

Available online at www.sciencedirect.com





Journal of Magnetism and Magnetic Materials 272-276 (2004) 244-245

www.elsevier.com/locate/jmmm

Crossover from anomalous to conventional antiferromagnetism in Pd-doped UPt₃ studied via cantilever magnetometry

C.P. Opeil^a, A. de Visser^b, M.J. Naughton^a, M.J. Graf^{a,*}

^a Department of Physics, Boston College, Chestnut Hill, MA 02467, USA ^b Van der Waals-Zeeman Inst., Valckenierstraat 65, 1018 XE Amsterdam, The Netherlands

Abstract

UPt₃ exhibits an anomalous form of antiferromagnetic (AF) order, which is thought to be fluctuating in time. Substitution of Pd for Pt induces conventional AF behavior. In an attempt to further probe the possible existence of an AF quantum critical point in $U(Pt_{1-x}Pd_x)_3$ at $x_c \sim 0.006$, we have made cantilever magnetometer measurements on single crystalline $U(Pt_{1-x}Pd_x)_3$ for x = 0.01, 0.02, and 0.05 in magnetic fields of 6T. While readily observable for the samples with x = 0.02 and 0.05, we find no evidence for the AF transition at x = 0.01, in contrast to μ SR measurements.

© 2003 Published by Elsevier B.V.

PACS: 71.27.+a; 74.70.Tx; 75.30.Kz

Keywords: Heavy-fermion; Quantum critical point; UPt3; Antiferromagnetism; Magnetometer

The unconventional heavy fermion superconductor $(T_c \sim 0.6 \text{ K})$ UPt₃ is characterized by antiferromagnetic (AF) order, observed through neutron scattering, that develops below $T^* = 6$ K and coexists with (and couples to) superconductivity (SC). This state is unconventional in the sense that the square of the ordered moment $m^2(T)$ grows quasilinearly with decreasing temperature and that the size of the ordered moment is extremely small, $m \sim 0.02 \,\mu_{\rm B}/{\rm U}$ -atom. Other bulk and local probe measurements such as NMR [1] and μ SR [2,3] fail to detect entry into this "small moment antiferromagnetic" (SMAF) phase. It has been suggested that the magnetic order fluctuates on timescales faster than the characteristic times of μ SR and NMR, but slower than that for neutron scattering, and so T^* represents a crossover temperature rather than a true ordering temperature. Doping UPt₃ with isoelectronic Pd reveals quick suppression of SC [4] and the emergence of the

conventional "large moment antiferromagnetic" phase (LMAF) for which at optimum doping (x = 0.05), the ordered moment is ~0.63 $\mu_{\rm B}$ /U-atom and $T_{\rm N} = 5.8$ K. The LMAF has the *same* magnetic structure as the SMAF, and its $T_{\rm N}(x)$ follows a Doniach-type diagram which extrapolates to $T_{\rm N} = 0$ K at $x_{\rm c} = 0.006$ [5]. Between $x_{\rm c}$ and x = 0.05 this phase line separates the SMAF phase from the LMAF phase. The origin and nature of the transition from SMAF to LMAF is unclear. Indeed, while μ SR shows that x = 0.01 samples have an LMAF transition at 1.8 K, bulk probes (e.g. resistivity, specific heat) detect no anomaly. The objective of this work is to investigate whether the change from SMAF to LMAF is a true phase transition or whether SMAF gradually evolves into LMAF.

We present measurements for three single crystalline $U(Pt_{1-x}Pd_x)_3$ samples with x = 0.01, 0.02, and 0.05, which have $T_N = 1.8$, 3.4, and 5.8 K, respectively, and ordered moments of 0.11, 0.35 and 0.63 μ_B/U -atom, respectively, as determined by neutron diffraction [6]. The samples were attached via Apiezon N-grease to a silicon cantilever magnetometer [7] (SCM) utilizing force

^{*}Corresponding author. Tel.: +1-627-552-4128; fax: +1-617-552-8478.

E-mail address: grafm@bc.edu (M.J. Graf).



