Neutron Scattering Studies of $Y_{1-x}U_xPd_3$ Compounds

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We have performed neutron scattering measurements on two alloys of $Y_{1-x}U_xPd_3$ ($x = 0.2, 0.45$). Our results indicate that the $x = 0.45$ compound orders antiferromagnetically with a moment of $0.7\mu_B$ per U atom and a Néel temperature (T_N) of 21 K. Although the $x = 0.2$ compound does not order above 0.2 K, critical fluctuations associated with antiferromagnetic ordering were observed on cooling from 77 to 0.2 K. The magnetic spectra for both compounds, obtained by polarized inelastic neutron scattering, were dominated by a quasielastic response close to 0 meV, suggesting a magnetic ground state.

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Strongly correlated electron materials, especially the high temperature (T_c) cuprate superconductors [1] and the heavy fermion rare earth and actinide compounds [2], have received considerable attention recently because they display novel thermodynamic, magnetic, and transport properties that cannot be described by conventional Fermi-liquid theory. While the microscopic origin of the non-Fermi-liquid (NFL) behavior in the high T_c cuprates remains unknown, progress has been made in understanding some of the heavy fermion compounds. In particular, the suppression of long-range magnetic or spin glass order to $T = 0$ has been suggested as the source of the NFL behavior in $CeCu_{5.9}Au_{0.1}$ [3] and $UCu_{3.5}Pd_{1.5}$ [4], although the origin of such a novel quantum phase transition remains obscure. On the other hand, there has been considerable debate about the microscopic origin of the NFL behavior in the $Y_{1-x}U_xPd_3$ system (for $x \leq 0.2$), which has the cubic Cu_3Au structure ($0 \leq x \leq 0.5$) [5,6]. Three conceptionally very different microscopic origins have been invoked: (1) Using a model proposed by Cox [7], Seaman *et al.* [5] explained their data on the basis of a two channel quadrupolar Kondo effect arising from the interaction between the conduction electrons and the electric quadrupole moment of a non-Kramers-doublet ground state of U in the crystalline electric field (CEF). (2) Based on the observed scaling behavior in the magnetic field and the temperature dependence of the specific heat and magnetization, a novel second order phase transition at $T = 0$ K was suggested [6]. When $T \rightarrow 0$ K, the long-range correlations between U atoms on the verge of magnetic order might lead to a breakdown of the Fermi-liquid description [8]. (3) Dilute magnetic impurities in a disordered metal might, at the proximity of a metal-insulator transition, suppress the Kondo temperature T_K sufficiently such that Fermi-liquid behavior expected for a conventional Kondo alloy never appears [9].

In this Letter, we present neutron scattering results for the $Y_{1-x}U_xPd_3$ system ($x = 0.2, 0.45$) which set significant constraints on a possible microscopic theory for the observed NFL behavior. In particular, we show that the $Y_{0.55}U_{0.45}Pd_3$ compound, previously thought to be a spin glass [5,10], orders antiferromagnetically with a moment of $(0.7 \pm 0.2)\mu_B$ per U atom and a magnetic structure identical to that of UPd_4 [11], which also has the $CuAu_3$ structure. The $Y_{0.8}U_{0.2}Pd_3$ compound, on the other hand, does not order above 0.2 K. However, critical fluctuations associated with antiferromagnetic ordering were observed on cooling from 77 to 0.2 K. The magnetic excitation spectra for both compounds, obtained by polarized inelastic neutron scattering (INS), were dominated by a quasielastic response close to 0 meV, suggesting a magnetic ground state.

The experiments were performed with the HB-1A, HB1, and HB3 triple-axis spectrometers at the High Flux Isotope Reactor at Oak Ridge National Laboratory. For elastic measurements, a pyrolytic-graphite (PG) monochromator and analyzer were used with a fixed final energy of either $E_f = 14.78$ or 13.5 meV and a collimation of 50′-40′-40′-60′ in the usual notation. PG filters eliminated higher order contamination of the beam. Previous INS studies on the $Y_{1-x}U_xPd_3$ system by different groups [12,13] using unpolarized neutrons have yielded different results. In particular, a large intensity from phonon and nuclear spin incoherent (NSI) scattering has prevented an unambiguous determination of the ground state of the U ions in the cubic CEF, which is critical in testing the validity of the quadrupolar Kondo scenario [7]. To avoid these problems, we have performed INS using the polarized beam on HB1 with the setup described previously [14]. For polarized beam scans it is often sufficient to measure the spin-flip scattering (SF) and the non-spin-flip scattering (NSF). In cases where the magnetic signal is on top of a large NSI background, the SF scattering can be measured with neutron polarization first parallel to the momentum transfer (horizontal guide field or HF) and then perpendicular (VF). The difference (HF-VF) will then give one-half of the magnetic intensity completely free of background effects [15].

