ELSEVIER

Physica B 230–232 (1997) 53–55

PHYSICA B

μSR study of antiferromagnetic order in UPt_3 alloyed with Pd

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, The Netherlands*
[b] *Institute for Particle Physics, ETH Zürich, CH-5232 Villigen PSI, Switzerland*

Abstract

We report on a μSR study of the evolution of antiferromagnetism in the heavy-fermion pseudobinary series $U(Pt, Pd)_3$. For pure UPt_3 and $U(Pt_{0.998}Pd_{0.002})_3$ no small-moment antiferromagnetism has been observed in the present experiment: the zero-field μSR data yield a temperature-independent Kubo–Toyabe line width $\sigma_{KT} = 0.06 \pm 0.01\ \mu s^{-1}$ ($T \leqslant 8$ K). As recent neutron-diffraction experiments on UPt_3 and $U(Pt_{0.998}Pd_{0.002})_3$ do reveal magnetic order, we suggest that the muons stop at high symmetry sites where the dipole fields cancel. However, for the large-moment compound $U(Pt_{0.95}Pd_{0.05})_3$, the μSR data unambiguously show magnetic signals: below T_N two frequencies appear ($\nu_1 \simeq 8.0$ MHz and $\nu_2 \simeq 2.0$ MHz, for $T \to 0$), indicating two magnetically inequivalent muon stopping sites.

Keywords: μSR spectroscopy; Antiferromagnetic order; $U(Pt, Pd)_3$; UPt_3

The heavy-electron material UPt_3 may serve as an exemplary compound to explore the magnetic instability in strongly correlated electron systems. Neutron-scattering experiments [1] demonstrate that pure UPt_3 orders antiferromagnetically at $T_N = 5$ K, albeit with an extremely small ordered moment ($|\mu| = 0.02 \pm 0.01 \mu_B$/U-atom). The magnetic properties of UPt_3 are remarkably sensitive to alloying. For instance, by substituting small amounts of Pt by isoelectronic Pd [2], or U by Th [3], antiferromagnetism with significantly enhanced ordered moments is induced. In the case of the pseudobinaries $U(Pt_{1-x}Pd_x)_3$, magnetic, thermal and transport measurements [2] indicate that this so-termed large-moment antiferromagnetism (LMAF) is limited to a narrow concentration range ($0.02 \leqslant x \leqslant 0.07$), with a maximum T_N of 5.8 K for $x = 0.05$. Neutron-diffraction experiments [4] on a 5 at% Pd single-crystalline sample showed that the size of the ordered moment equals $0.6 \pm 0.2 \mu_B$/U-atom. The magnetic ordering consists of a doubling of the nuclear unit cell along the a^*-axis, with the magnetic moment directed along the a^*-axis. The same magnetic structure has been identified for the small-moment antiferromagnetism (SMAF) in pure UPt_3.

The first indications for a tiny ordered moment in UPt_3 came from early μSR experiments performed by Heffner and co-workers [5]: a significant increase of the zero-field Kubo–Toyabe relaxation rate (σ_{KT}) was observed below ~ 5 K, which was attributed to a weak static moment of the order of 10^{-3}–$10^{-2} \mu_B$/U-atom. Recently, we have embarked upon a series of μSR studies, in order to examine the evolution of magnetism on the microscopic level across the $U(Pt Pd)_3$ series. In this paper we report our first results.

* Corresponding author.

0921-4526/97/$17.00 Copyright © 1997 Elsevier Science B.V. All rights reserved
PII S0921-4526(96)00543-1

Polycrystalline samples $U(Pt_{1-x}Pd_x)_3$ with $x = 0.000$, 0.002 and 0.050 were prepared by arc-melting the constituents in stoichiometric amounts on a water-cooled copper crucible in a continuously titanium gettered high-purity argon atmosphere. The arc-melted samples were annealed at 900°C for a period of 7 d. Four thin slices (thickness 0.8 mm, area $6 \times 10\,mm^2$) were cut from the annealed buttons by means of spark-erosion. The surface layer, defected by spark-cutting, was removed by polishing (down to 0.3 μm) with diamond paste. The samples were glued on a silver support by GE varnish, in order to cover the desired area for the μSR experiments: $12 \times 20\,mm^2$. The samples were characterized by electrical resistivity measurements. In accordance with our previous data [6], the superconducting transition temperature T_c^+ amounts to 533 and 389 mK, and ρ_0 equals 0.88 and 2.49 μΩ cm, for 0 and 0.2 at% Pd, respectively, T_N for the 5 at% Pd sample amounts to 6.3 K, which is slightly higher than reported previously (5.8 K) [2].

