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Neutron-diffraction study of antiferromagnetic order in $U(Pt, Pd)_3$

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Abstract

Neutron-diffraction experiments have been carried out on single-crystalline samples of the heavy-fermion pseudobinaries $U(Pt_{1-x}Pd_x)_3$ ($x \leqslant 0.05$) in order to investigate the evolution of magnetic order. At low Pd contents ($x = 0.002$ and 0.005), small-moment magnetic order is observed below ~ 6 K, just like in pure UPt_3. For $x = 0.02$ and 0.05, the ordered moment is about one order of magnitude larger compared to UPt_3. The compound with $x = 0.01$ represents a most intriguing intermediate case as it exhibits a cross-over behaviour from small-moment to large-moment antiferromagnetic order.

Keywords: Neutron diffraction; Antiferromagnetic order; $U(Pt, Pd)_3$

In the past years, it has been recognized that an unusual type of small-moment magnetic order arises quite generally in heavy-electron materials. In the case of UPt_3, the small-moment antiferromagnetic order (SMAF) is of special interest, because it possibly couples to the unconventional superconducting order parameter which generates the multicomponent superconducting phase diagram. So far, the tiny moment has been observed convincingly by neutron diffraction only [1,2]. Below ~ 5 K, antiferromagnetic (AF) order sets in with an extremely small ordered moment. The magnetic order is unusual in the sense that the magnetic Bragg intensity grows approximately linearly with decreasing temperature, and the size of the ordered moment, $|\mu|$, amounts to $0.02 \pm 0.01\mu_B$/U-atom in the limit $T \to 0$ K. The AF ordering vector is $[\frac{1}{2}, 0, 0]$ and the moments point along the a^* axes in the hexagonal structure.

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By alloying UPt_3, e.g. by substituting Pt by isoelectronic Pd [3], one can easily induce magnetic moments one or two orders of magnitude larger. Magnetic, thermal and transport measurements [3] performed on pseudobinary $U(Pt_{1-x}Pd_x)_3$ indicate that this so-termed large-moment antiferromagnetism (LMAF) is restricted to the narrow concentration range $0.02 \leqslant x \leqslant 0.07$. At optimal doping ($x = 0.05$) the maximum T_N amounts to 5.8 K, while the ordered moment equals $0.6 \pm 0.2\mu_B$/U-atom [4]. The magnetic structure of UPt_3 is identical to the one reported for the Pd doped compounds. Here, we report the first results of a neutron- diffraction study conducted to investigate the evolution of magnetism in pseudobinary $U(Pt_{1-x}Pd_x)_3$.

Single-crystalline samples $U(Pt_{1-x}Pd_x)_3$, with $x = 0.002, 0.01, 0.02$ and 0.05, were prepared in a tri-arc furnace using the Czochralski technique, while a crystal with $x = 0.005$ was prepared in a mirror furnace using the horizontal floating zone method. The experiments were performed on

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samples with dimensions of approximately $4 \times 4 \times 4$ mm^3, which were cut from the as-grown crystals by spark erosion. The samples were annealed at 950 °C for a period of 5 days.

The Bragg positions of UPt_3 (space group $P6_3/mmc$) are labelled using reciprocal lattice units, where $a^* = b^* = 4\pi/(a\sqrt{3}) = 1.264$ Å^{-1} and $c^* = 2\pi/c = 1.283$ Å^{-1}. At SILOE (CEA), $U(Pt_{1-x}Pd_x)_3$ samples with $x = 0.01$, 0.02 and 0.05 were studied, using the DN1 triple-axis spectrometer. The crystals were mounted with the c^*-axis vertical, i.e. perpendicular to the scattering plane. At the ILL, experiments were carried out using the IN14 triple-axis spectrometer. The $x = 0.005$ sample was mounted with the c^*-axis vertical, while the $x = 0.002$ and 0.01 samples were mounted with the b^*-axis vertical. Triple-axis spectrometers with the analyzer set to zero-energy transfer were used in order to separate the elastic Bragg scattering from possible low-energy magnetic excitations. Throughout this paper the magnetic Bragg intensities are given by the maximum peak values. The widths of the Bragg peaks were almost constant. A full account of the experimental conditions will be published elsewhere [5].

