ARTICLES

Non-Fermi-liquid behavior and magnetic order in the $U_{1-x}Y_xPd_2Al_3$ system

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Measurements of the low-temperature electrical resistivity ρ, specific heat C, and magnetic susceptibility χ of the $U_{1-x}Y_xPd_2Al_3$ system reveal non-Fermi-liquid behavior for $x \gtrsim 0.7$. For $x = 0.8$, the low-temperature physical properties can be expressed as $\rho(T) = \rho(0)[1 - a(T/T_0)]$, $C(T)/T = -(R/bT_0)\ln(T/b'T_0)$ or $T^{-1+\lambda}$ ($\lambda = 0.82$), and $\chi(T) = \chi(0)[1 - c(T/T_0)^{1/2}]$ where $-a$, b, b', and c are positive constants and $T_0 \approx 30$ K. In addition, we observed concentration x-dependent features in the specific heat and magnetic susceptibility that appears to be due to magnetic order and extrapolate linearly to $T = 0$ K at $x \approx 0.7$. [S0163-1829(98)06147-5]

I. INTRODUCTION

The breakdown of the Fermi liquid (FL) paradigm in a certain class of strongly correlated *f*-electron materials has attracted considerable interest[1–3] during the past several years. The materials which exhibit non-Fermi-liquid (NFL) behavior are typically Ce- or U-based intermetallic compounds which are diluted by substituting a nonmagnetic element on either the rare earth/actinide or ligand sites.[1] In these materials, the NFL regime can be suppressed[4–7] or induced[4,6–8] through the application of hydrostatic pressure or applied magnetic fields. A variety of theoretical routes to the NFL regime have been proposed which are based on chemical disorder,[9,10] single or multichannel Kondo effects of magnetic or electric origin,[9–12] ordering of magnetic dipole or electric quadrupole moments,[13–17] or an interplay of these phenomena.[18]

Several years ago, NFL behavior was observed[1,19] in the $U_{1-x}Th_xPd_2Al_3$ system, where an increase in the tetravalent thorium concentration x suppresses the antiferromagnetic order found in the parent compound, UPd_2Al_3, revealing NFL behavior at thorium concentrations $x \gtrsim 0.4$. The substitution of Th reduces the Néel temperature only slightly before the feature in $C(T)$ associated with antiferromagnetic order disappears. In contrast, studies of the $U_{1-x}Y_xPd_2Al_3$ system in the range $0 \leq x \leq 0.5$ revealed that the substitution of trivalent Y for U suppresses the Néel temperature of UPd_2Al_3 much more rapidly than substitutions of tetravalent Th for U.[20] In this paper, we report measurements of the temperature T dependence of the electrical resistivity ρ, specific heat C, and magnetic susceptibility χ of the $U_{1-x}Y_xPd_2Al_3$ system in the concentration range $0 \leq x \leq 0.8$, within which single phase pseudoternary compounds could be prepared. The primary objective of this work was to determine whether NFL behavior occurs in the $U_{1-x}Y_xPd_2Al_3$ system at concentrations x above the critical concentration at which the magnetic ordering temperature vanishes, and to characterize the T dependences of ρ, C, and χ in the NFL regime.

II. EXPERIMENTAL DETAILS

The polycrystalline $U_{1-x}Y_xPd_2Al_3$ samples were prepared by arc melting the constituent elements on a copper hearth in an argon atmosphere and then annealing in argon for 7 days at 900 °C. X-ray powder diffraction measurements indicated that the samples consist of a single phase with the hexagonal $PrNi_2Al_3$ structure. The specific heat $C(T)$ was measured from 0.5 K to 20 K in a ^{3}He calorimeter using a semiadiabatic heat pulse technique. Magnetization M as a function of T and magnetic field H was measured from 1.8 to 300 K using a Quantum Design SQUID magnetometer in magnetic fields up to 1 T and from 0.4 to 2 K using a ^{3}He Faraday magnetometer in magnetic fields up to 5 T. The magnetic susceptibility $\chi(T)$ was obtained from the linear part of the M versus H isotherms at low fields $\mu H/k_B T \ll 1$, where μ is the magnetic moment and k_B is Boltzmann's constant. This procedure was especially important at low temperatures where the M versus H curves display negative curvature. The electrical resistivity $\rho(T)$ was measured by means of a standard four-wire ac technique (16 Hz) in the range $0.02 \leq T \leq 2.5$ K using a ^{3}He-^{4}He dilution refrigerator and in the range $1.7 \leq T \leq 300$ K in a ^{4}He cryostat.

