Interplay between magnetism and non-Fermi-liquid behavior in $Sc_{1-x}U_xPd_3$

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Low-temperature electrical resistivity $\rho(T)$ and specific heat $C(T)$ measurements on $Sc_{1-x}U_xPd_3$ for $0.2 \leqslant x \leqslant 0.35$ are reported, demonstrating non-Fermi-liquid temperature dependence. For temperatures $0.05\ \mathrm{K} \leqslant T \leqslant 2.5\ \mathrm{K}$, the resistivity follows $\rho(T)=\rho(0)[1-a(T/T_K)^n]$, with n decreasing from $n=1.3$ at $x=0.2$ to $n=0.5$ at $x=0.35$. The electronic specific heat $\Delta C(T)$ follows $\Delta C(T)/T=-(b\mathrm{R}/T_K)\ln(b'T/T_K)$ above the spin-glass freezing temperature, with values of T_K that decrease with increasing x, in accord with the Fermi-level tuning phenomenon previously observed in this system. Applied magnetic fields produce an increase in the resistivity of samples with $x<0.3$ at the lowest temperatures.

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A decade has passed since experiments[1,2] on the strongly correlated *f*-electron system $Y_{1-x}U_xPd_3$ inspired worldwide interest in materials that appear to deviate from the ''standard model'' of condensed-matter physics appropriate at very low temperatures, Landau's Fermi-liquid (FL) theory. The observation of weak power-law and logarithmic divergences in the temperature dependence of the magnetic susceptibility, the specific heat divided by temperature, and the electrical resistivity at low temperatures in the $Y_{1-x}U_xPd_3$ system motivated a search for further examples of this so-called non-Fermi-liquid (NFL) behavior. The proliferation of new systems that exhibit NFL behavior attests to the widespread occurrence of deviations from FL theory.

In the initial work of Seaman *et al.*,[1] the NFL behavior of $Y_{1-x}U_xPd_3$ was attributed to a two-channel quadrupolar Kondo effect (QKE). The observation of a logarithmic divergence in the electronic specific heat divided by temperature

$$\Delta C(T)/T=-(b\mathrm{R}/T_K)\ln(b'T/T_K), \tag{1}$$

and a nearly linear electrical resistivity

$$\rho(T)=\rho(0)[1-a(T/T_K)^n], \tag{2}$$

with $n\approx 1$ were in agreement with the rough predictions of the QKE model, which identifies T_K with the Kondo temperature. Exact conformal field theory calculations[3] later showed that $\rho(T)$ should diverge as Eq. (2) with $n=0.5$ in the two-channel QKE model. This $\sqrt{T}$ divergence is expected to be observable below $0.05T_K$, while in the range $0.05T_K \leqslant T \leqslant T_K$, $\rho(T)$ should be approximately linear in temperature.[4,5] In contrast, $\rho(T)$ in a single-channel, spin-$\frac{1}{2}$ Kondo effect should saturate as T^2 for $T \ll T_K$, consistent with FL theory. Subsequent studies of $Y_{1-x}U_xPd_3$ demonstrated[6] that $\rho(T)$ is nearly linear for $x=0.1$, 0.15, and 0.2 for $T>0.1$ K ($T/T_K \sim 0.07\%$, 0.1%, and 0.2%, respectively). Because of the lack of agreement with the resistivity prediction, it is possible that the QKE model is not the appropriate description of $Y_{1-x}U_xPd_3$. However, the resistivity was found to scale with x and T_K, which suggests that the origin of NFL behavior in this system is a single-ion mechanism, such as a multichannel Kondo effect.

One requirement for the QKE model is a nonmagnetic ground state. The ground state of $Y_{1-x}U_xPd_3$ has been investigated by several groups by means of neutron-scattering experiments.[7–9] According to Lea, Leask, and Wolf,[10] the U^{4+} ninefold degenerate $J=4$ Hund's rule multiplet is split in the presence of a cubic crystalline electric field (CEF) into Γ_4 and Γ_5 triplets, a Γ_1 singlet, and a Γ_3 doublet. Initially, the ground state of $Y_{0.8}U_{0.2}Pd_3$ was determined to be a nonmagnetic Γ_3, with magnetic Γ_5 and Γ_4 excited states at 5 and 16 meV, respectively. The inelastic neutron-scattering spectra exhibited a small quasielastic line width $\Delta/2 \lesssim 0.1$ meV that is significantly smaller than the $k_BT_K \sim 4$ meV value expected for a magnetic Kondo effect.[7] Another experiment[8] corroborated the identification of the ground state as Γ_3, but suggested slightly different excited states. These results were interpreted as further evidence for a QKE in $Y_{1-x}U_xPd_3$. In contrast, polarized inelastic neutron-scattering measurements[9] indicated a different arrangement with a magnetic Γ_5 as the ground state for $x=0.45$ and possibly for $x=0.2$, which would preclude the existence of a QKE.

