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μ SR study of spin-glass freezing in $Y_{1-x}U_xPd_3$ and UPd_4 Disappearance of the ordered moment in the 'Kondo regime'

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Muon spin relaxation (μ SR) measurements have been performed on polycrystalline specimens of $Y_{1-x}U_xPd_3$ with x = 0.1, 0.2, 0.4 and on UPd₄. Spin-glass-like magnetic order was observed in UPd₄ at $T_g \simeq 13$ K and in $Y_{0.6}U_{0.4}Pd_3$ at $T_g \simeq 12$ K. The average static random internal field H_{int} at the muon site is found to decrease very rapidly with U concentration x, indicating that the ordered static U moment rapidly disappears with decreasing U concentration around $x \simeq 0.2$. Our results are consistent with the conjecture that Kondo screening eliminates the magnetic moment around this concentration.

It has been found [1] that the alloy system $Y_{1-x}U_xPd_3$ displays a variety of magnetic behaviors ranging from apparent localized U ions as $x \rightarrow 1$ through possible spin glass behavior for $0.3 \le x \le 0.5$. At x = 0.2, electrical resistivity, magnetic susceptibility and specific heat measurements have been interpreted in terms of the two-channel quadrupolar Kondo effect [1]. It is therefore worthwhile to use a microscopic probe of local magnetic fields – μ SR to observe the change in magnetic behavior with U concentration x.

Muon Spin Relaxation (μ SR) is an extremely sensitive method of measuring local magnetic fields in solids, which has been described elsewhere [2]. The experiments reported here were performed at the M15 and M20 surface muon channels at TRIUMF. The polycrystalline specimens of $Y_{1-x}U_xPd_3$ with various U concentrations x = 0.1, 0.2, 0.4 and UPd₄ were prepared by arc melting in an ultrahigh purity argon atmosphere. The specimens of Y_{0.6}U_{0.4}Pd₃ and UPd₄ were measured down to 3 K using a ⁴He gas flow cryostat, while specimens of $\mathbf{Y}_{0.8}\mathbf{U}_{0.2}\mathbf{Pd}_3$ and $Y_{0.9}U_{0.1}Pd_3$ were measured down to several tens of mK using an Oxford dilution refrigerator. Both zero field and longitudinal field measurements (with a field applied parallel to the initial muon spin polarization) were performed to distinguish between muon spin depolarization due to static and dynamic local magnetic fields. Weak transverse field measurements were also made in order to check the solid-angle factor $N_{\rm E}/N_{\rm B}$ of the forward and backward counters.

Figure 1 shows the time-dependent muon spin polarization for $Y_{1-x}U_xPd_3$ with x = 0.1, 0.2, 0.4 and for UPd, at different temperatures in zero field. In $Y_{0,4}U_{0,4}Pd_3$ and UPd_4 , the polarization at the lowest temperature decreases to its minimum value in a very short time, less than 0.05 µs in UPd₄ and less than 0.08 μ s in Y_{0.6}U_{0.4}Pd₃, and then recovers to about 1/3 of the initial value. This is a clear signature of spin glass order [2]. The depolarization at the lowest temperature becomes much slower in Y_{0.8}U_{0.2}Pd₃, indicating a much smaller average static internal field compared to $Y_{0.6}U_{0.4}Pd_3$ and UPd_4 . In $Y_{0.9}U_{0.1}Pd_3$, most of the depolarization at the lowest temperature is due to the nuclear dipolar magnetic field. The very small difference in the polarization between 2.1 K and 20 mK shows that static magnetic order disappears almost completely when the U concentration is decreased to 0.1. Since the average static internal field is proportional to the ordered U moment in the ground state, fig. 1 demonstrates that the ordered static U moment rapidly disappears with decreasing U concentration around $x \simeq 0.2$.

Compared to usual d-electron spin glass systems, such as AuFe or CuMn, with much more dilute magnetic moments, the U concentration in the $Y_{0,6}U_{0,4}Pd_3$ and UPd₄ compounds is relatively high.

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Fig. 1. Muon spin relaxation spectra of $Y_{1-x}U_xPd_3$ and UPd_4 compounds observed in zero field. The fast decrease of polarization followed by recovery to about 1/3 in $Y_{0.6}U_{0.4}Pd_3$ and UPd_4 is a clear signature of spin glass order. The spectra of $Y_{0.8}U_{0.2}Pd_3$ and $Y_{0.9}U_{0.1}Pd_3$ include a background signal, which is about 20% of the total signal.

