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μSR study of magnetism in heavy-fermion $U(Pt,Pd)_3$

A. de Visser[a,*], R.J. Keizer[a], R. van Harrevelt[a], A.A. Menovsky[a], J.J.M. Franse[a], A. Amato[b], F.N. Gygax[b], M. Pinkpank[b], A. Schenck[b]

[a] *Van der Waals-Zeeman Institute, University of Amsterdam, Valckenierstraat 65, 1018 XE Amsterdam, Netherlands*
[b] *Institute for Particle Physics, ETH Zürich, PSI, CH-5232 Villigen, Switzerland*

Abstract

We report on a μSR study of the evolution of antiferromagnetism across the $U(Pt_{1-x}Pd_x)_3$ series. Zero-field experiments on polycrystalline samples with $x = 0.01$ and 0.02 reveal spontaneous μ^+ precession frequencies below the Néel temperatures $T_N \approx 1.8$ K and 3.5 K, respectively. These frequencies relate to the so-called large-moment antiferromagnetism recently detected by neutron diffraction for $x = 0.01$ and 0.02. Transverse-field data ($B = 0.6$ T), taken on a single-crystalline sample with $x = 0.05$, reveal an unusual temperature and angular-dependent Knight shift. This points to a complex direction dependence of the moments at the uranium sites.

Keywords: Muon spin relaxation; Antiferromagnetism; Heavy-fermion compounds

The pseudobinary $U(Pt_{1-x}Pd_x)_3$ series forms an exemplary system to study the magnetic instability in heavy-electron compounds [1]. Neutron-diffraction experiments [2] demonstrated recently that two intimately connected magnetic-ordering phenomena take place. For $x \leqslant 0.01$, small-moment antiferromagnetism (SMAF) invariably sets in at $T_N \approx 6$ K, with an atypical quasi-linear temperature dependence of the order parameter, $m^2(T)$. The ordered moment, $m(T \to 0\ \text{K})$, grows with increasing Pd contents. For $x \geqslant 0.01$, the so-termed large-moment antiferromagnetism (LMAF) is observed. The LMAF is characterised by (i) a conventional $m^2(T)$, (ii) $T_N(x)$ increases up to 6.2 K for $x = 0.05$ and (iii) $m(x)$ grows rapidly up to 0.62 μ_B/U-atom for $x = 0.05$. The compound with $x = 0.01$ presents an intermediate case: SMAF sets in at ~ 6 K, while LMAF sets in at ~ 1.8 K.

Recently, we have reported zero-field μSR experiments on polycrystalline samples of the SMAF compounds UPt_3 and $U(Pt_{0.998}Pd_{0.002})_3$, and the LMAF compound $U(Pt_{0.95}Pd_{0.05})_3$ [3]. The SMAF ($T_N \approx 6$ K) did not show up in the μSR spectra, which suggests that the moment as observed by neutron diffraction [2] is rapidly fluctuating, or that the muons stop at a site where the dipolar fields cancel. For $x = 0.05$, two spontaneous μ^+ precession frequencies were observed for $T < T_N = 6.2$ K with values of 2.0 and 8.0 MHz in the limit $T \to 0$ K. This points to the presence of two μ^+-sites. Both frequency components have approximately the same amplitude, which suggests that the muons stop at both sites with the same probability.

In this paper we report (i) zero-field (ZF) μSR experiments on polycrystalline samples with intermediate Pd concentrations ($x = 0.01$ and 0.02) in order to study the evolution of the LMAF, and (ii) transverse-field (TF = 0.6 T) experiments on single-crystalline $U(Pt_{0.95}Pd_{0.05})_3$ conducted to determine the μ^+ stopping sites.

For $x = 0.02$ ($T_N = 3.5$ K and $m = 0.35 \pm 0.05\ \mu_B$/U-atom [2]) ZF data were taken for $T > 1.8$ K. Two spontaneous μ^+ precession frequencies were observed for $T < T_N$ with values ($T \to 0$ K) of 1.0 and 6.9 MHz. This points to the presence of two μ^+-sites, just as for $x = 0.05$. Again, both frequency components have approximately the same amplitude, which suggests that the muons stop at both sites with the same probability. For $x = 0.01$ ($T_N = 1.8$ K and $m = 0.11 \pm 0.03\ \mu_B$/U-atom [2]) ZF experiments were performed in the temperature interval 0.1–6 K. Only one spontaneous μ^+ precession frequency (ν) was observed. The μSR signals could adequately be fitted using the following three-component function:

* Corresponding author: Tel.: 31-20-5255732; fax: 31-20-5255788; e-mail: devisser@phys.uva.nl.