III. RESULTS

Displayed in Fig. 1(a) are specific heat divided by temperature, C/T, versus T data on a logarithmic T scale between 0.5 and 20 K for $U_{1-x}Y_xPd_2Al_3$ samples with $x = 0.4$, 0.5, 0.6, 0.7, and 0.8. For $x = 0.7$ and 0.8, the $C(T)/T$ data are consistent with a logarithmic temperature dependence below ~ 4 K, which is characteristic of many other NFL systems.[1] For the samples with $x = 0.4$, 0.5, and 0.6, there is a kink in the C/T versus T curve at temperatures of 3, 2.2, and 1.2 K, respectively. For higher concentrations ($x = 0.7$, 0.8), there is no indication of the kink in C/T versus T above 0.5 K, the low temperature limit of the $C(T)$ measurements. Shown in Fig. 1(b) are χ versus T data for $U_{1-x}Y_xPd_2Al_3$ samples with $x = 0.1$, 0.2, 0.4, and 0.5 mea-
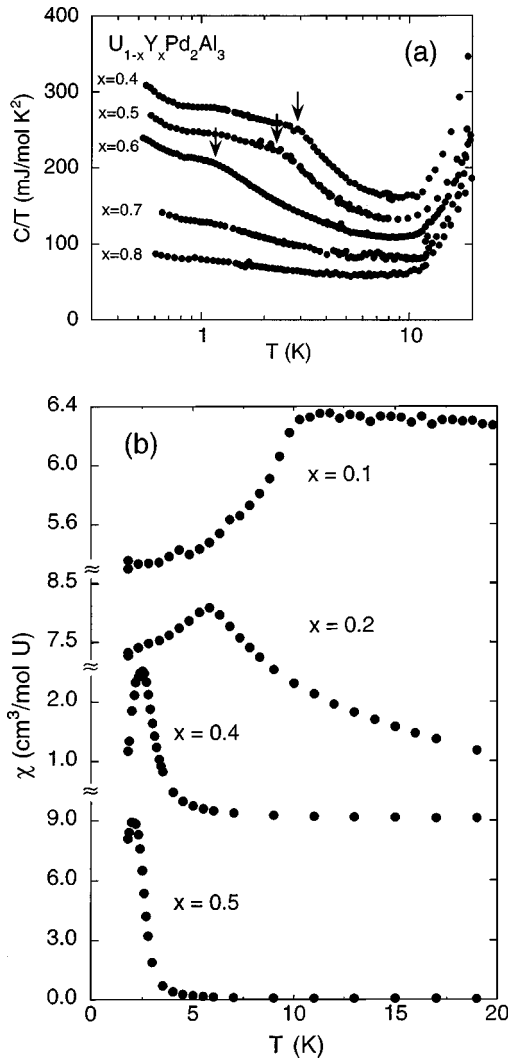
FIG. 1. (a) Specific heat divided by temperature C/T vs T on a logarithmic T scale of $U_{1-x}Y_xPd_2Al_3$ for several x values between 0.4 and 0.8. Arrows indicate the position of the kinks associated with the onset of magnetic order. (b) Magnetic susceptibility χ vs T of $U_{1-x}Y_xPd_2Al_3$ for several x values between 0.1 and 0.5 measured at 20 G. The data for $x=0.1$ and 0.2 have been multiplied by 1000.

sured in a field of 20 G where $\chi \equiv M/H$. These data reveal maxima in $\chi(T)$, apparently associated with antiferromagnetic order, at temperatures of 12, 6, 2.5, and 2 K, respectively, which are close to the temperatures of the kinks in the C/T versus T data. The Néel temperatures T_N inferred from the $C(T)$ and $\chi(T)$ data in Figs. 1(a) and 1(b) are indicated in the temperature T versus concentration x phase diagram for $U_{1-x}Y_xPd_2Al_3$ displayed in Fig. 2. It appears that as the concentration x of the Y substituent is increased, the antiferromagnetic transition that occurs at 14 K in the parent compound, UPd_2Al_3, systematically shifts to lower temperatures and vanishes near $x=0.7$. This scenario is consistent with the depression of the Néel temperature previously observed in the $U_{1-x}Y_xPd_2Al_3$ system for $x \lesssim 0.5$.[20,21] Neutron diffraction studies of the magnetic structure as a function of x are planned for a future investigation.

