Physica B 186–188 (1993) 291–293
North-Holland

PHYSICA B

High-field magnetoresistance of heavy-fermion UPd_2Al_3

A. de Visser[a], K. Bakker[a], L.T. Tai[a], A.A. Menovsky[a], S.A.M. Mentink[b], G.J. Nieuwenhuys[b] and J.A. Mydosh[b]

[a]*Van der Waals–Zeeman Laboratorium, Universiteit van Amsterdam, The Netherlands*
[b]*Kamerlingh Onnes Laboratorium, Rijksuniversiteit Leiden, The Netherlands*

We report on high-field ($B < 38$ T) magnetoresistance experiments on a single-crystalline sample of the heavy-fermion antiferromagnet UPd_2Al_3 ($T_N = 14$ K) at liquid-helium temperatures. The antiferromagnetic phase boundary, previously detected by magnetization measurements at $B_c = 18$ T for a field in the basal plane ($T = 1.3$ K), is reflected as a sharp peak in the longitudinal ($I \| a$) and as a precipitous drop in the transverse ($I \| c$) magnetoresistance.

1. Introduction

Recently, the study of the interplay of antiferromagnetism and superconductivity in heavy-fermion systems received a considerable impetus from the discovery of the antiferromagnetic superconductors UPd_2Al_3 ($T_N = 14$ K, $T_c = 2$ K) [1] and UNi_2Al_3 ($T_N = 4.6$ K, $T_c = 1$ K) [2]. The formation of heavy quasiparticles in both compounds is evidenced by the moderately enhanced linear coefficient of the normal-state specific heat ($\gamma = 150$ mJ/mol K^2 for UPd_2Al_3 [1] and $\gamma = 120$ mJ/mol K^2 for UNi_2Al_3 [2]). The heavy quasiparticles take part in the superconducting condensate as follows from the large jumps in the specific heat at T_c. The analysis of the upper critical field (B_{c2}) yields in both cases a quasiparticle mass (m^*) of ~70 times the free electron mass [1,2]. Since both uranium compounds crystallize in the hexagonal $PrNi_2Al_3$ structure, strongly anisotropic electronic and magnetic parameters are expected. Therefore, experiments should preferably be performed on single-crystalline samples. Recently, we succeeded in preparing UPd_2Al_3 in a single-crystalline form [3] and in the following we report on a 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 $c(T)$ [1,3], a kink in the magnetic susceptibility $\chi(T)$ [1,3], and a kink in the electrical resistivity $\rho(T)$ [1]. Neutron diffraction experiments on polycrystalline material [4] revealed a magnetic structure consisting of ferromagnetic sheets that are coupled antiferromagnetically along the hexagonal axis (c-axis), i.e. a doubling of the nuclear unit cell with an ordering vector $\boldsymbol{k} = [0, 0, \frac{1}{2}]$. The ordered uranium moment amounts to $0.85 \pm 0.03 \mu_B$/U-atom. Recent μSR experiments down to 50 mK revealed that antiferromagnetic order and superconductivity coexist [5].

Magnetic susceptibility data taken on single-crystalline samples revealed that the magnetic properties are strongly anistropic [3,6]. The basal plane is the easy direction for magnetization. For $B \| a$ deviations from a Curie–Weiss behaviour ($\mu_{eff} = 3.6 \mu_B$/U atom) appear below 150 K, leading to a broad maximum centered at 40 K, while a kink is found at $T_N = 14$ K. For $B \| c$ $\chi(T)$ is only weakly temperature dependent. The magnetic anisotropy is also reflected in the high-field magnetization data $M(B)$ (see fig. 1) [3]. At

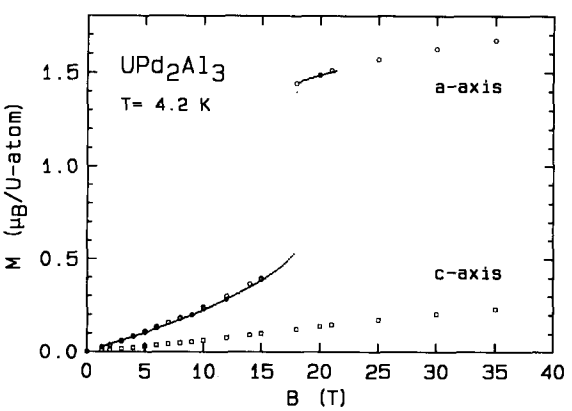

Fig. 1. Magnetization of UPd_2Al_3 along the a- and c-axis at a temperature of 4.2 K. The open circles and squares are taken from step pulses. The dots are taken from smooth pulses (only increasing field data).

Correspondence to: A. de Visser, Van der Waals–Zeeman Laboratorium, Universiteit van Amsterdam, Valckenierstraat 65, 1018 XE Amsterdam, The Netherlands.

