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Publication date
1994

Published in
Physica B-Condensed Matter

[Link to publication](#)

Citation for published version (APA):

Mentink, S. A. M., Nieuwenhuys, G. J., Menovsky, A. A., Mydosh, J. A., Drost, A., Frikkee, E., Bando, Y., Takabatake, T., Boni, P., Fischer, P., Furrer, A., Amato, A., & Schenck, A. (1994). Crystal symmetry and electronic properties of heavy-fermion MPd₂Al₃ (M=Ce, U). *Physica B-Condensed Matter*, 199&200, 143-144.

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Crystal symmetry and electronic properties of heavy-fermion MPd_2Al_3 ($\text{M} = \text{Ce}, \text{U}$).

S.A.M. Mentink^{a,*}, G.J. Nieuwenhuys^a, A.A. Menovsky^a, J.A. Mydosh^a, A. Drost^b,
E. Frikkee^b, Y. Bando^c, T. Takabatake^c, P. Böni^d, P. Fischer^d, A. Furrer^d,
A. Amato^e, A. Schenck^e

^aKamerlingh Onnes Laboratory, Leiden University, 2300 RA Leiden, The Netherlands

^bDutch Energy Research Foundation ECN, 1755 ZG Petten, The Netherlands

^cFIAS, Hiroshima University, Higashi-Hiroshima 724, Japan

^dLaboratory for Neutron Scattering, ETH&PSI, CH-5232 Villigen PSI, Switzerland

^eInstitut für Mittelenergiephysik der ETH-Zürich, CH-5232 Villigen PSI, Switzerland

Abstract

We have determined the Kondo temperature and crystal-field splitting of the new heavy-fermion compound CePd_2Al_3 by thermopower and inelastic neutron scattering. The absence of static, magnetically ordering cerium moments in single crystals is demonstrated by zero-field μ^+ SR. Small deviations of aluminum stoichiometry yield significant variations of T_N of CePd_2Al_3 and T_c of UPd_2Al_3 in different samples.

Soon after the discovery of the new heavy-fermion superconductor UPd_2Al_3 [1], its cerium-based counterpart CePd_2Al_3 was shown to be a heavy-electron antiferromagnet [2, 3]. We estimated the relevant energy scales, viz. the Néel temperature $T_N = 2.7$ K, Kondo temperature $T_K = 19$ K and crystal-field splitting energy $\Delta_1 = 33$ K, of CePd_2Al_3 from bulk experiments. Strong sample dependences between polycrystals and single-crystalline samples were found in the values of T_c for UPd_2Al_3 [4] and T_N for CePd_2Al_3 [5]. In fact, single crystals of CePd_2Al_3 do not order magnetically down to 0.3 K.

The thermopower, $S(T)$, was measured at Hiroshima University, from 1.7 to 300 K. The data on both polycrystal and single crystal along the principal axes of the

hexagonal crystal structure are displayed in Fig. 1. $S(T)$ is positive in the entire temperature range, with large absolute values. There is no tendency towards changing sign at low T . This observation allows for two interpretations. First, T_K and Δ_1 are close together, resulting in a position of the Kondo resonance just above the Fermi level. Second, ferromagnetic correlations of the cerium moments in the basal plane give the positive $S(T)$, as found for all ferromagnetic Kondo lattices, i.e. isostructural CePd_2Ga_3 [6]. The maximum of $S(T)$ occurs at about the same temperature for all samples, $T_{\text{max}} \approx 21$ K. This, in contrast, to the position of the maximum in the electrical resistivity, $\rho(T)$, which shifts from 32 K in polycrystals towards 6.3 K (a) and 5.0 K (c) [3, 5]. Thus, the maximum in $S(T)$ seems to be directly related to the crystal-field excitation at $\Delta_1 = 33$ K, which does not change for the various samples. For the polycrystal, no effect was observed at $T_K = 2.7$ K.

* Corresponding author.

Fig. 1. Thermopower of poly- and single-crystalline CePd_2Al_3 .

Fig. 2. Inelastic neutron cross-section of CePd_2Al_3 , corrected by the data of LaPd_2Al_3 , at $T = 12$ K (●) and $T = 100$ K (○).

Both T_K and Δ_1 can be directly measured with inelastic neutron scattering. We measured the energy-loss spectra (up to 55 meV) on annealed powder at PSI, with fixed final energy $E_f = 14.96$ meV, at temperatures from 12 to 160 K. The data taken on the nonmagnetic homolog LaPd_2Al_3 have been subtracted, as shown in Fig. 2 for $T = 12$ K and $T = 100$ K. A best fit is obtained when using Lorentzian line shapes. The width Γ , extrapolated to zero temperature (Γ_0), gives a first estimate of the Kondo temperature, while the position of the maximum, E_{max} , measures the excitation energy of the first excited CEF state. We find $\Gamma_0 = 1.92$ meV, corresponding to $T_K = 22.3(7)$ K. At 12 K, $E_{\text{max}} = 2.95$ meV, giving $\Delta_1 = 34(1)$ K [7]. These values are in excellent agreement with our earlier estimates [3]. Higher resolution is needed to determine accurately the quasielastic contribution. The absence of a second excitation up to 55 meV sets a lower limit to the overall splitting of ≈ 600 K. Interestingly, CePd_2Ga_3 was shown to possess a very similar CEF split ground state [6].

We investigated the sample-dependent occurrence of AF order in CePd_2Al_3 by zero-field μ^+ SR experiments, performed at PSI, on both poly- and single-crystalline samples; see Fig. 3. The upturn of the muon relaxation rate of the polycrystal indicates $T_N = 2.7$ K. This upturn

Fig. 3. Zero-field muon relaxation rate for polycrystalline (○) and single crystalline (●, $P_\mu \parallel a$) CePd_2Al_3 .

is absent in the single crystal (with muon polarization $P_\mu \parallel a$), suggesting that, at 1.5 K, the cerium moments are still fluctuating (strongly confined to the basal plane) at a rate above 10^7 Hz.

The absence of magnetic order in CePd_2Al_3 and the variation of T_c 's in UPd_2Al_3 originate from small differences in Al-concentration, which we established by Electron Probe Microanalysis. Ordering polycrystals are always slightly richer in Al than single crystals. In single crystals of UPd_2Al_3 , we found macroscopic (100 μm) oscillations in Al concentration along the growth direction ($\parallel a$). $\rho(T)$ along and perpendicular to this direction displays multistep and single-step T_c 's, respectively (not shown). These observations indicate that the Al atoms play a crucial role in establishing the magnetic coupling along the c -axis in CePd_2Al_3 , and the superconductivity in UPd_2Al_3 .

This work is partially supported by the Dutch Foundations FOM and NWO.

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