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The samples used in the experiments were polycrystalline ingots of $Y_{0.55}U_{0.45}Pd_3$ and $Y_{0.8}U_{0.2}Pd_3$ (total mass between 28 and 45 g each) prepared at UCSD. X-ray diffraction patterns for both samples show single phase Cu_3Au structure. However, electron microprobe measurements revealed fluctuations in uranium concentration on a length scale of ~ 10 μm, similar to those reported by Süllow *et al.* [16]. The samples were crushed into small pieces and mounted on the cold fingers of Displex helium refrigerators in aluminum cans filled with helium exchange gas. For measurements below 3 K, a dilution refrigerator with a copper sample can was used. In our measurements, momentum transfers are expressed in reciprocal-lattice units (rlu), where $\mathbf{a}^* = \mathbf{b}^* = \mathbf{c}^* = 2\pi/a = 1.54$ Å^{-1} and the horizontal scattering plane was arbitrarily defined to coincide with (h, h, l) zones of the crystal for notation convenience.

Earlier thermodynamic [5] and muon spin relaxation (μSR) [10] studies of $Y_{1-x}U_xPd_3$ have indicated that the samples with U concentrations $0.3 \leq x \leq 0.5$ exhibit spin-glass (SG) freezing below a temperature T_{SG} which increases monotonically with x. In view of the fact that none of these techniques are structurally sensitive, we have performed elastic and inelastic scattering measurements on $Y_{0.55}U_{0.45}Pd_3$ which was thought to have a T_{SG} of 21 K [5]. We found that the compound orders antiferromagnetically. Figure 1 shows the elastic $(h, h, 0)$ scans at two different temperatures which probe the static correlations ($\hbar\omega = 0$). The scans are featureless at 30 K while there are well-defined maxima at (0.5,0.5,0) below 21 K. Further polarized beam measurements confirm that this reflection is SF in nature. In addition to $(0.5, 0.5, 0)$, we observed the $(0.5, 0.5, 1)$ reflection. The magnetic unit cell of $Y_{0.55}U_{0.45}Pd_3$, therefore, doubles the chemical one in the $(h, h, 0)$ plane and repeats it along the $(0, 0, l)$ direction. This structure is identical to that of UPd_4, which has an ordered moment of $(0.8 \pm 0.1)\mu_B$ [11].

To determine whether the correlation is truly long range, we have measured the (0.5,0.5,0) reflection with several collimation configurations and a neutron energy of 14.78 meV. The final result is shown in Fig. 1(b). The elastic peak has a width (FWHM) of 0.007 ± 0.0003 rlu; the corresponding resolution is 0.0066 ± 0.0002 rlu as measured by $\lambda/2$ scattering from the nuclear (1,1,0) Bragg reflection at 30 K. Therefore the magnetic peak width is resolution limited to within the error of the measurements, and the minimum coherence length for the static correlations is 400 Å. The ordered U moment, obtained by normalizing the magnetic peak intensities at 4.4 K with respect to the weak nuclear reflection (1,0,0), is $(0.7 \pm 0.2)\mu_B$. This result is consistent with $\mu \simeq 1\mu_B$ for $Y_{0.6}U_{0.4}Pd_3$ estimated by μSR measurements [10].

Having established the magnetic structure of $Y_{0.55}U_{0.45}Pd_3$, we now focus on the temperature dependence of the order parameter and magnetic fluctuations. Earlier bulk susceptibility measurements at this composition reveal irreversible behavior between