In order to characterize the physical properties in the NFL regime, we undertook a more extensive study of the
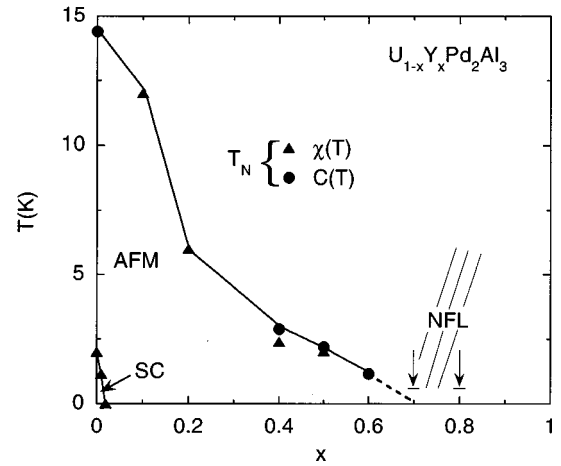

FIG. 2. Temperature T vs yttrium concentration x phase diagram indicating antiferromagnetically ordered (AFM) and non-Fermi-liquid (NFL) regimes based on measurements of the magnetic susceptibility and specific heat. The superconductivity (SC) data are from Geibel *et al.* (Ref. 21).

$U_{0.2}Y_{0.8}Pd_2Al_3$ sample. Shown in Fig. 3 is a plot of C/T versus $\log_{10} T$ for this sample where the data have been fitted to power-law (dashed line) and logarithmic (solid line) temperature dependences between 0.6 and 5 K. Both the power law $C(T)/T \propto T^{-1+\lambda}$ and logarithmic $C(T)/T \propto -\ln T$ behaviors describe the data quite well. Using the reduced chi square, χ_ν^2, to test the quality of the fits, we find $\chi_\nu^2=1.08$ for the power-law fit and $\chi_\nu^2=1.15$ for the logarithmic fit. Although the power law seems to describe the data slightly better according to the values of χ_ν^2, it is not possible to clearly establish which functional form provides the best description of the data.

The NFL behavior of the sample with $x=0.8$ was further characterized by $\rho(T)$ and $\chi(T)$ measurements. Displayed in Fig. 4 are ρ versus T data for $U_{0.2}Y_{0.8}Pd_2Al_3$ between 100 mK and 300 K. With decreasing temperature, $\rho(T)$ decreases nearly linearly with T from 300 K down to ~ 100 K, below which the slope, $d\rho/dT$, increases dramatically as the temperature is lowered. Shown in the inset are data for temperatures below 15 K with a linear fit of the resistivity repre-
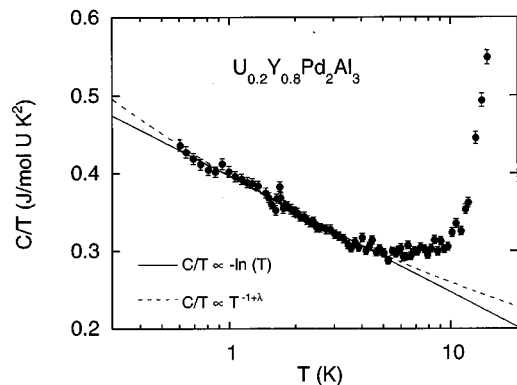