Fig. 1 shows the magnetic Bragg intensities measured at $Q = [1, \frac{1}{2}, 0]$ for the large-moment compounds $U(Pt_{1-x}Pd_x)_3$ with $x = 0.02$ and 0.05. The vertical axis has been scaled to yield $(|\mu|/\mu_B)^2$, by comparing the intensity of the magnetic peaks with the [1, 1, 0] nuclear reflection. For both compounds, the T-variation of the ordered moment can be expressed as $|\mu|^2 \propto 1 - (T/T_N)^\alpha$. For $x = 0.05$ we find $|\mu| = 0.62 \pm 0.05\mu_B$/U-atom, $T_N = 5.9$ K and $\alpha = 3.0$, while for $x = 0.02$ we obtain $|\mu| = 0.35 \pm 0.05\mu_B$/U-atom, $T_N = 3.5$ K and $\alpha = 1.8$.

In Fig. 2, we show the magnetic Bragg intensity measured at $Q = [\frac{1}{2}, 0, 1]$ for $x = 0.002$ and at $Q = [1, \frac{1}{2}, 0]$ for $x = 0.005$. Here, the value for the ordered moments has been determined by comparing the magnetic Bragg intensities with the [1, 0, 1] and [1, 1, 0] nuclear peaks, respectively. Both compounds display SMAF as in pure UPt_3. The value of $|\mu|^2$ starts to rise slowly below $T_N \simeq 6$ K. From 4 K down to 1.5 K the Bragg intensities vary approximately linearly in temperature. In the limit $T \to 0$ K, $|\mu|$ amounts to 0.048 ± 0.01 and $0.038 \pm 0.01\mu_B$/U-atom, for $x = 0.005$ and 0.002, respectively.

The T-dependence of the magnetic Bragg intensity of the 1 at% Pd sample, shown in Fig. 3,

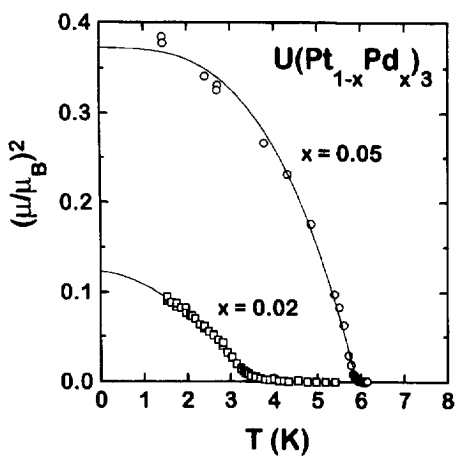

Fig. 1. Temperature variation of $|\mu|^2$ measured at the Bragg peak $Q = [1, \frac{1}{2}, 0]$, for $U(Pt_{1-x}Pd_x)_3$ with $x = 0.05$ (○) and $x = 0.02$ (□). The solid lines yield fits to $|\mu|^2 \propto 1 - (T/T_N)^\alpha$ (see text).

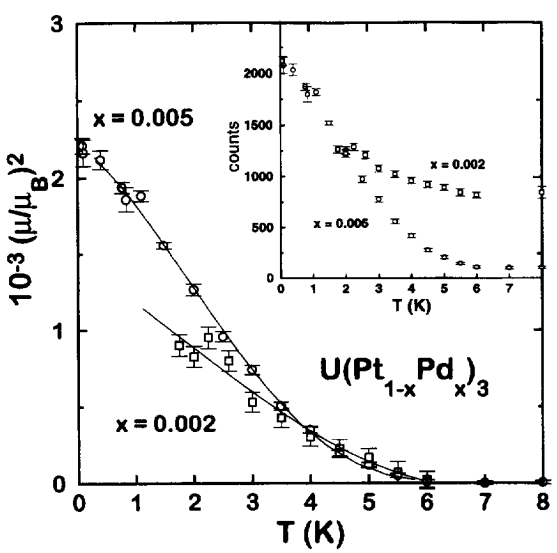

Fig. 2. Temperature variation of $|\mu|^2$ measured at the Bragg peak $Q = [\frac{1}{2}, 0, 1]$ and at $Q = [1, \frac{1}{2}, 0]$ for $U(Pt_{1-x}Pd_x)_3$ with $x = 0.005$ (○) and $x = 0.002$ (□), respectively. The solid lines are to guide the eye. The insert shows the measured counts per 60 min for $x = 0.002$ and per 6 min for $x = 0.005$.