Considerable debate about the origin of NFL behavior has raised the question of whether the physical properties of the polycrystalline samples are intrinsic electronic properties or extrinsic properties associated with metallurgical inhomogeneity. The $Y_{1-x}U_xPd_3$ system has been shown[11,12] to possess inhomogeneities in the U concentration on micron length scales. A recent investigation found uranium in $Sc_{1-x}U_xPd_3$ to be much more homogeneously distributed than in $Y_{1-x}U_xPd_3$ and demonstrated that the low-temperature physical properties of these two systems are quite similar despite significant differences in their metallurgy.[13] The $Sc_{1-x}U_xPd_3$ system has nearly the same T vs x phase diagram as that of $Y_{1-x}U_xPd_3$ (see Ref. 13), with a slightly higher value of T_K for a given value of x.[14]

In this paper, we present measurements of the electrical resistivity and specific heat of $Sc_{1-x}U_xPd_3$ for concentrations $0.2 \leqslant x \leqslant 0.35$, motivated by the substantially cleaner metallurgy of this system. Our results show NFL behavior characterized by a logarithmic divergence in $C(T)/T$ and an electrical resistivity that varies as Eq. (2) with $n=0.5$ for $x=0.3$ and 0.35. In conjunction with the results of Ref. 14, these results represent an instance in which the temperature dependences of $\rho(T)$, $\chi(T)$, and $C(T)/T$ of a material are

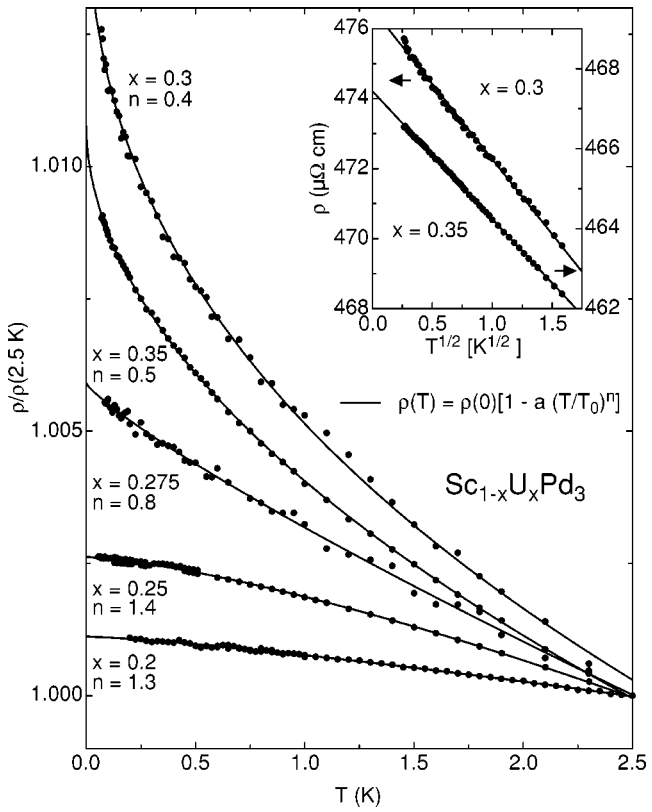

FIG. 1. Normalized electrical resistivity $\rho(T)/\rho(2.5\ \mathrm{K})$ of $Sc_{1-x}U_xPd_3$ for $0.2 \leqslant x \leqslant 0.35$ vs temperature T. Here $T_0 \equiv T_K$. The resistivity for $x=0.3$ and 0.35 is plotted vs $\sqrt{T}$ in the inset.

all consistent with the QKE and suggest that the QKE may be responsible for NFL behavior in $Sc_{1-x}U_xPd_3$.

