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Letter to the Editor

High-field magnetization of heavy-fermion UPd_2Al_3

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We report on high-field magnetization ($M(B)$) measurements ($B \leq 35$ T) of antiferromagnetic heavy-fermion UPd_2Al_3 at liquid helium temperatures. The data taken on well-characterized single-crystalline samples reveal a large anisotropy. For a field directed in the hexagonal plane ($B \| a$-axis) a sharp jump, $\Delta M = 0.94 \mu_B$/U-atom, occurs at $B_c = 18.0$ T, while for a field along the hexagonal axis (hard axis) the magnetization is small and almost linear up to 35 T. The jump at $B_c = 18.0$ T for $B \| a$ reflects the antiferromagnetic phase boundary.

1. Introduction

Recently, the select class of heavy-fermion compounds that undergo both an antiferromagnetic and a superconducting phase transition has been extended by two new members: UNi_2Al_3 ($T_N = 4.6$ K, $T_c = 1$ K) [1] and UPd_2Al_3 ($T_N = 14$ K, $T_c = 2$ K) [2]. For both compounds heavy-fermion superconductivity is evidenced by the fairly large jumps in the specific heat at T_c. The analysis of the upper critical field (B_{c2}) [1, 2], employing the moderately enhanced linear coefficient of the (normal-state) specific heat ($\gamma = 120$ mJ/mol K^2 for UNi_2Al_3 and $\gamma = 150$ mJ/mol K^2 for UPd_2Al_3), yields in both cases an effective mass (m^*) of about 70 times the free electron mass. These ternary uranium compounds crystallize in the hexagonal $PrNi_2Al_3$ structure. As, in general, the electronic and magnetic parameters of (non-cubic) heavy-fermion compounds are strongly anisotropic, experiments should preferably be performed on single crystals. Recently, we succeeded in preparing single-crystalline samples of UPd_2Al_3 and we here report on our first study of the antiferromagnetic state.

The antiferromagnetic phase transition of UPd_2Al_3 at a Néel temperature $T_N = 14$ K is evidenced by a pronounced λ-type anomaly in the specific heat (c), a kink in the magnetic susceptibility (χ) and a kink in the electrical resistivity (ρ) [2]. The kink in $\rho(T)$ at T_N suggests a classical antiferromagnetic ordering, in contrast to magnetic ordering in the heavy-fermion compounds URu_2Si_2 ($T_N = 17.5$ K) [3] and $U(Pt_{0.95}Pd_{0.05})_3$ ($T_N = 5.8$ K) [4], where an increase in ρ below T_N signals the presence of a spin-density wave. A neutron diffraction study on polycrystalline material revealed an ordered uranium moment of $(0.85 \pm 0.03)\mu_B$ [5]. The ordering consists of ferromagnetic sheets in the basal plane that are coupled antiferromagnetically along the hexagonal axis (c-axis), i.e. a doubling of the nuclear unit cell with an ordering vector $\boldsymbol{k} = [0, 0, 0.5]$. Recent μSR experiments down to 50 mK reveal that antiferromagnetic order and superconductivity coexist [6].

In this paper we report on magnetization mea-

surements on single-crystalline UPd_2Al_3 at liquid helium temperatures, i.e. at temperatures well below T_N. As the Néel temperature is rather large one expects to need a rather large magnetic field in order to suppress the long-range antiferromagnetic order. Therefore, experiments up to $B = 35$ T have been performed in the High Magnetic Field Facility of the University of Amsterdam. Furthermore, we report on specific heat, susceptibility and resistivity data in order to characterize our single-crystalline samples.

2. Experimental

Polycrystalline UPd_2Al_3 was prepared by arc-melting together the metallic components (depleted U (2N5), Pd(5N) Johnson Matthey, Al (6N) Johnson Matthey) in a stoichiometric composition in a water-cooled copper crucible under a continuously titanium gettered argon atmosphere. A single-crystalline rod was prepared in a tri-arc furnace also in a titanium gettered argon atmosphere using the Czochralski technique. Microprobe measurements on several parts of the crystal showed the correct homogeneous chemical composition UPd_2Al_3 within the experimental accuracy. The lattice parameters of the as-grown crystal, determined by X-ray analysis, amount to 5.365 Å (a-axis) and to 4.191 Å (c-axis), in good agreement with previous results [2]. The single-crystalline rod (wrapped in tantalum) was annealed in a quartz tube under a protective argon atmosphere at a temperature of 900°C for 7 days, leaving the lattice parameters essentially unchanged. Specific heat measurements were performed on the bulk part of the rod. Subsequently, several specimens, with well-defined a- and c-axes, were cut from the rod, by means of spark erosion, for electrical resistivity, magnetic susceptibility and high-field magnetization experiments.