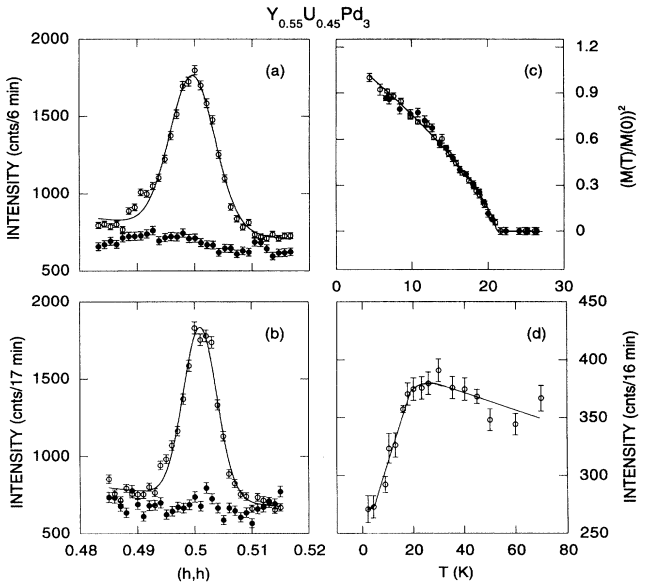

FIG. 1. (a) Elastic scattering with a PG filtered beam and 40′-20′-20′-60′ collimation at 10 K (∘) and 30 K (•). (b) Identical scans using 40′-10′-20′-60′ collimation. Solid lines are Gaussian fits to the data. (c) Reduced magnetization vs temperature on warming up (∘) and cooling down (•), obtained by subtracting the background and normalizing the data to the intensity at 4.4 K. (d) Temperature dependence of the inelastic scattering at $Q = (0.49, 0.49, 0)$ rlu and $\hbar\omega = 0.83$ meV. The lines are guides to the eye.

magnetic field cooling and zero field cooling below 21 K. To test whether the order parameter has hysteresis, we measured the magnetic elastic (0.5,0.5,0) peak intensity as a function of temperature at zero field. Figure 1(c) shows that the results are identical for warming up (open circles) and cooling down (solid circles), consistent with a second order phase transition. The magnetization vs temperature, described by $[M(T)/M(0)]^2 \sim (1 - T/T_N)^{0.7}$ with $T_N = 21.1 \pm 0.1$ K, is shown as a solid line in Fig. 1(c). Figure 1(d) displays the temperature dependence of the magnetic fluctuations, measured at Q offset slightly from $(0.5, 0.5, 0)$ to eliminate Bragg contamination and a neutron energy loss of $\hbar\omega = 0.83$ meV. As expected, the fluctuations show a maximum at T_N and then decrease drastically for $T < T_N$ as the system becomes progressively more ordered [17].

Although it is interesting to study $Y_{0.55}U_{0.45}Pd_3$, more important questions, such as the ordering status and the CEF energy level scheme of the U $5f$ electrons, need to be answered for the $Y_{0.8}U_{0.2}Pd_3$ compound in order to understand the microscopic origin of the observed NFL behavior. To this end, we have performed elastic and inelastic measurements. Figure 2(a) shows the difference obtained between a Q scan of an integral of $S(Q, \omega)$ measured using the two-axis mode at 1.25 K with a neutron energy of 13.5 meV and the same scan taken at 77 K where the magnetic fluctuations spread throughout the zone. It is clear from this figure that magnetic

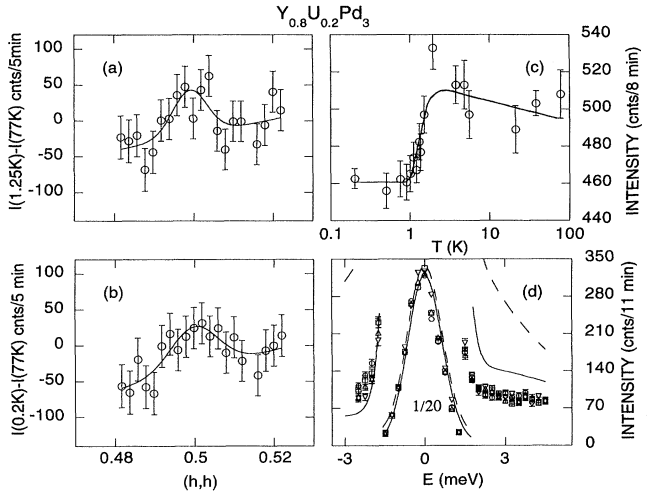

FIG. 2. (a) Q dependence of the integral of $S(Q,\omega)$ (between 0 and 13.5 meV) in which data obtained at 77 K are subtracted from 1.25 K data. (b) Differences between 0.2 and 77 K. (c) Temperature dependence of the scattering at (0.49, 0.49, 0) rlu and $\hbar\omega = 0.5$ meV. (d) Constant-Q scans at $Q = (0.5, 0.5, 0)$ rlu with $E_i = 14.586$ meV and $T = 70$ K (∘), 50 K (□), 30 K (△), and 10 K (▽). Solid lines in (a)–(c) are guides to the eye.

fluctuations develop at (0.5, 0.5, 0) rlu as the temperature is lowered from 77 to 1.25 K [18]. Figure 2(b) displays similarly subtracted data between 0.2 and 77 K, which again shows a peak at (0.5,0.5,0) rlu. Elastic scans indicate that no magnetic order exists above 0.2 K.