New polycrystalline samples were prepared as described previously.[13] Powder-x-ray diffraction measurements confirmed that the samples had the cubic Cu_3Au structure with no trace of impurity phases. Electrical resistivity measurements were made in a transverse geometry using ^{3}He-^{4}He dilution refrigerators at UCSD and at the NHMFL-LANL and a Linear Research LR 700 four-wire ac resistance bridge operating at 12 Hz. Specific heat measurements were performed using a UCSD-built semiadiabatic heat-pulse calorimeter.

Normalized electrical resistivity data for $Sc_{1-x}U_xPd_3$ with $0.2 \leqslant x \leqslant 0.35$ for temperatures below 2.5 K are displayed in Fig. 1. The resistivities of all of the samples increase with decreasing temperature on this scale, consistent with the behavior of samples with $x=0.2$ and 0.3 measured earlier[14] at temperatures above 1.2 K. The solid lines are fits of Eq. (2) to the data and the values of n extracted from these fits are listed below each concentration. The value of the exponent n decreases from $n=1.3$ for the sample with $x=0.2$ to $n=0.4$ for $x=0.3$. It is interesting to note that this power law is apparent only below ~ 1 K. Previous resistivity measurements[14] above 1.2 K could be described quite well by Eq. (2) with a value of $n \approx 1$. The inset shows the resistivity for samples with $x=0.3$ and 0.35 plotted vs $\sqrt{T}$ as well as straight lines representing a $\sqrt{T}$ temperature dependence that describe the data quite well over the entire temperature range of the measurements.

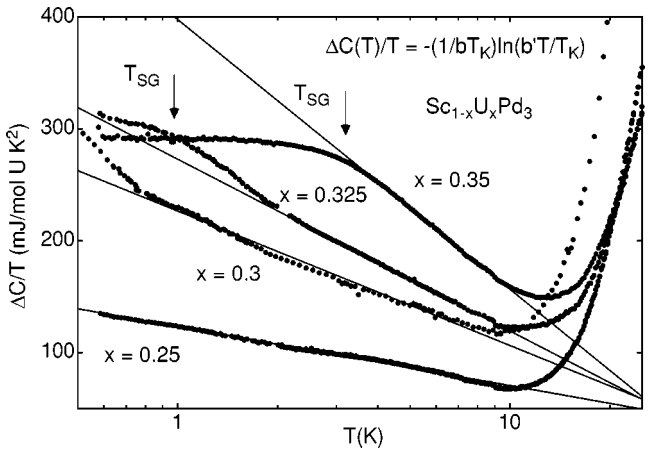

FIG. 2. Uranium-ion contribution to the specific heat $\Delta C(T)/T$ vs temperature T on a semilogarithmic scale for $Sc_{1-x}U_xPd_3$ with $0.25 \leqslant x \leqslant 0.35$.

The uranium-ion contribution to the specific heat of $Sc_{1-x}U_xPd_3$ for $T \leqslant 25$ K is shown in Fig. 2 plotted on a logarithmic temperature scale for $0.25 \leqslant x \leqslant 0.35$. The data for $Sc_{0.7}U_{0.3}Pd_3$ are from a previous experiment[14] and are shown for comparison. The value of $\Delta C(T)$ was calculated by subtracting an estimated value of the phonon contribution βT^3 from the measured value of $C(T)$. Values of β for each compound were determined by scaling the literature value[15] of β for $ScPd_3$ by the molecular weight of each compound. The upturns in $\Delta C(T)/T$ for $T > 10$ K are most likely due to either an incorrect subtraction of the phonon contribution or a Schottky anomaly associated with CEF energy levels. For $Sc_{0.75}U_{0.25}Pd_3$, the data are described well by a logarithmic temperature dependence, indicated in the figure by the solid line, between 10 K and 0.6 K, the lowest temperature of the measurement. As x is increased, the region over which the logarithmic temperature dependence describes the data becomes smaller, presumably due to the onset of spin-glass (SG) freezing that occurs at increasingly higher temperatures. The slopes of the logarithmic regions increase due to a decrease in the Kondo temperature T_K. The T_K values calculated from the fits decrease nearly exponentially with increasing x from $T_K=91$ K for $x=0.25$ to $T_K=20$ K for $x=0.35$ and are in good agreement with values of T_K from previous $\rho(T)$ measurements[14] above 1.2 K. The arrows, labeled T_{SG}, identify features in the $\Delta C(T)/T$ data that we attribute to the onset of SG freezing. In $Sc_{0.65}U_{0.35}Pd_3$, the feature in the data associated with SG freezing occurs at $T_{SG} \approx 3$ K, close to the irreversibility temperature $T_{irr} \approx 2.6$ K determined by dc magnetic susceptibility measurements.[16] The data for $Sc_{0.675}U_{0.325}Pd_3$ deviate from the fit at temperatures slightly above T_{SG}, possibly due to the onset of magnetic fluctuations. A similar deviation occurs for $Sc_{0.7}U_{0.3}Pd_3$ and comparable behavior is also observed in $Y_{1-x}U_xPd_3$.