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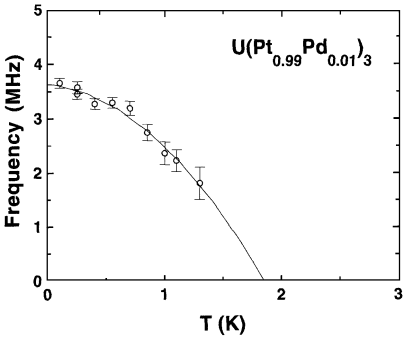

Fig. 1. Spontaneous μ^+ precession frequency for polycrystalline $U(Pt_{0.99}Pd_{0.01})_3$. The solid line is to guide the eye.

$$AG(t) = A_1 e^{-\lambda_1 t} + A_2(\tfrac{1}{3} e^{-\lambda'_2 t} + \tfrac{2}{3} e^{-\lambda_2 t} \cos(2\pi\nu t)) + A_3 G_{KT}(t), \quad (1)$$

where $G_{KT}(t)$ is the Kubo–Toyabe relaxation function. The second term is the normal depolarization function for a polycrystalline magnet. Adding the first term considerably improves the fit, which is taken as evidence that a second type of magnetically active stopping sites is present in the ordered state. The third term is attributed to the muon depolarization due to nuclear moments, a contribution which is generally present in the $U(Pt,Pd)_3$ pseudobinaries [3]. The temperature variation of ν and A_i ($i = 1, 2, 3$) is shown in Figs. 1 and 2, respectively. In the fitting procedure we took $\sum_i A_i$ fixed and, because $A_1 \approx A_2$, we also fixed $A_1 = A_2$. At ~ 2 K, A_1 and A_2 vanish, while ν drops to zero at about 1.8 K. This supports the idea of a cross-over from SMAF to LMAF at $T_N = 1.8$ K, as inferred from the neutron-diffraction data [2]. In the limit $T \to 0$ K, $\nu = 3.6$ MHz, which is much larger than the value of $\nu(x = 0.01) = 1.4$ MHz, calculated under the assumption that the frequencies scale with the ordered moments: $\nu(x = 0.01)/\nu(x = 0.05) = m(x = 0.01)/m(x = 0.05)$, where we used $\nu(x = 0.05) = 8$ MHz, $m(x = 0.01) = 0.11\ \mu_B$/U-atom and $m(x = 0.05) = 0.62\ \mu_B$/U-atom.

In an attempt to determine the μ^+-sites for the $U(Pt,Pd)_3$ series, we have measured the Knight shift (K_a, K_c) of single-crystalline $U(Pt_{0.95}Pd_{0.05})_3$ in the temperature interval 20–300 K in a transverse field of 0.6 T along the a- and the c-axis (hexagonal structure). A small anisotropy of the order of 0.1% is observed. At raising the temperature two unusual features are observed: (i) K_a and K_c cross near 140 K, and (ii) K_a exhibits a local maximum near 140 K. Since the Knight shift is not simply proportional to the magnetic susceptibility, extraction of the components of the dipolar tensor and the determination of the stopping sites is hampered. The complex character of the local magnetism is also reflected in the angular dependence of the Knight shift measured

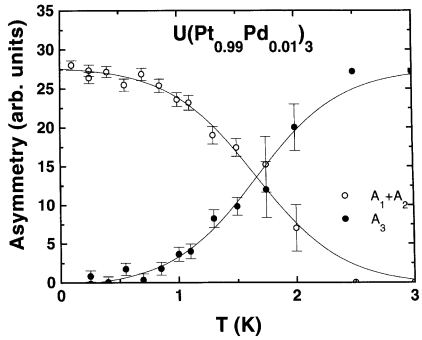

Fig. 2. Asymmetries $A_1 + A_2$ and A_3 for polycrystalline $U(Pt_{0.99}Pd_{0.01})_3$, determined by fitting the ZF muon depolarization spectra to Eq. (1). The solid lines are to guide the eye.

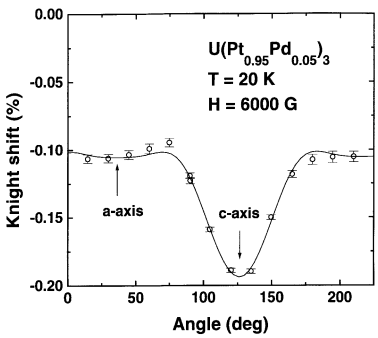

Fig. 3. Angular dependence of the Knight shift (TF = 0.6 T) of $U(Pt_{0.95}Pd_{0.05})_3$ in the (a,c) plane at $T = 20$ K.

in the (a, c) plane at $T = 20$ K (see Fig. 3). A fit to $K(\theta)$ invokes Legendre polynomial functions of the eighth order (see solid line in Fig. 1), which is highly unusual. Such an angular dependence cannot originate from the angular dependence of dipole fields, and must involve a complex direction (and possibly field) dependence of the moments at the uranium sites. The only other uranium compound for which such an intricate behaviour has been reported so far is U_2Zn_{17} [4].

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