FIG. 3. Specific heat divided by temperature C/T vs T on a logarithmic T scale of $U_{0.2}Y_{0.8}Pd_2Al_3$. The solid and dashed lines represent fits of the data to power-law and logarithmic temperature dependences, respectively, over the same temperature range.
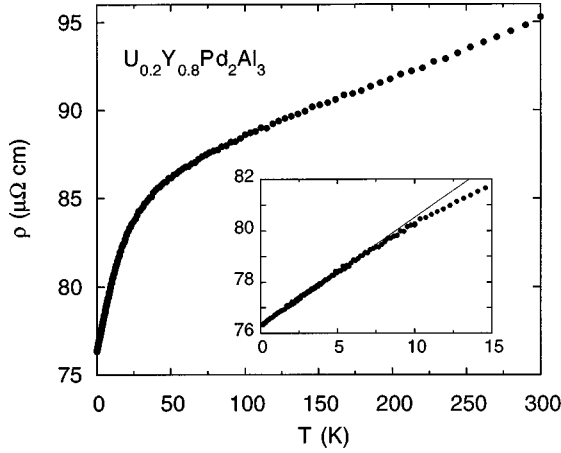
FIG. 4. The electrical resistivity ρ vs T of $U_{0.2}Y_{0.8}Pd_2Al_3$. The inset shows an enlarged view of the data for $T \leqslant 15$ K. The solid line is a linear fit to the low-temperature data.

sented by a solid line. The fit is good from the lowest measured temperature, 0.1 K, to ~7 K, which encompasses the temperature range over which NFL behavior was observed in the specific-heat measurement. The positive slope seen here is different from many other NFL systems such as $U_{1-x}Th_xPd_2Al_3$ and $Y_{1-x}U_xPd_3$,[1,2] which have a resistivity linear in temperature with a negative slope. A linear resistivity with a positive slope has been observed in a few NFL materials such as $U_{0.9}Th_{0.1}Be_{13}$ ($0.23 \leqslant T \leqslant 0.7$ K) (Ref. 5) and $CeCu_{5.9}Au_{0.1}$ ($0.02 \leqslant T \leqslant 0.5$ K).[22]

Shown in Fig. 5 is a plot of χ^{-1} versus T for $U_{0.2}Y_{0.8}Pd_2Al_3$, where the solid line represents a Curie-Weiss law that has been fitted to the data from 300 K down to 70 K. The fit yields a Curie-Weiss temperature $\Theta = -25.3$ K and an effective moment $\mu_{eff} = 2.76\mu_B/U$. The inset of Fig. 5 shows χ versus T data for $U_{0.2}Y_{0.8}Pd_2Al_3$
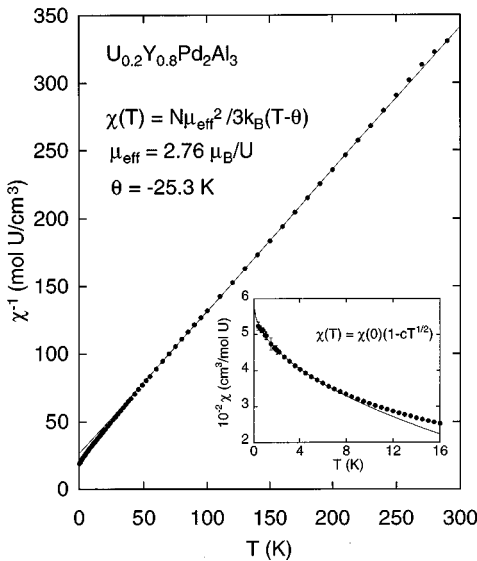

FIG. 5. The inverse magnetic susceptibility χ^{-1} vs T of $U_{0.2}Y_{0.8}Pd_2Al_3$. The solid line is a fit of the high-temperature data to a Curie-Weiss law. The inset shows χ vs T below $T = 16$ K. The solid line is a fit of the data to the expression $\chi(T) = \chi(0)(1 - cT^{1/2})$.

below 16 K, where the solid line is a fit of the data to the expression $\chi(T) = \chi(0)(1 - cT^{1/2})$. The fit describes the observed temperature dependence from the lowest measured temperature of 0.4 to ~7 K, consistent with the temperature range over which NFL behavior is observed in the $\rho(T)$ and $C(T)$ data.