To determine further the temperature dependence of the magnetic fluctuations, we have measured the scattering intensity at (0.49, 0.49, 0) rlu with neutron energy loss of $\hbar\omega = 0.5$ meV. Figure 2(c) displays the results which reveal a clear suppression of the 0.5 meV fluctuations, indicating that the characteristic fluctuation energy is less than 0.5 meV below 2 K. For a conventional magnetic Kondo metal, one expects the quasielastic form $\chi'' \sim \omega\Gamma/(\omega^2 + \Gamma^2)$ with $\Gamma = \Gamma_0 + aT^{1/2}$, where Γ_0 is the characteristic energy scale at $T = 0$ K [19]. Therefore, if the observed logarithmic resistivity increase with decreasing temperature in $Y_{0.8}U_{0.2}Pd_3$ [5] is due to a magnetic Kondo effect, one would expect quasielastic scattering on an energy scale of $k_B T_K$ (i.e., $T_K = 42$ K $\sim$ 3.6 meV), and the width of the quasielastic scattering should vary as a function of temperature. Figure 2(d) shows $S(Q,\omega)$ at several temperatures. The solid and dashed lines in the figure represent the resulting theoretical form, evaluated by using $\Gamma = 3.6$ meV and a resolution-limited Gaussian peak to account for the nonmagnetic scattering estimated from polarization analysis, at 10 and 70 K, respectively. As we can see from the figure, a conventional magnetic Kondo effect clearly cannot describe our data, which show little quasielastic scattering beyond 2 meV and no temperature dependence. For the quadrupolar Kondo effect [7] to be responsible for the NFL behavior, the nonmagnetic Γ_3 doublet has to be the ground state for U^{4+} ions in the cubic CEF of the host YPd_3 [5,12]. Below, we present polarized beam measurements for both compounds, which elucidate the nature of the ground state.

Figure 3 summarizes the results for $Y_{0.55}U_{0.45}Pd_3$, obtained with polarized neutrons on HB1. Frame (a) displays the SF scattering with HF at 10 K, which shows clear CEF peaks at 5 and 39 meV [20]. Frame (b) is SF scattering using HF and VF with a lower instrument resolution to boost intensity. The subtraction of VF from HF, shown in frame (c), gives one-half the magnetic signal [15]. The important result, which is difficult to obtain with unpolarized neutrons, is that the dominating magnetic feature peaks at $\hbar\omega = 0$. According to Lea *et al.* [21], the ground state of U (assuming a $5f^2$ electron configuration with $J = 4$) ions in a cubic CEF can be either a Γ_1 singlet, a Γ_3 nonmagnetic doublet, or a Γ_5 triplet. The observation of a narrow resolution-limited magnetic peak at $\hbar\omega = 0$ already indicates a Γ_5 magnetic ground state if we assume that the quasielastic scattering due to coupling of the nonmagnetic ground state to excited states is small [2,7], as Γ_1-Γ_1 or Γ_3-Γ_3 transitions are disallowed [22]. To obtain a quantitative comparison with the data, we have calculated $S(Q,\omega)$ by normalizing the cross section to the magnetic intensity at $\hbar\omega = 0$ [23], using the appropriate scattering law [24]. Assuming a Γ_5 ground state, and Γ_3 and Γ_4 excited states at 5 and 39 meV, we obtain the solid lines in Figs. 3(a) and 3(c) by using appropriate transition probabilities [22] and thermal population factors. It is clear that a Γ_5 ground state

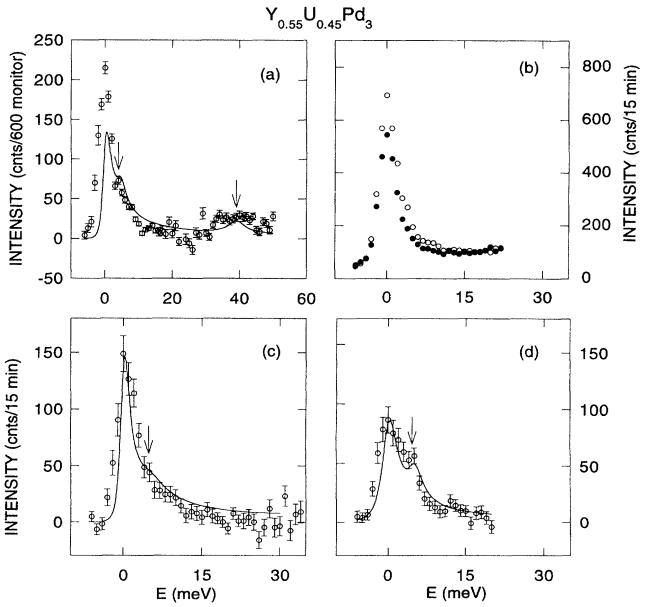

FIG. 3. (a) SF scattering with background (measured with analyzer turned off) subtracted using HF at $T = 10$ K. Data were taken at $Q = (0, 0, 1.2)$ rlu for $\hbar\omega \leq 24$ meV and at $Q = (0, 0, 1.7)$ rlu elsewhere. (b) SF scattering at 10 K using HF (∘) and VF (•). Error bars in the data are less than the size of the symbol. (c) SF scattering, HF-VF, at $T = 10$ K and (d) at 30 K. Solid lines are calculations discussed in the text.