Applied magnetic fields cause an increase in the electrical resistivity at low temperatures as shown in Fig. 3 for $Sc_{0.75}U_{0.25}Pd_3$. In the main figure, the resistivity is displayed for magnetic fields up to 50 kOe and at temperatures below 2.5 K. The solid lines are fits of Eq. (2) to the data for

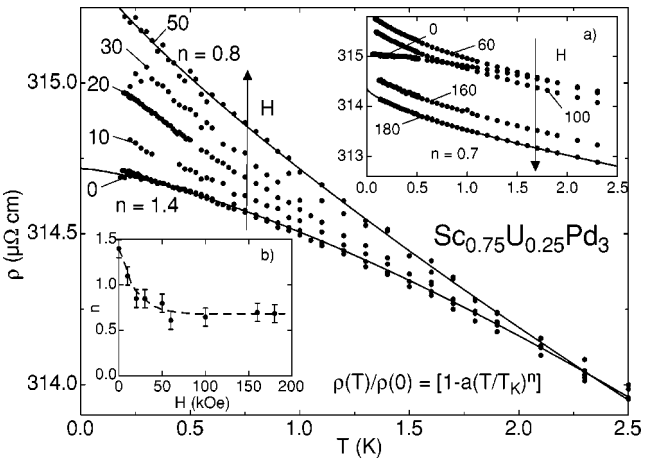

FIG. 3. Electrical resistivity $\rho(T)$ of $Sc_{0.75}U_{0.25}Pd_3$ vs temperature T in applied magnetic fields 0 kOe $\leqslant H \leqslant$ 50 kOe. The arrows in the main panel and in inset (a) indicate the direction of increasing magnetic field. The upper inset (a) shows the resistivity vs temperature for applied magnetic fields 0 kOe $\leqslant H \leqslant$ 180 kOe. The lower inset (b) shows the magnetic field H dependence of the exponent n derived from fits of the data in the main figure and in inset (a) to Eq. (2).

$H=0$ and 50 kOe. In zero field, the resistivity varies as Eq. (2) with $n=1.4$. As the field is increased, the resistivity at the lowest temperature increases such that for an applied field of 50 kOe, n is decreased to $n=0.8$. The resistivity in larger applied fields is shown in the upper inset (a) for fields up to 180 kOe. The resistivity is found to increase at the lowest temperatures in fields up to 60 kOe with negligible magnetoresistance above ~ 1.5 K. For $H>60$ kOe, a small negative magnetoresistance is observed over the entire temperature range of the measurement that shifts the resistivity curves down nearly parallel to the curve for 60 kOe. The power-law fits to the resistivity data for $H>60$ kOe are quite similar as demonstrated by the solid line in the inset which is a fit of Eq. (2) to the data for $H=180$ kOe. The exponent calculated from this fit, $n=0.7$, is very close to the value that was obtained from the fit of the data measured in the 50 kOe field. The lower inset (b) shows the H dependence of the exponent n derived from the fits in the main figure and in inset (a). The value of n decreases rapidly for applied fields up to 50 kOe and then is relatively constant up to 180 kOe. It should be noted that the resistivity in applied magnetic fields showed no sign of crossing over to the $n=2$ temperature dependence of a Fermi liquid.

Qualitatively similar behavior for $x=0.2$ and 0.275 was also observed in the presence of magnetic fields where n was seen to decrease with increasing field. However, the effect of the magnetic field was different for the $x=0.3$ and 0.35 samples for which the resistivity did not increase with field at low temperatures as shown in Fig. 3. Instead, a negative magnetoresistance was observed for all applied fields similar to what was observed in the $x=0.25$ sample for fields above 60 kOe.