Resistivity measurements yield a residual resistance of 10 and 23 μΩcm for a current along the a- and c-axis, respectively. The residual resistance ratios, $R(300\,\mathrm{K})/R(0\,\mathrm{K})$, equal 18 ($a$-axis) and 8 ($c$-axis). The superconducting transition temperature varies from 1.3 up to 1.75 K for different parts of the crystal. A variation of T_c has also been observed in single crystals grown elsewhere [7] and is probably related with subtle changes in the aluminium concentration. The Néel temperature determined from the resistivity equals 14.4 ± 0.1 K.

The magnetization measurements were performed on two cylindrical samples (diameter 3 mm, length 4 mm) that were cut along the a- and c-axis. The magnetization was measured by integrating the signal from a pick-up coil in which the samples were positioned. Long pulsed magnetic fields (up to 35 T) were produced by the High Magnetic Field Facility of the University of Amsterdam [8]. The pulse duration amounts up to 1 s. Different pulse shapes can be chosen, e.g., step-like pulses (with constant magnetic fields for about 100 ms) and smooth pulses. In order to minimize eddy current heating, the samples are immersed directly in the liquid helium.

Magnetic susceptibility and specific-heat measurements were performed at the Kamerlingh Onnes Laboratory of the University of Leiden using standard techniques.

3. Results

Magnetic susceptibility data taken for a field ($B = 2$ T) along the a- and c-axis reveal that the basal plane is the easy direction for magnetization (fig. 1). The data in fig. 1 are in good agreement with the results reported in ref. [9]. A Curie–Weiss behaviour with an effective moment of $3.6\mu_B$/U-atom ($B \| a$ and $B \| c$) is observed only in a limited temperature range ($T > 150$ K). The paramagnetic Curie temperature (Θ_p) equals -33 K for the a-axis and -215 K for the c-axis. At lower temperatures for $B \| a$ a broad maximum is centered at 40 K, while a kink at 14.7 ± 0.1 K reflects the Néel temperature.

The specific heat, measured with an adiabatic method in the temperature interval $1.5\,\mathrm{K} < T < 30\,\mathrm{K}$, reveals a large λ-type anomaly at T_N in agreement with the results reported in ref. [2] (see fig. 2). Below 10 K the specific heat can satisfactorily be described by a linear and a cubic

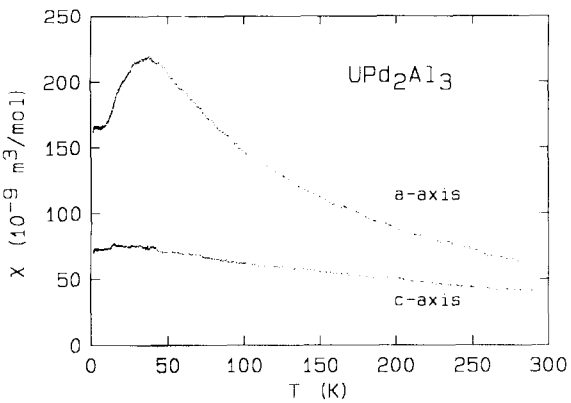

Fig. 1. Magnetic susceptibility of UPd_2Al_3 for a field (B = 2 T) along the a- and c-axis as indicated. The Néel temperature is reflected by a kink in $\chi(T)$ at 14.7 K for the a-axis.