IV. DISCUSSION

In many of the f-electron systems that exhibit NFL behavior that have been studied previously, the electrical resistivity $\rho(T)$, specific heat $C(T)$, and magnetic susceptibility $\chi(T)$ at low temperatures $T \ll T_0$, where T_0 is a characteristic temperature, scale with T_0 and have the following forms:

$$\rho(T) = \rho(0)[1 - a(T/T_0)], \tag{1}$$

$$C(T)/T = -(R/bT_0)\ln(T/b'T_0), \tag{2}$$

$$\chi(T) = \chi(0)[1 - c(T/T_0)^{1/2}], \tag{3}$$

where a can be positive or negative, and $|a|$, b, b', and c are constants of the order of unity.[1] For the $U_{1-x}Y_xPd_2Al_3$ system, the logarithmic temperature dependence of $C(T)/T$ is consistent with the data for $x=0.7$ and 0.8, while the forms of both $\rho(T)$ and $\chi(T)$ are consistent for $x=0.8$.

In some systems, such as $Y_{1-x}U_xPd_3$, the value of $b=4$ was taken from the two channel spin-$\frac{1}{2}$ Kondo model[2] which yields a specific heat given by Eq. (2) (with T_0 identified with the Kondo temperature T_K) from which T_0 can be determined from the slope $\partial[C(T)/T]/\partial \ln T$; i.e.,

$$T_0 = -R/b\{\partial[C(T)/T]/\partial \ln T\}. \tag{4}$$

Equation (4) yields values of $T_0 = 32$ K for $U_{0.2}Y_{0.8}Pd_2Al_3$ and $T_0 = 23$ K for $U_{0.3}Y_{0.7}Pd_2Al_3$. If T_0 could be identified with the Kondo temperature and the valence of U was +4, the decrease in T_0 with increasing uranium concentration would be consistent with the "Fermi level tuning" scenario established for the $Y_{1-x}U_xPd_3$ system as described in Ref. 2. Using the value $T_0 = 32$ K for $U_{0.2}Y_{0.8}Pd_2Al_3$, the fits of Eqs. (1) and (3) to the $\rho(T)$ and $\chi(T)$ data yield $a = -0.18$ and $c = 0.87$. The values of $|a|$ and c are of the order of unity and similar to the values derived from the $Y_{1-x}U_xPd_3$ system, where $a = 0.23$ and $c = 0.36$. In the $Y_{1-x}U_xPd_3$ system, these values of $|a|$, b, and c describe the $\rho(T)$, $C(T)$, and $\chi(T)$ data over an appreciable range of x and T_0.[2] No theoretical model presently known can account for the behavior of $\rho(T)$, $C(T)$, and $\chi(T)$ given by Eqs. (1)–(3). The quadrupolar Kondo model, which is the electric analog of the two channel spin-$\frac{1}{2}$ Kondo model, yields expressions for $C(T)$ and $\chi(T)$ of the form represented by Eqs. (2) and (3). However, the prediction for the electrical resistivity, which varies as $\rho(T) \sim 1 - a(T/T_0)^{1/2}$, does not agree with the linear T dependence of $\rho(T)$ reported here for the $U_{1-x}Y_xPd_2Al_3$ system and observed in virtually all of the other chemically substituted f-electron systems that display NFL behavior at low temperature.[1]

Some theoretical models, such as the quadrupolar Kondo model, are single ion in nature. In systems such as $Y_{1-x}U_xPd_3$ and $U_{1-x}Th_xPd_2Al_3$, scaling of the physical properties with the substituent concentration x and an effec-

tive Kondo temperature over an appreciable range of concentrations suggests that the NFL behavior in these systems is the result of a single ion mechanism. Even though our results show a change in T_0 with increasing uranium concentration, we cannot eliminate the scaling scenario at this point. As was discussed above, the change in T_0 might be related to a ''Fermi level tuning'' mechanism similar to that found in the $Y_{1-x}U_xPd_3$ system.[2] This issue can only be addressed through further transport, thermal, and magnetic measurements, as well as photoemission experiments, on the $U_{1-x}Y_xPd_2Al_3$ system.