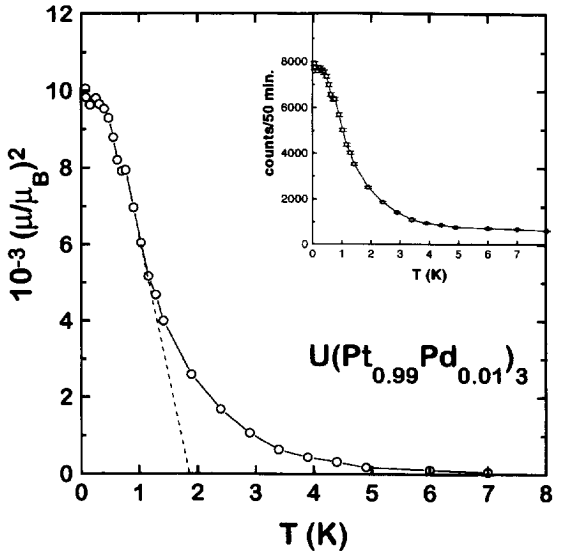

Fig. 3. Temperature variation of $|\mu|^2$ measured at the Bragg peak $Q = [\frac{1}{2}, 0, 1]$ for $U(Pt_{1-x}Pd_x)_3$ with $x = 0.01$ (○). The solid line is to guide the eye. The insert shows the measured counts per 50 min.

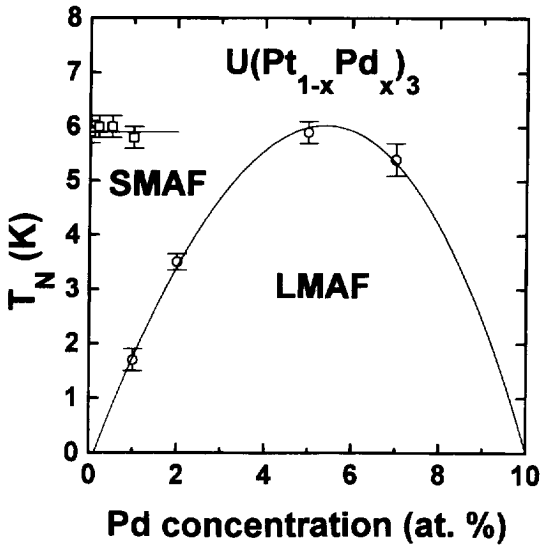

Fig. 4. Néel temperature versus Pd concentration of $U(Pt_{1-x}Pd_x)_3$. SMAF and LMAF denote small-moment and large-moment antiferromagnetism, respectively. Note that for $x = 0.01$ two Néel temperatures can be defined. The data point for pure UPt_3 has been taken from Ref. [2].

is quite intriguing: $|\mu|^2$ grows slowly below ~ 6 K, rises rapidly below 2 K and saturates below ~ 0.5 K. The rapid rise near 2 K suggests a cross-over from the small-moment to the large-moment state. For $T \to 0$ K, $|\mu|$ reaches a value of $0.11 \pm 0.03\mu_B$/U-atom. T_N for the LMAF is estimated at 1.8 K (see dashed line in Fig. 3).

Our neutron diffraction experiments evidence magnetic order for all samples investigated ($x \leqslant 0.05$). The Néel temperatures for the various samples are collected in Fig. 4. On decreasing the Pd concentration below 5 at%, T_N and $|\mu|$ gradually drop. The phase boundary for the LMAF extrapolates to $T_N = 0$ for $x \simeq 0.00$, locating the magnetic instability in or near pure UPt_3. On the other hand, at low Pd concentrations ($x \leqslant 0.01$) SMAF invariably sets in at ~ 6 K, just like in pure UPt_3. Probably, the phase line for the SMAF extends to higher Pd contents, but for $x \geqslant 0$ it becomes more and more difficult to discriminate experimentally between SMAF and LMAF. For the 1 at% Pd sample, a cross-over regime has been identified. This compound yields Néel points, at 6 and 1.8 K, for the small and large-moment magnetic order, respectively.

The current results stress the special character of the SMAF. The magnetic Bragg intensity has an unusual temperature dependence, which is likely due to the presence of magnetic correlations. On decreasing the Pd concentration the magnetic correlations become more short-ranged as in the pure compound where $\xi \sim 250$ Å [1]. Also our recent μSR experiments [6] underline the special character of the SMAF, because it does not show up as a spontaneous frequency in the spectra. An extensive discussion of the magnetism in the $U(Pt,Pd)_3$ series on the microscopic level will be published elsewhere [5].

Acknowledgements

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