The two-channel QKE model has several attributes that make it an attractive candidate to describe the physics of both $Sc_{1-x}U_xPd_3$ and $Y_{1-x}U_xPd_3$. Previous work has demonstrated that the predictions of the QKE model for the specific heat [Eq. (1)] and the magnetic susceptibility are in good agreement with the behavior observed in $Y_{1-x}U_xPd_3$ and $Sc_{1-x}U_xPd_3$.[13,17,18] Values of T_K determined from low-temperature fits to the specific heat, electrical resistivity, and magnetic susceptibility of $Y_{1-x}U_xPd_3$ are also in good agreement with the values of T_K found from electrical resistivity and magnetic susceptibility measurements at temperatures above 4 K.[6] This agreement, together with the fact that the low-temperature properties were found to scale with x,[6] provides compelling evidence that a single-ion mechanism is responsible for the NFL physics. It is thus appealing to attribute the $\sqrt{T}$ dependence of $\rho(T)$ demonstrated in Fig. 1 to the QKE model. In fact, the existence of a $\sqrt{T}$ dependence in $\rho(T)$ at temperatures below 2 K in the sample with $x=0.3$ is in excellent agreement with Cox and Mackivic's prediction[4] since 5% of $T_K \approx 40$ K is 2 K.

There are a few aspects of the $Sc_{1-x}U_xPd_3$ system that at first examination seem to preclude the existence of the QKE effect. The existence of a spin-glass state for the sample with $x=0.35$ below ~ 4 K, coexisting with the NFL behavior, would seem to be at odds with a single-ion model. However, inhomogeneities inherent due to the substituted nature of this system could result in some U ions clustering to form a spin glass whereas others are isolated and participate in single-ion NFL behavior. Second, a single-ion model such as the QKE implies that the physical properties should have the same temperature dependence as the uranium concentration is varied. In contradiction to this basic premise of a single-ion model, our results indicate that n increases with decreasing x. This issue can be resolved by considering that the ground state of the $Y_{1-x}U_xPd_3$ system varies with x. Since the QKE model is consistent with our results for the samples with $x=0.3$ and 0.35, it is possible that the QKE model is only valid in a certain range of uranium concentrations. A crossover of the uranium-ion ground state in the presence of a CEF from a nonmagnetic ground state, for $x \geqslant 0.3$, to a magnetic ground state as x is decreased below $x=0.3$ could explain why the QKE is not observed at lower U concentration. The physical properties of the samples with the nonmagnetic ground state would be dominated by the QKE, while the properties of samples with x less than the crossover value would reflect a magnetic ground state and could evolve towards Fermi-liquid behavior.

Evidence of a ground-state crossover in $Y_{1-x}U_xPd_3$ was found in recent measurements[19] that imply Γ_3 is the ground state in only a certain U-concentration regime. The authors of Ref. 19 assigned the Γ_3 ground state to $Y_{0.55}U_{0.45}Pd_3$ and observed that the movement of a low-energy excitation in the neutron-scattering spectra of samples with $0.2 \leqslant x \leqslant 0.45$ towards zero-energy transfer with decreasing x suggests that the CEF parameters move toward the Γ_3–Γ_5 crossing point on the Lea, Leask, and Wolf diagram[10] for $J=4$. This observation led to the proposal that the Γ_3 and Γ_5 states are degenerate and coexist as the ground state for $x=0.2$. Furthermore, the authors concluded that a single-ion mechanism was the dominant cause of the magnetic properties in the $x=0.2$ compound. Additional evidence for a crossover in the

ground state of $Y_{1-x}U_xPd_3$ near $x=0.2$ is found in thermopower data[20] that change sign as U concentration is increased from $x=0.2$ to 0.3. Assuming that $Sc_{1-x}U_xPd_3$ is similar to $Y_{1-x}U_xPd_3$ in this respect, it is possible that a crossover in the CEF ground state for x slightly below $x=0.3$ could be responsible for the evolution in the value of n observed in Fig. 1. Inelastic neutron-scattering measurements of $Sc_{1-x}U_xPd_3$ would be required to test this hypothesis.