4.2 K, $M(B)$ is close to linear for $B \| c$, while for $B \| a$ the magnetization increases faster than linearly and shows a sharp jump at $B_c = 18.0$ T. The size of the jump amounts to $\Delta M = 0.92 \mu_B$/U-atom). Therefore, we suggested [3] that the anomaly at B_c reflects the antiferromagnetic phase boundary.

In this paper we report on an investigation of the magnetoresistance at crossing the antiferromagnetic phase boundary. The experiments were performed at $T = 1.4$ and 4.2 K, for various orientations of the magnetic field with respect to the crystal axes and current, in order to elucidate the magnetic anisotropy.

2. Experimental

A single-crystalline rod of UPd_2Al_3 was prepared in a titanium-gettered argon atmosphere using the Czochralski technique and annealed for 7 days at 900°C. Specific heat measurements on the annealed sample yield a sharp anomaly at $T_N = 14.1$ K [3]. Specimens for the (magneto)resistance experiments were cut from the single-crystalline batch by means of spark erosion. The dimensions of the specimens amount to $1 \times 1.25 \times 2.5$ mm^3, with the long direction either along the a- or c-axis. Resistivity measurements yield residual resistances of 9.8 and 22.8 μΩ cm for currents along the a- and c-axes, respectively. The residual resistance ratios, $R(300\,\text{K})/R(0\,\text{K})$, equal 18 ($a$-axis) and 8 ($c$-axis).

The magnetoresistance experiments were performed in the High Magnetic Field Installation of the University of Amsterdam that produces pulsed magnetic fields up to 40 T with a total pulse duration of 1 s [7]. Different pulse shapes can be chosen as the field profile can be programmed. The magnetoresistance was measured with a standard DC four-probe method ($I = 0.4$ A). In order to minimize eddy current heating the samples were immersed in the liquid helium. The magnetic field was oriented with respect to the crystal axes with an accuracy of a few degrees.

3. Results

The longitudinal magnetoresistance of UPd_2Al_3 for both the current and field direction along the a-axis is shown in fig. 2 in a plot of $\Delta\rho/\rho = (\rho(B) - \rho(0))/\rho(0)$ versus B. The values for $\rho(0)$ amount to 10.0 and 12.6 μΩ cm at 1.4 and 4.2 K, respectively, where $\rho(0)$ at 1.4 K is given by the normal-state resistivity. The transverse magnetoresistance for a current along the c-axis and a field along the b-axis is shown in fig. 3, where $\rho(0)$ amounts to 23.0 and 26.8 μΩ cm at 1.4 and

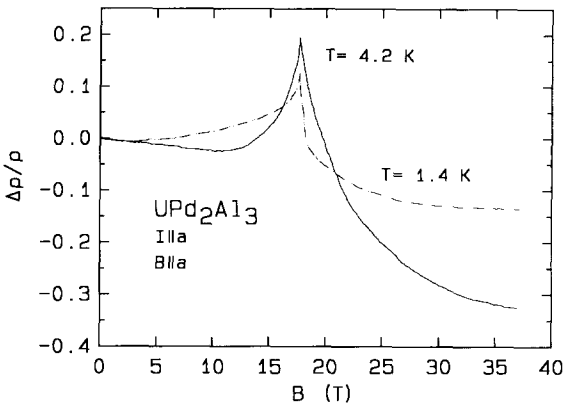

Fig. 2. Longitudinal magnetoresistance of UPd_2Al_3 ($B \| I \| a$) at temperatures indicated.

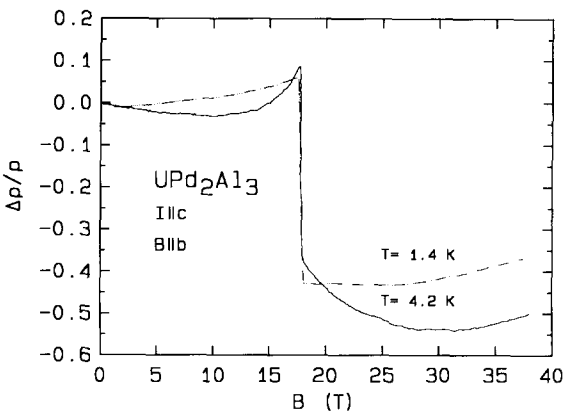

Fig. 3. Transverse magnetoresistance of UPd_2Al_3 ($B \| b$, $I \| c$) at temperatures indicated.

4.2 K, respectively. The data shown in figs. 2 and 3 were taken with so-called smooth pulses [7]. Data acquisition was done at decreasing field. The anomaly at B_c appears as a sharp maximum at 17.8 T for $B \| I \| a$, while it appears as a precipitous drop (a 50% decrease) at 17.6 T for $B \| b$ and $I \| c$. On decreasing the temperature from 4.2 to 1.4 K, B_c remains constant, but the transitions become somewhat sharper. The origin of the small rise of $\Delta\rho$ in the high-field limit for the transverse configuration is not clear, but is probably an artefact of the experiment.