The muon spin polarization in AuFe and CuMn spin glass systems has been treated assuming a Gaussian field distribution at each muon site and a probability function $\rho(\Delta) = \sqrt{2/\pi}(a/\Delta^2) \exp(-a^2/2\Delta^2)$ to account for different ranges of magnetic fields Δ at different muon sites [2]. The total field distribution $P(H_i)$ (i = x, y, z) in the dilute systems is approximated by a Lorentzian distribution. We follow the same procedures in ref. [2] to calculate the polarization function $G_z(t)$ with a slightly modified probability function $\rho(\Delta)$. In order to compare the average static internal field in the $Y_{1-x}U_xPd_3$ and UPd₄ compounds, we choose HWHM (half width at half maximum) of the static internal field distribution $P(H_i)$.

The muon spin polarization of UPd₄ and $Y_{1-x}U_xPd_3$ has been fitted with the fixed functional form of $\rho(\Delta)$ obtained from fitting the polarization at the lowest temperature to $G_z(t)$. The HWHM of the static internal field distribution is shown in fig. 2. For UPd₄ and $Y_{0.6}U_{0.4}Pd_3$, there is sharp increase in the HWHM when the temperature is decreased below T_g , with values of HWHM(T = 2.6 K) = 450 ± 20 G and $T_g = 13 \pm 1.5$ K in UPd₄, and HWHM(T = 3.45 K) = 260 ± 10 G and $T_g = 11.5 \pm 0.5$ K in $Y_{0.6}U_{0.4}Pd_3$. In



Fig. 2. HWHM (half width at half maximum) of the static internal magnetic field distribution $P(H_i)$ as a function of temperature in $Y_{1-x}U_xPd_3$ and UPd₄. For $Y_{0.6}U_{0.4}Pd_3$ and UPd₄, there is a sharp increase in the HWHM when the temperature is decreased below the freezing temperature T_g . The HWHM of the static internal field distribution $P(H_i)$ in $Y_{0.6}U_{0.4}Pd_3$ and UPd₄ is more than 100 times larger than that in $Y_{0.8}U_{0.2}Pd_3$.

 $Y_{0.6}U_{0.4}Pd_3$, by applying a small field parallel to the initial muon spin polarization, we also performed decoupling measurements at low temperatures and near the freezing temperature T_g . The longitudinal field measurements indicate that there is still some static magnetic order at 11.0 K, but no static magnetic order above 11.5 K, consistent with the freezing temperature T_g of 11.5 K in the $Y_{0.6}U_{0.4}Pd_3$ sample.



Fig. 3. (a) Low-temperature HWHM $(T \rightarrow 0)$ of the static internal field distribution $P(H_i)$ as a function of U concentration. (b) Phase diagram determined by μ SR. These results support the conjecture that the Kondo screening eliminates the ordered static U moment in $Y_{1-x}U_xPd_3$ around U concentration $x \approx 0.2$.

The muon spin polarization of Y_{0.8}U_{0.2}Pd₃ and $Y_{0,9}U_{0,1}Pd_3$ has been fitted after subtraction of a temperature-independent background signal, obtained from fitting the polarization at the highest temperature. For $Y_{0.8}U_{0.2}Pd_3$, we obtain HWHM(T =0.05 K) = 2.0 ± 0.04 G. As shown in fig. 2, the temperature dependence of the HWHM of the static internal field distribution in Y_{0.8}U_{0.2}Pd₃ is very unusual, and it is difficult to define a freezing temperature T_{g} . This is possibly because the interaction between U ions at this U concentration is too weak to retain the usual spin glass behavior, or because the U concentration is not homogeneous throughout the specimen. The onset of the very small static internal field H_{int} occurs near 2.5 ± 1 K, as indicated by the open circle in fig. 3. For $Y_{0.9}U_{0.1}Pd_3$, the upper limit of the HWHM of the internal field distribution is estimated to be 0.15 G.

Figure 3 shows the low-temperature HWHM of the static internal field distribution and the freezing temperature T_g as functions of the U concentration x in the $Y_{1-x}U_xPd_3$ and UPd_4 compounds. We see that the freezing temperature T_g decreases with decreasing U concentration x. The HWHM of the static internal

field distribution in $Y_{0.6}U_{0.4}Pd_3$ and UPd_4 is more than one hundred times larger than in $Y_{0.8}U_{0.2}Pd_3$. This is consistent with the conjecture that the ground state of the U ions is nonmagnetic in $Y_{1-x}U_xPd_3$ for U concentration $x \le 0.2$, where the physical properties have been interpreted in terms of the two-channel quadrupolar Kondo effect. Details of this work will be published elsewhere.

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