Remarkably, n is essentially the same for the samples with $x=0.35$ and 0.3, although the NFL behavior in the resistivity of the former occurs at temperatures well below T_{SG}, while nearly the same behavior occurs in the latter at temperatures for which there is no evidence of SG freezing. It is striking to note the proximity of the U concentration where T_{SG} extrapolates to $T=0$ K, determined by the $C(T)$ measurements, and the U concentration at which n decreases to $n\approx 0.5$. This might indicate a $T=0$ K phase transition, but it is possible that the extrapolation of T_{SG} is an indication of the U concentration where the magnetic Γ_5 ground state falls below the energy of the nonmagnetic Γ_3 as x is decreased. The prediction of the QKE model seems best fulfilled in $Sc_{1-x}U_xPd_3$ at the border of ($x=0.3$) and within ($x=0.35$) the SG regime, where excitations above a nonmagnetic ground state produce magnetism that coexists with non-Fermi-liquid behavior, similar to the behavior suggested for $Y_{1-x}U_xPd_3$ by μSR[21] and neutron scattering.[19]

The NFL behavior has previously been induced with the application of magnetic fields in the $CeCu_{6-x}Ag_x$ system.[22] The behavior observed in $Sc_{0.75}U_{0.25}Pd_3$, shown in Fig. 3, might be interpreted as the induction of NFL behavior by the application of a magnetic field, although the temperature exponent in zero field, $n=1.4$, is quite different from FL behavior with $n=2$. It seems likely that the exponent $n\approx 0.6$ observed for $H\geqslant 50$ kOe reflects the same NFL behavior as is seen in zero field for $x=0.3$ and 0.35. If a crossover to a magnetic ground state occurs near $x=0.3$, then the application of a magnetic field for samples with $x<0.3$ may cause a field-induced crossover in the CEF ground state from Γ_5 to Γ_3, although the mechanism for such a crossover is not readily apparent. For samples in which the ground state is magnetic in this scenario, the application of a magnetic field seems to induce an NFL temperature dependence that is similar to that observed in zero field in the samples with $x=0.3$ and 0.35, which presumably have a nonmagnetic ground state. This transformation does not occur in samples that already have a nonmagnetic ground state, and we observe a small negative magnetoresistance consistent with a nonmagnetic ground state typical of a QKE. The NFL temperature dependence observed in these materials seems to be quite robust in large magnetic fields as there is no evidence for a crossover to FL behavior.

While it is possible that applying a magnetic field to compounds with $x<0.3$ induces the same type of magnetism, characterized by $n\approx 0.5$, as is present in zero field in compounds with $x\geqslant 0.3$, we have no knowledge of a model that predicts such a unique temperature dependence in $\rho(T)$. Furthermore, $\rho(T)$ of samples with $x\geqslant 0.4$ actually decreases with decreasing temperature below either T_{SG} or T_N.[14] This behavior suggests that the effect of magnetism on $\rho(T)$ is actually quite different from the measured $n\approx 0.5$ temperature dependence shown in Fig. 1. A more detailed investigation of $\rho(T)$ of $Y_{1-x}U_xPd_3$ for $x>0.2$ will be required to determine if the resistivity of that system is different from what is observed in the present work on $Sc_{1-x}U_xPd_3$. It is currently unknown if samples of $Y_{1-x}U_xPd_3$ with $0.2\leqslant x\leqslant 0.3$ also exhibit an exponent near $n=0.5$ as x is increased towards the regime in which SG freezing is observed.

Several alternatives to single-ion mechanisms have been posited to explain NFL behavior in the $Y_{1-x}U_xPd_3$ system. An early proposal considered fluctuations of an order parameter near a second-order $T=0$ K phase transition.[2] Scenarios in which disorder leads to NFL behavior have more recently been suggested. The Kondo-disorder model, in which spatial fluctuations in T_K of local moments lead to NFL characteristics,[23] was shown to be an unlikely description of NMR measurements of $Y_{0.8}U_{0.2}Pd_3$.[24] The Griffiths-McCoy theory of NFL behavior, which attributes NFL properties to the formation of magnetic clusters due to system inhomogeneity and competition between Kondo and Ruderman-Kittel-Kasuya-Yosida interactions,[25] has been shown to be consistent with $C(T)$ and $\chi(T)$ measurements of $Y_{1-x}U_xPd_3$.[26] However, based upon recent measurements of $Sc_{1-x}U_xPd_3$, in particular, the single-ion scaling of the physical properties with x and the $T^{1/2}$ dependence of the electrical resistivity at low temperatures for samples with $x=0.3$ and 0.35, we submit an argument in favor of a single-ion description of the $Sc_{1-x}U_xPd_3$ and $Y_{1-x}U_xPd_3$ systems, with the QKE as the predominant cause of NFL behavior.

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