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Published in:
Physica B-Condensed Matter

Link to publication

Citation for published version (APA):

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Crystal symmetry and electronic properties of heavy-fermion MPd$_2$Al$_3$ (M = Ce, U).

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Abstract

We have determined the Kondo temperature and crystal-field splitting of the new heavy-fermion compound CePd$_2$Al$_3$ by thermopower and inelastic neutron scattering. The absence of static, magnetically ordering cerium moments in single crystals is demonstrated by zero-field $\mu$SR. Small deviations of aluminum stoichiometry yield significant variations of $T_K$ of CePd$_2$Al$_3$ and $T_N$ of UPd$_2$Al$_3$ in different samples.

Soon after the discovery of the new heavy-fermion superconductor UPd$_2$Al$_3$ [1], its cerium-based counterpart CePd$_2$Al$_3$ 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$_2$Al$_3$ from bulk experiments. Strong sample dependences between polycrystals and single-crystalline samples were found in the values of $T_K$ for UPd$_2$Al$_3$ [4] and $T_N$ for CePd$_2$Al$_3$ [5]. In fact, single crystals of CePd$_2$Al$_3$ 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 $\Delta_1$ 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$_2$Ga$_3$ [6]. The maximum of $S(T)$ occurs at a 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.

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Both $T_K$ and $\Delta_1$ 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$_2$Al$_3$ 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 $\Gamma$, extrapolated to zero temperature ($\Gamma_0$), gives a first estimate of the Kondo temperature, while the position of the maximum, $E_{\text{max}}$, measures the 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 $\approx 600$ K. Interestingly, CePd$_2$Ga$_3$ was shown to possess a very similar CEF split ground state [6].

We investigated the sample-dependent occurrence of AF order in CePd$_2$Al$_3$ by zero-field $\mu^+$ 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 $T_\mu = 2.7$ K. This upturn is absent in the single crystal (with muon polarization $P_\mu || a$), 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$_2$Al$_3$ and the variation of $T_\mu$'s in Upd$_2$Al$_3$ 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$_2$Al$_3$, we found macroscopic (100 μm) oscillations in Al concentration along the growth direction (||a), $p(T)$ along and perpendicular to this direction displays multistep and single-step $T_\mu$'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$_2$Al$_3$, and the superconductivity in Upd$_2$Al$_3$.

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

References

[7] A more detailed account of these experiments will be given in a forthcoming paper.