Fig. 1. Temperature-dependent magnetization of single crystal $U(Pt_{1-x}Pd_x)_3$ with $B\parallel$ to the *a*-axis with concentrations as indicated. The x = 0.01 curve has been offset + 200 emu/mol-U for clarity.

mode with B perpendicular to the cantilever plane and parallel to the sample's crystalline a-axis, and positioned in a field gradient. The SCM capacitance is monitored by an ultra-sensitive autobalancing bridge operating at 1 kHz. The SCM was mounted on a thin sapphire plate, thermally connected to the cold finger of a ³He refrigerator and monitored with field calibrated thermometers. Magnetization measurements were obtained by zero field cooling the samples from room temperature to below 10 K, raising the magnetic field to 6 T, and then slowly decreasing the temperature from 8 to < 1 K. The observed Néel temperatures for x = 0.05 and 0.02 are shifted from the zero-field values by 10%. We assume that the application of 6T will not effect the possible LMAF transition for the x = 0.01 sample, although earlier work suggests that T_N might be more significantly depressed [8].

The molar magnetization M for several Pd concentrations at B = 6 T is plotted vs. T in Fig. 1. The absolute values for M are roughly within a factor of 2 of the accepted values, [9] with the accuracy being limited by the variations in the demagnetization factors, misalignment of the crystals, and mixing of the force and torque contributions from the SCM. The zero-field Néel temperatures from neutron scattering for these samples [6] are marked by arrows, and correspond to abrupt changes in the slope in M(T) for x = 0.05 and 0.02. No break of slope occurs in the x = 0.01 sample. To highlight the transitions, in Fig. 2 we plot dM/dT vs. $T/T_{\rm N}(B=0)$. The curves are offset for clarity. Prominent transitions are evident for the x = 0.05 and 0.02 samples. In contrast, no transition is evident for the x = 0.01 sample.

Experimentally we find the decrease of M below T_N for x = 0.05 and 0.02 scales approximately as m^2 , and extrapolating this behavior to x = 0.01 we estimate that the anomaly should be readily observable by our SCM.



Fig. 2. dM/dT vs. $T/T_N(B = 0)$ for x = 0.01, 0.02 and 0.05 in a field of 6 T. The curves are offset for clarity, and the x = 0.01 curve is multiplied by 4 to highlight the lack of an anomaly near T_N .

We have also looked at the $B \| c$ -axis for x = 0.01 and find no evidence for the transition. This result is consistent with less sensitive specific heat measurements, which also clearly show a transition for x = 0.02 but not for x = 0.01.

In conclusion, we could not detect the transition to the LMAF phase at x = 0.01, despite what appears to be sufficient sensitivity and a clear indication of magnetic ordering at $T_N \sim 1.8$ K as determined by μ SR [2] and neutron diffraction [6]. Thus we cannot rule out the possibility that the SMAF evolves gradually from the LMAF, e.g., at x = 0.01 the SMAF temporal fluctuations have been slowed sufficiently by Pd "impurities" to appear static on μ SR timescales, but still undetectable by our DC magnetization measurements. We are expanding these studies to include a more systematic variation of the transition with x, as well as extending the measurement to include frequency dependence of the magnetic response.

Work funded by Petroleum Research Fund of the American Chemical Society (34579-AC5), DOE (DEF 60202ER63404), Sigma Xi Grant-in-Aid of Research.

References

- [1] H. Tou, et al., Phys. Rev. Lett. 77 (1996) 1374.
- [2] R.J. Keizer, et al., J. Phys.: Condens. Matter 11 (1999) 8591.
- [3] P. Dalmas de Réotier, et al., Phys. Lett. A 205 (1995) 239.
- [4] M.J. Graf, et al., Phys. Rev. B 60 (1999) 3056;
 M.J. Graf, et al., Physica B 259–260 (1999) 666.
- [5] A. de Visser, et al., Phys. Rev. Lett. 85 (2000) 3005.
- [6] R.J. Keizer, et al., Phys. Rev. B 60 (1999) 6668.
- [7] M. Chaparala, et al., AIP Conf. Proc. 273 (1992) 407.
- [8] F.S. Tautz, et al., Physica B 223&224 (1996) 178.
- [9] A. de Visser, et al., Physica B 147 (1987) 81.