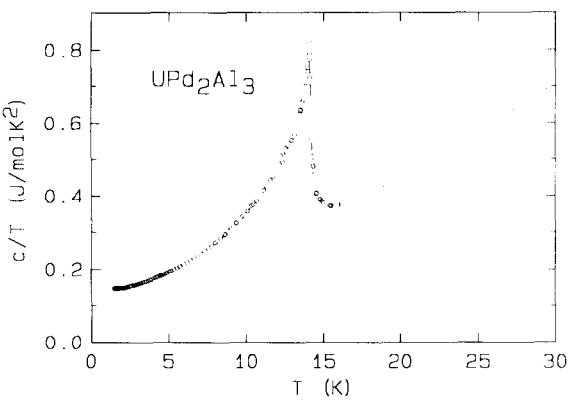

Fig. 2. Specific heat of single-crystalline UPd_2Al_3 in a plot of c/T versus T for $T < 30$ K. The antiferromagnetic transition (T_N = 14.1 K) appears as a λ-type anomaly. The solid line is a guide to the eye.

term: $c = \gamma T + \beta T^3$ with $\gamma = 150$ mJ/mol K^2 and $\beta = 1.53$ mJ/mol K^4. The cubic term is probably dominated by a contribution from magnons. The entropy associated with the antiferromagnetic phase transition amounts to $0.62R \ln 2$, indicating reduced ordered moments.

A more detailed account of the resistivity, susceptibility and specific-heat measurements will be published elsewhere [10].

High-field magnetization experiments have been performed at 4.2 K for a field directed along both the a- and c-axis, while at 1.3 K data have only been taken for $B \| a$. The measurements have been performed on the same samples as used for the susceptibility measurements. The results are shown in fig. 3. At 4.2 K the magnetization for $B \| c$ is close to linear, while for $B \| a$ the magnetization increases faster than linear and shows a very sharp jump at $B_c \sim 18.0$ T. For an accurate determination of B_c the step-like pulses have been complemented by a smooth pulse. In this symmetric smooth pulse the field was first raised up to 21.3 T (23.3 T at T = 1.3 K) at a constant rate of 40 T/s, and then diminished to zero at the same rate. A small hysteresis was observed: with decreasing field the transition takes place at 17.9 T. The width of the transition (ΔB) is 0.2 T. Data taken at 1.3 K showed essentially the same result, although the jump at B_c is

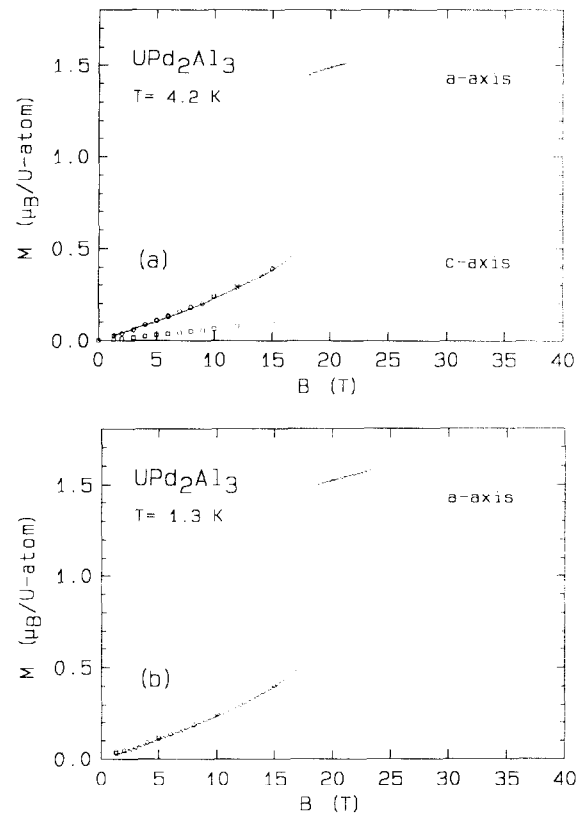

Fig. 3. High-field magnetization of UPd_2Al_3 along the a- and c-axis at a temperature of (a) 4.2 K and (b) 1.3 K (only a-axis data). The open circles and squares are taken from step pulses. The dots from smooth pulses (only increasing field data). The transition at B_c = 18.0 T reflects the antiferromagnetic phase boundary.

2% larger. Above the transition the moment tends to saturate. In a field of 35 T a moment of $\sim 1.7\mu_B$/U-atom results.