Zero-field μSR experiments have been performed on pure UPt_3 in the T-interval 2.7–7.0 K and for $x = 0.002$ in the T-interval 0.9–8.0 K. The muon depolarization is well described by the standard Kubo–Toyabe function. The Kubo–Toyabe line width, σ_{KT}, is shown for $x = 0$ and $x = 0.002$ in Figs. 1 and 2, respectively. Inspecting Figs. 1 and 2, we conclude that for both compounds σ_{KT} is almost T-independent and amounts to $0.06 \pm 0.01\ \mu s^{-1}$. For $x = 0.002$, data taken in a transverse field of 100 G, gave best fits using a Gaussian relaxation function. The resulting relaxation rate, σ_G, is comparable to σ_{KT} and displays no significant T-dependence either (Fig. 2).

Surprisingly, our data for polycrystalline UPt_3 differ from the earlier data by Heffner et al. [5], who reported a doubling of σ_{KT} from a value of $0.06\ \mu s^{-1}$ just above 5 K to a value of $0.12\ \mu s^{-1}$ for $T \to 0$ K. In other words, our μSR data do not confirm static weak magnetic order in the samples with $x = 0$ and $x = 0.002$. However, recent neutron-diffraction experiments [7] on single-crystalline $U(Pt, Pd)_3$ with comparable small values of x, do reveal magnetic order. A similar result was recently obtained by Dalmas de Réotier et al. [8] for high-purity single-crystalline UPt_3: weak magnetic order was detected by neutron scattering, but was not found in the μSR signals obtained on the same sample. A possible explanation for this might be a total cancellation of the dipole fields at the muon stopping site(s). Such a cancellation may indeed occur in "perfect" crystals. The data by Heffner et al. could then be explained by assuming that their sample was not "perfect".

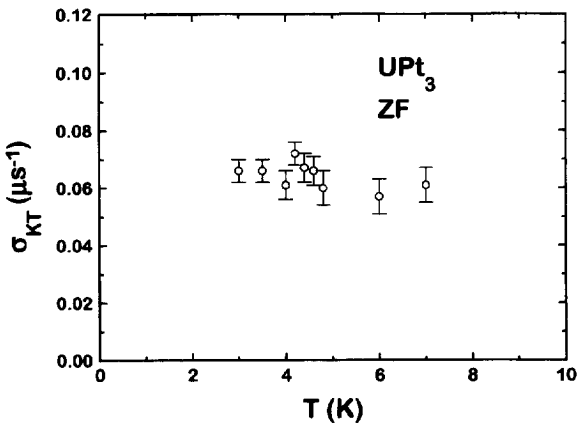

Fig. 1. Zero-field Kubo–Toyabe line width for polycrystalline UPt_3.

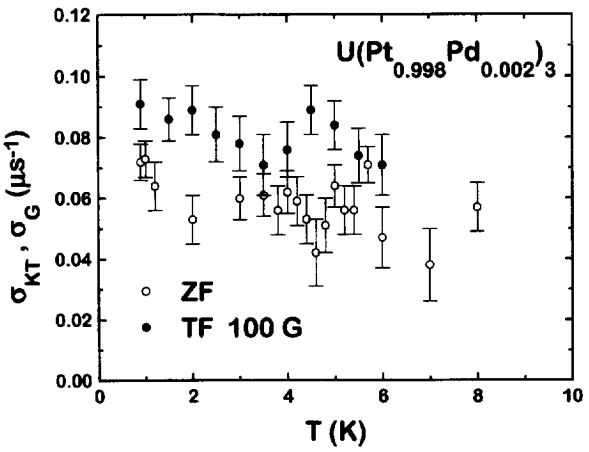

Fig. 2. Zero-field Kubo–Toyabe line width (○) and transverse field (100 G) Gaussian line width (●) for polycrystalline $U(Pt_{0.998}Pd_{0.002})_3$.