4. Discussion

Comparing the data in figs. 2 and 3 we observe that for both field directions in the basal plane ($a \perp b$) the transition occurs at almost the same field. This indicates that the basal-plane anisotropy is negligible or at

least very small. The value for $B_c = 17.6$–17.8 T deduced from figs. 2 and 3 agrees well with the value deduced from the magnetization measurements (B_c = 17.9 T with decreasing field). Our high-field data indicate that the magnetization process is rather complex, although it is of the single-step type. The large jump in the magnetization at B_c, with a size equal to the ordered moment, suggests that a spin-flip takes place. However, the absence of a marked basal-plane anisotropy implies that at low fields the sublattice magnetization is free to rotate towards a direction perpendicular to the field. Therefore one expects (in a simple two-sublattice picture) that the alignment of the sublattice magnetizations with increasing field would occur gradually, which is contradicted by the experiment. However, because UPd_2Al_3 is a heavy-fermion compound, the Kondo effect might complicate the magnetization process and transitions between different magnetic structures might only take place after the suppression of the Kondo screening in field.

The observed anisotropy in the magnetoresistance reflects the magnetic structure. For $I \| c$ the large drop in $\Delta\rho$ at B_c is consistent with the return to nuclear periodicity, while for $I \| a$ the (broad) anomaly indicates the presence of magnetic fluctuations in the hexagonal plane.

Recently, additional anomalies observed in the magnetization, magnetoresistivity and magnetostriction [8] were taken as evidence that UPd_2Al_3 exhibits a magnetic phase diagram with three different antiferromagnetic phases for $B \perp c$, with critical fields (for $T \rightarrow 0$) of 0.6, 4.2 and 18 T. We cannot comment on the low-field phase because our measurements were only performed for fields above 1 T. We did not find indications of the intermediate phase from our magnetoresistance data. In ref. [8] the phase boundary at 4.2 T ($T \rightarrow 0$) was observed as a pronounced maximum in the transverse magnetoresistance (both I and B in the basal plane). Instead our transverse magnetoresistance data show a negative contribution at low fields (fig. 3), which might possibly be explained by the different I–B configurations. Magnetostriction experiments [8] demonstrated that the phase boundary at 4.2 T is accompanied by an unusually large hysteresis. We therefore propose that the phase boundary at 4.2 T reported in ref. [8] is not between different antiferromagnetic structures but originates from the re-orientation of antiferromagnetic domains. Neutron diffraction experiments in field would be highly welcome to elucidate this point.

The research of one of us (AdV) was made possible by a fellowship of the Royal Netherlands Academy of Arts and Sciences. Part of this work has been performed within the research program of the Amsterdam–Leiden Materials Research Cooperation (FOM–ALMOS).

References

[1] C. Geibel, C. Schank, S. Thies, H. Kitazawa, C.D. Bredl, A. Böhm, M. Rau, A. Grauel, R. Helfrich, U. Ahlheim, G. Weber and F. Steglich, Z. Phys. B 85 (1991) 1.

[2] C. Geibel, S. Thies, D. Kaczorowski, A. Mehner, A. Grauel, B. Seidel, U. Ahlheim, R. Helfrich, K. Petersen, C.D. Bredl and F. Steglich, Z. Phys. B 83 (1991) 305.

[3] A. de Visser, H. Nakotte, L.T. Tai, A.A. Menovsky, S.A.M. Mentink, G.J. Nieuwenhuys and J.A. Mydosh, Physica B 179 (1992) 84.

[4] A. Krimmel, P. Fischer, B. Roessli, H. Maletta, C. Giebel, C. Schank, A. Grauel, A. Loidl and F. Steglich, Z. Phys. B 86 (1992) 161.

[5] A. Amato, R. Feyerherm, F.N. Gygax, A. Schenck, M. Weber, R. Caspary, P. Hellmann, C. Schank, C. Geibel, F. Stehlich, D.E. MacLaughlin, E.A. Knetsch and R.H. Heffner, Europhys. Lett. 19 (1992) 127.

[6] C. Geibel, U. Ahlheim, C.D. Bredl, J. Diehl, A. Grauel, R. Helfrich, H. Kitazawa, R. Köhler, R. Modler, M. Lang, C. Schank, S. Thies, F. Steglich, N. Sato and T. Komatsubara, Physica C 185–189 (1991) 2651.

[7] R. Gersdorf, F.R. de Boer, J.C. Wolfrat, F.A. Muller and L.W. Roeland, in: High Field Magnetism, ed. M. Date (North-Holland, Amsterdam, 1983) p. 277.

[8] A. Grauel, A. Böhm, H. Fischer, C. Geibel, R. Köhler, T. Komatsubara, R. Modler, N. Sato, C. Schank, F. Steglich and G. Weber, Phys. Rev. B 46 (1992) 5818.