps $\Delta 2\Theta = 0.1°$. No magnetic intensities could be observed above 60 degrees.

In the following, we concentrate on the results on UPd_2Al_3. In Fig. 1 (lower part) the difference of the intensities $I(1.7\,\text{K}) - I(20\,\text{K})$ is shown, yielding the reflections of magnetic origin only. These can be indexed unambigiously by assuming ferromagnetic sheets with the magnetic moments of uranium lying in the easy $a-b$ plane and being coupled antiferromagnetically along the c-axis. This magnetic order corresponds to a doubling of the chemical unit cell with an ordering wave vector $\mathbf{k} = [0, 0, 0.5]$. Note that the lattice parameter c corresponds to the shortest U-U distance. A standard Rietveld analysis performed for both, the nuclear as well

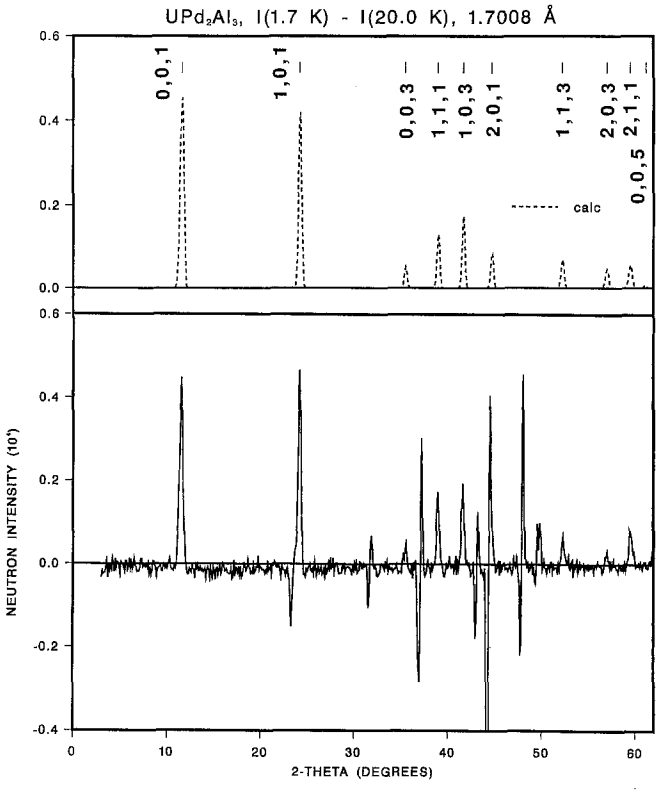

Fig. 1. Upper part: Calculated magnetic intensities of UPd_2Al_3 according to the antiferromagnetic structure shown in Fig. 1. Lower part: measured magnetic intensities corresponding to the difference diagram $I(1.7\,\text{K}) - I(20\,\text{K})$. The magnetic peaks are indexed with respect to the magnetic unit cell

as for the magnetic structure, revealed an ordered magnetic moment of the uranium atom $\mu_U = (0.85 \pm 0.03)\,\mu_B$. These moments are oriented perpendicular to the c-axis, in agreement with the results of susceptibility measurements on single crystals, reported recently [5]. The calculated intensities are shown in the upper part of Fig. 1. A comparison with the measured intensities in the lower part of the figure displays nicely the quality of our Rietveld analysis. The R-value for the magnetic structure was 15.1%.

The observed nuclear intensities were found to deviate by about 15% from those calculated for the hexagonal $PrNi_2Al_3$ type structure (space group P6/mmm) as reported in the literature and confirmed by X-ray diffraction [1, 2, 6]. A possible explanation lies in a preferred orientation of crystallites, which is a well known problem in this class of compounds and has already been inferred from susceptibility and dilatation measurements on UPd_2Al_3 [2, 5].

In addition, the sample was investigated on the two-axis spectrometer P2AX at the Saphir reactor. Using a neutron wavelength $\lambda = 2.337$ Å, we followed the temperature dependence of the first magnetic Bragg peak down to $T = 1.5$ K. The magnetic intensity of this (001) reflection is almost constant for $T \leqq 5$ K (Fig. 2). No changes could be detected on crossing the normal-to-superconducting phase boundary. With increasing temperature, a transition from the simple antiferromagnetic to an incommensurate (IC) structure was detected near 15 K. The IC structure with a propagation vector $\mathbf{k} = [0, 0, 0.521 \pm 0.002]$ was confirmed by measuring four additional magnetic reflections. With increasing temperature, the IC structure is stable for temperatures $15\,\text{K} \leqq T \leqq 20\,\text{K}$ and coexists with the commensurate structure in a narrow temperature regime. The temperature dependence of the first magnetic Bragg reflection and its satellites, monitored upon heating, defines the corresponding phase boundaries, as shown in Fig. 2. Further experiments are underway to clarify, why the IC phase appears almost suppressed on cooling and why T_N is shifted by about 1 K compared to bulk measurements [2, 5]. Another interesting open question concerns the absence of well-defined frequencies in μ^+SR for $T < T_N$, suggesting either that all μ^+ stopping sites are symmetric or that the magnetic structure is an incommensurate one [7].

For the UNi_2Al_3 compound, no long-range antiferromagnetic order could be observed which gives an upper limit for the corresponding ordered moment of approximately $0.2\,\mu_B$. This is consistent with μSR-measurements, which revealed an ordered antiferromagnetic moment of about $0.1\,\mu_B$ [7], well below the sensitivity of this powder-diffraction experiment. A Rietveld analysis of the nuclear Bragg peaks again hints at some preferred orientation of crystallites in the polycrystalline samples as discussed before for UPd_2Al_3.

We conclude by emphasizing the large ordered moment in UPd_2Al_3 of order $1\,\mu_B$/U-atom which exceeds that in other U-based heavy fermion superconductors, e.g. URu_2Si_2 [8], by orders of magnitude. Most remarkably, this large μ_U value was detected down to $T = 1.5$ K, i.e. below $T_c = 1.8$ K. Thus, UPd_2Al_3 is the first compound in which heavy fermion superconductivity coexists with antiferromagnetic ordering between local U-derived moments of ordinary size.

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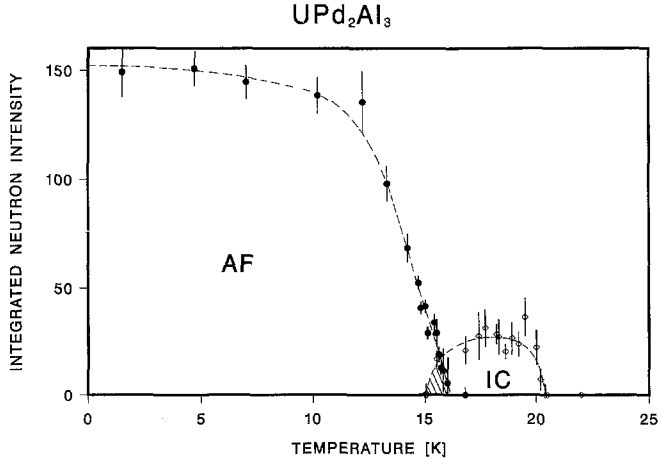

Fig. 2. Temperature dependence of the integrated magnetic intensities of UPd_2Al_3 for increasing temperature. AF: antiferromagnetic order with $\mathbf{k} = [0, 0, 0.5]$; IC: incommensurate structure with $\mathbf{k} = [0, 0, 0.521]$; shaded area: coexistence region. The dashed lines are guides to the eye

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