Zero-field μSR experiments have been carried out on $U(Pt_{0.95}Pd_{0.05})_3$ in the T-interval 3–10 K. In the ordered state ($T < T_N = 6.3$ K), the

relaxation spectra show two precession frequencies. A typical spectrum, taken at $T = 3.0$ K, is shown in Fig. 3. Best fits are obtained using two exponentials. The T-variation of the resulting frequencies is shown in Fig. 4. For $T \to 0$ K, we find $\nu_1 \simeq 8.0$ MHz and $\nu_2 \simeq 2.0$ MHz. The presence of two frequency components indicates two magnetically inequivalent muon stopping sites. The two exponentials have approximately the same amplitude, which suggests that muons stop at both sites with the same probability. Interestingly, almost identical frequencies were observed for $U_{0.95}Th_{0.05}Pt_3$ [5], which orders antiferromagnetically at $T_N = 6.2$ K as well (with the same magnitude for the ordered moment, i.e. $0.6\mu_B$/U-atom). This strongly suggests identical stopping sites in both materials.

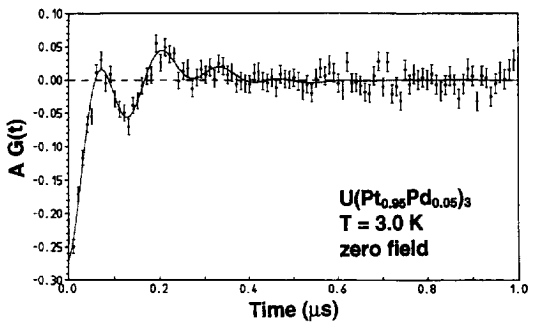

Fig. 3. Zero-field μSR signal for polycrystalline $U(Pt_{0.95}Pd_{0.05})_3$ at $T = 3.0$ K.

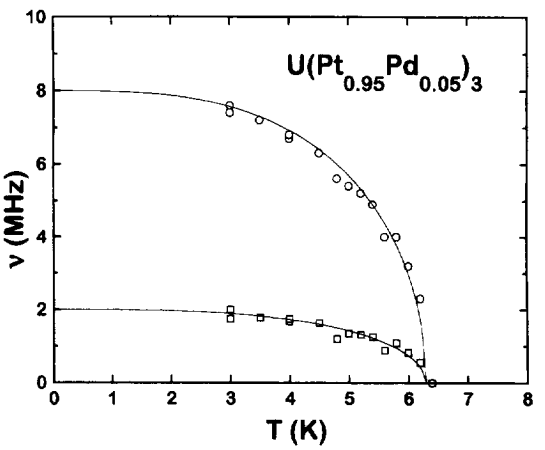

Fig. 4. Zero-field frequency components for polycrystalline $U(Pt_{0.95}Pd_{0.05})_3$.

Our new μSR results demonstrate a salient difference between the small- and large-moment antiferromagnetic order in the $U(Pt, Pd)_3$ series. The SMAF does not show up in the μSR signals. Since the magnetic structures of pure UPt_3 and the Pd-doped compounds are identical, one would expect UPt_3 to exhibit a spontaneous frequency of about 0.2–0.3 MHz (the ordered moment is a factor 30 smaller compared to the 5 at% Pd compound). The fact that the internal magnetic fields cancel at the muon stopping sites in the small-moment compounds, possibly indicates that the SMAF is of a different nature than concluded from the neutron-diffraction experiments. μSR studies on compounds with intermediate Pd concentrations are underway, as well as a determination of the muon stopping site(s) for $U(Pt_{0.95}Pd_{0.05})_3$.

Acknowledgements

This work was supported by the Dutch Foundation for Fundamental Research of Matter (FOM).

References

[1] G. Aeppli, E. Bucher, C. Broholm, J.K. Kjems, J. Baumann and J. Hufnagl, Phys. Rev. Lett. 60 (1988) 615.
[2] A. de Visser, A. Menovsky and J.J.M. Franse, Physica B 147 (1987) 81.
[3] A.P. Ramirez, B. Batlogg, A.S. Cooper and E. Bucher, Phys. Rev. Lett. 57 (1986) 1072.
[4] P.H. Frings, B. Renker and C. Vettier, J. Magn. Magn. Mater. 63&64 (1986) 202.
[5] R.H. Heffner et al., Phys. Rev. B 39 1989) 11 345.
[6] A. de Visser, P. Haen, T. Vorenkamp, M. van Sprang, A.A. Menovsky and J.J.M. Franse, J. Magn. Magn. Mater. 76&77 (1988) 112.
[7] A. de Visser et al., these Proceedings (SCES'96), Physica B (1996).
[8] P. Dalmas de Réotier, A. Huxley, A. Yaouanc, J. Flouquet, P. Bonville, P. Imbert, P. Pari, P.C.M. Gubbens and A.M. Mulders, Phys. Lett A 205 (1995) 239.