4. Discussion

The most striking result of the high-field experiments is the almost step-like increase in the magnetization occurring at $B_c = 18.0$ T for $B \| a$. The size of the jump amounts to $\Delta M = 0.92\mu_B$/*U*-atom at 4.2 K ($0.92\mu_B$/U-atom at 1.3 K), which is almost equal to the size of the ordered moment ($0.85\mu_B$/U-atom). Therefore, we believe that the anomaly at B_c reflects the antiferromagnetic phase boundary. However, a study of the temperature variation of B_c over a wider temperature range is necessary to confirm this. The magnitude of the magnetic moment induced in a field of 35 T ($|\boldsymbol{\mu}| = 1.7\mu_B$/U-atom) is already quite large for uranium compounds, which indicates that the magnetization process is of a single-step type. The neutron-diffraction studies [5] revealed that the ordered moments lie in the *ab*-planes, which is supported by the high-field magnetization data of fig. 3. The magneto-crystalline in-plane anisotropy has not been investigated in detail yet, but is probably small [7], suggesting that the transition is not of the simple spin-flop or spin-flip type as might occur in the case of a simple two-sublattice antiferromagnet with in-plane anisotropy.

The high-field transition in UPd_2Al_3 has a quite different nature than the metamagnetic(-like) transitions observed in other uranium heavy-fermion compounds. In UPt_3 [11] and in $U(Pt_{0.95}Pd_{0.05})_3$ [12] a rather broad increase in the magnetization is found at liquid helium temperatures at a field of ~20 and ~12 T, respectively, also directed in the hexagonal plane. For both alloys $\Delta M \simeq 0.5\mu_B$/U-atom and the moment in a field of 35 T amounts to $\sim 1\mu_B$/U-atom. The value for ΔM is comparable to the size of the antiferromagnetically ordered moment observed by neutron diffraction [13] for the 5% Pd compound ($T_N = 5.8$ K), indicating that a moment of this size can be stabilized either by alloying with Pd or by applying strong magnetic fields. The metamagnetic-like transition in the $U(Pt, Pd)_3$ series is primarily attributed to a quenching of antiferromagnetic inter-site fluctuations. In URu_2Si_2 a three-step magnetization process is observed at $T = 1.5$ K for fields in the range 35–40 T directed along the tetragonal *c*-axis [14, 15]. The total increase of the magnetization at the three jumps corresponds to $1\mu_B$/U-atom and in a field of 41 T a moment of $1.4\mu_B$/U-atom results. This high-field magnetization process is one of the puzzling magnetic properties of URu_2Si_2. The antiferromagnetically ordered moment ($T_N = 17.5$ K) was found to be extremely small ($(0.03 \pm 0.01)\mu_B$/U-atom) [16]. Apparently, first a destruction of the heavy-fermion state with field (i.e. a reduction of the Kondo screening) has to take place before transitions between different magnetic structures can occur.

Until now little information is available about the crystalline electric field (CEF) parameters of UPd_2Al_3. The analysis of the f-electron specific heat ($T < 150$ K), which was obtained after subtracting the lattice contribution measured on $ThPd_2Al_3$, could not resolve unambiguously the CEF-level scheme [17]. Moreover, the high-temperature effective moment ($3.6\mu_B$/U-atom) as determined from the susceptibility data cannot discern between a uranium f^2 ($3.58\mu_B$/U-atom) or f^3 ($3.62\mu_B$/U-atom) configuration. Besides, the Curie–Weiss law holds only in a very limited temperature range. The influence of the crystalline electric field on the magnetic properties of URu_2Si_2 and UPt_2Si_2 was investigated by Nieuwenhuys [18] in a mean-field model assuming a $5f^2$ configuration with a singlet ground state and a singlet first excited level. In the case of UPt_2Si_2 the magnetic properties, including the high-field magnetization curves, can reasonably be fitted with such a model [18, 19], while the model appears to be only qualitatively successful for URu_2Si_2 [18]. Similar calculations for UPd_2Al_3 will be carried out in the near future [10].

In summary, we have detected the antiferromagnetic phase boundary of UPd_2Al_3 at $B_c = 18.0$ T at liquid helium temperatures for a field along the *a*-axis in the hexagonal plane. The

jump in the magnetization at B_c is almost equal to the size of the ordered moment.

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