is consistent with the data. To prove further that the ground state is consistent with Γ_5, we made polarized, HF-VF measurements at 30 K shown in Fig. 3(d). For a $\Gamma_5 = 0$ meV ground state and a $\Gamma_3 = 5$ meV first excited state, we expect the population of the Γ_5 and Γ_3 states to change from 99.5% and 0.5% at 10 K to 87.3% and 12.7% at 30 K, thereby reducing the $\Gamma_5 \rightarrow \Gamma_5$ ($\hbar\omega = 0$), $\Gamma_5 \rightarrow \Gamma_3$ ($\hbar\omega = 5$), and $\Gamma_5 \rightarrow \Gamma_4$ ($\hbar\omega = 39$) transition intensities. On the other hand, for a Γ_3 ground state, we would expect an increase in intensity at $\hbar\omega = 0$ due to the population increase in Γ_5. As we can see from the solid line in frame (d), obtained by repeating the CEF scheme calculation of Fig. 3(c) at $T = 30$ K, our data clearly favor a Γ_5 ground state for $Y_{0.55}U_{0.45}Pd_3$.

To complete our understanding of the $Y_{1-x}U_xPd_3$ system, we also made polarized measurements on $Y_{0.8}U_{0.2}Pd_3$. In contrast to $Y_{0.55}U_{0.45}Pd_3$, the magnetic signals from $Y_{0.8}U_{0.2}Pd_3$ are much weaker. Figure 4 summarizes the results. It is clear from the figure that the magnetic spectrum is still dominated by a peak at $\hbar\omega = 0$, identical to that of the $Y_{0.55}U_{0.45}Pd_3$ compound. Unpolarized neutron scattering data indicate that, in addition to the CEF peaks at $\hbar\omega = 0, 5$ meV, there is a CEF peak at 39 meV. Therefore we conclude that the CEF scheme of $Y_{0.8}U_{0.2}Pd_3$ is identical to that of the $Y_{0.55}U_{0.45}Pd_3$ compound, but with the magnetic signal strongly suppressed.

In conclusion, we have shown that $Y_{0.55}U_{0.45}Pd_3$ is an antiferromagnet with a Néel temperature of 21 K and an ordered moment of $(0.7 \pm 0.2)\mu_B$. The ground state of the U ions in the cubic CEF is consistent with a magnetic Γ_5 triplet description. Although $Y_{0.8}U_{0.2}Pd_3$ does not order above 0.2 K, critical fluctuations around the antiferromagnetic position were observed on cooling from 77 to 0.2 K. The magnetic spectrum was similar to that of the $Y_{0.55}U_{0.45}Pd_3$ compound. No quasielastic scattering $\hbar\omega \sim 3.6$ meV was observed. These results put severe constraints on the possible microscopic theory for the observed NFL behavior.

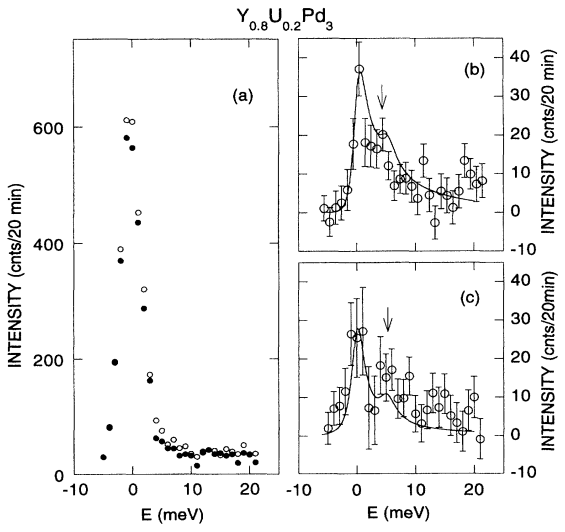

FIG. 4. (a) Constant-Q scans at $Q = (0, 0, 1.2)$ rlu and $T = 10$ K using HF (○) and VF (●). Data shown have the background subtracted. (b) SF scattering, HF-VF, at $T = 10$ K and (c) at 30 K. Solid lines are calculations discussed in the text.

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