An interesting aspect of the NFL behavior in the $U_{1-x}Y_xPd_2Al_3$ system is that it occurs above the concentration x at which magnetic order seems to vanish. This is often the case for systems which exhibit NFL behavior, including the $UCu_{5-x}Pd_x$ (Refs. 23 and 24) and $CeCu_{6-x}Au_x$ (Ref. 25) systems, which have Néel temperatures that steadily decrease towards $T=0$ K with increasing substituent concentration. It is tempting to associate this observation with the presence of a quantum critical point at $T=0$ K which gives rise to NFL behavior.[13–18] Further experiments at lower temperatures and on samples with concentrations x in the neighborhood of the concentration where the Néel temperature extrapolates to $T=0$ K ($x \approx 0.7$ in the case of the $U_{1-x}Y_xPd_2Al_3$ system) are required in order to determine if the quantum critical point can provide a consistent explanation of NFL behavior in these systems. The proximity of the NFL regime to magnetic ordering in the $U_{1-x}Y_xPd_2Al_3$ system suggests that the NFL behavior may be associated with fluctuations of an order parameter near a second-order phase transition.[13–17] However, a quantum critical point can also originate from an unconventional moment screening process such as the multichannel Kondo effect.[11,12,26]

Recently, an alternative model describing the NFL properties of rare-earth and actinide materials has been proposed.[18] This theory attributes the NFL behavior to a Griffiths' phase that is associated with magnetic clusters resulting from the interplay between the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction and the Kondo effect in the presence of magnetic anisotropy and disorder inherent in alloyed materials. The model predicts that the electronic specific-heat coefficient and the magnetic susceptibility diverge as $C(T)/T \propto \chi(T) \propto T^{-1+\lambda}$ with $\lambda < 1$. This theory has had some success in describing the behavior of $C(T)$ and $\chi(T)$ in other systems which exhibit NFL behavior.[27] Earlier in this paper, we analyzed the specific-heat data of the $x=0.8$ sample in terms of both a power law and a logarithmic temperature dependence and found that they described the data equally well. The power-law fit to the specific-heat data yields $\lambda=0.82$, which is similar to the values found for other NFL systems.[27] However, the magnetization data could not be fitted with a power law as predicted by the Griffiths'

phase model over any appreciable temperature range. This prevented us from deriving a value for λ from the $\chi(T)$ data. As was mentioned above, a power law which included a constant saturating term, $\chi(T) = \chi(0)[1 - c(T/T_0)^{1/2}]$, did provide an excellent fit of the data. It should also be noted that because of the polycrystalline nature of the samples, the magnetization data for the $x=0.8$ sample represent an average of the magnetization of crystallites with hexagonal structures which are oriented in different directions. Measurements on single-crystal specimens are needed to clarify whether both specific-heat and magnetization measurements give consistent values of λ for this system. No prediction for the temperature dependence of the electrical resistivity has yet emerged from the Griffiths' phase model.

V. SUMMARY

Measurements of $\rho(T)$, $C(T)$, and $\chi(T)$ on the $U_{1-x}Y_xPd_2Al_3$ system indicate that it belongs to an ever-growing class of compounds that exhibit NFL behavior. In particular, the physical properties of $U_{0.2}Y_{0.8}Pd_2Al_3$ exhibit the characteristic temperature dependences seen in many other NFL systems: an electrical resistivity $\rho(T)$ that is linear in temperature, a power-law or logarithmic divergence in the temperature dependence of the electronic specific-heat coefficient, $C(T)/T$, and a magnetic susceptibility $\chi(T)$ that varies as $1 - c(T/T_0)^{1/2}$. Comparison of the data with both the multichannel Kondo model and the Griffiths' phase model finds that both can describe some of our results, but neither can successfully account for all of the data presented. A clear trend in the data is the suppression of the Néel temperature T_N with increasing yttrium concentration x with T_N falling below the low-temperature limit of our experiment (~ 0.5 K) for $x \approx 0.7$, the Y concentration where we begin to observe non-Fermi-liquid behavior. These observations suggest the existence of a quantum critical point at $T=0$ K which might be related to the onset of NFL behavior in this system. The existence of such a quantum critical point and its implications for the physics of NFL materials needs to be investigated further by performing additional experiments at lower temperatures and at neighboring substituent concentrations x.

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