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Fermi- and non-Fermi-liquid ground states in $M_{1-x}U_xPd_3$ (M = Sc, Y, La, Pr, Zr, Th) systems

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Abstract

A growing number of chemically substituted intermetallic compounds of Ce and U exhibit non-Fermi-liquid (NFL) behavior in their low temperature physical properties and apparently constitute a new class of strongly correlated f-electron materials. In this paper, we update the experimental situation for the archetypal NFL f-electron system $Y_{1-x}U_xPd_3$ and briefly describe recent experiments on the related systems $M_{1-x}U_xPd_3$ (M = Sc, La, Pr, Zr, Th).

1. Introduction

A new class of strongly correlated f-electron materials whose low temperature physical properties display non-Fermi-liquid (NFL) behavior has emerged during the past several years [1,2]. The presently known materials that belong to this group are Ce and U intermetallic compounds containing nonmagnetic elemental substituents. The physical properties of these materials exhibit weak power law or logarithmic divergences in temperature that scale with a characteristic temperature T_0. This suggests the existence of a quantum critical point at $T = 0$ K, possible origins of which include an unconventional moment screening process, such as a multichannel Kondo effect [3], and fluctuations of an order parameter in the vicinity of a second-order phase transition at $T = 0$ K [4].

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In this paper, we update the experimental situation for the $Y_{1-x}U_xPd_3$ system, the first f-electron system in which NFL behavior was discovered [5], and present some recent experimental results on the related systems $M_{1-x}U_xPd_3$ (M = Sc, Y, La, Pr, Zr, Th).

2. The $Y_{1-x}U_xPd_3$ system – an update

The most recent version of the low temperature T–U concentration x phase diagram of the $Y_{1-x}U_xPd_3$ system is shown in Fig. 1 [6]. Of particular interest are the physical properties of $Y_{1-x}U_xPd_3$ in the cubic Cu_3Au phase that extends from $x = 0$ to $x \approx 0.55$. Magnetization $M(T)$ measurements, performed under field-cooling and zero-field cooling conditions on $Y_{1-x}U_xPd_3$ samples in the range $0.2 \leqslant x \leqslant 0.55$, exhibit irreversible behavior below an irreversibility temperature T_{irr}, reminiscent of spin-glass freezing. A plot of T_{irr} versus x in Fig. 1 delineates a region in which spin-glass and/or antiferromagnetic (AFM) ordering occurs. The kink in the T_{irr} versus

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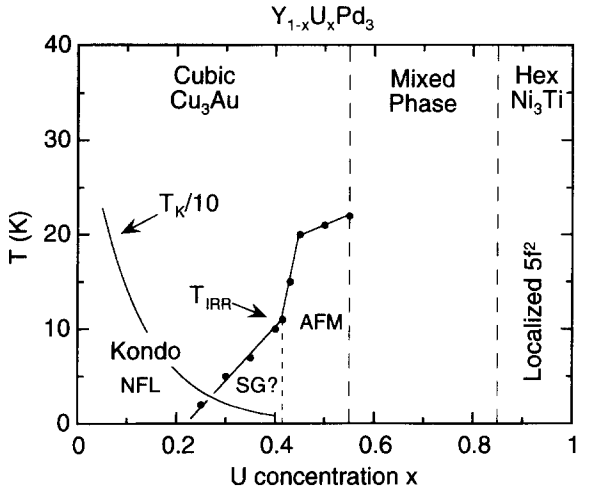

Fig. 1. Low temperature–U concentration phase diagram of $Y_{1-x}U_xPd_3$.

x curve at $x \approx 0.42$ may reflect a change from spin-glass ordering at lower values of x to long-range AFM ordering at higher values of x (long-range AFM ordering of the U moments has recently been observed by neutron scattering experiments on a sample with $x = 0.45$ [7]).

Transport, thermal, and magnetic measurements have revealed the existence of an unconventional Kondo effect in the region $0 < x \leqslant 0.55$ [5,8]. As indicated in Fig. 1, the Kondo temperature T_K decreases rapidly with x. This can be explained in terms of a phenomenon, discovered in photoemission studies of $Y_{1-x}U_xPd_3$ and referred to as "Fermi level tuning" [9], in which the U^{4+} 5f binding energy $\varepsilon_{5f} = E_F - E_{5f}$, where E_F is the Fermi energy and E_{5f} is the energy of the U^{4+} 5f state, increases by ~ 1 eV as x increases from 0 to 1 [1,8]. The increase of ε_{5f} with x can be understood in terms of increase of E_F with x as tetravalent U is substituted for trivalent Y. The nearly linear increase of ε_{5f} with x should cause a rapid decrease in T_K since

$$T_K \sim T_F \exp[-1/N(E_F)|\mathcal{J}|] \sim T_F \exp[-\varepsilon_{5f}/\langle V_{kf}^2\rangle N(E_F)] \quad (1)$$

where T_F is the Fermi temperature, $N(E_F)$ is the density of states at E_F, $\mathcal{J} \sim -\langle V_{kf}^2\rangle/\varepsilon_{5f}$ is the exchange interaction parameter, and V_{kf} is the hybridization matrix element. From this result, it is evident that a decrease in $\langle V_{kf}^2\rangle$ or $N(E_F)$ will also cause T_K to decrease, a point to which we will return later. Assuming a linear increase of ε_{5f} with x, $\varepsilon_{5f} \sim \varepsilon_0 + \varepsilon_1 x$, it can be shown that

$$T_K \sim (T_K)_0 \exp(-\alpha x) \quad (2)$$

where $(T_K)_0$ is the value of T_K for $x = 0$ and $\alpha = \varepsilon_1/\langle V_{kf}^2\rangle N(E_F)$.

The electrical resistivity $\rho(T)$, specific heat $C(T)$, and magnetic susceptibility $\chi(T)$ of the $Y_{1-x}U_xPd_3$ system exhibit NFL behavior at low temperatures $T \ll T_K$ and scale with T_K, where the value of T_K has been inferred from the high temperature $T \geqslant T_K$ behavior of $\rho(T)$ and $\chi(T)$ [1,5,8]. The U 5f electron contributions to these properties, $\Delta\rho(T)$, $\Delta C(T)$ and $\Delta\chi(T)$, in the limit $T \ll T_K$ are described well by the following expressions:

$$\Delta\rho(T)/\Delta\rho(0) = 1 - a(T/T_K), \quad (3)$$

$$\Delta C(T)/T = (-bR/T_K)\ln[b'(T/T_K)], \quad (4)$$

and

$$\Delta\chi(T)/\Delta\chi(0) = 1 - c(T/T_K)^{1/2}. \quad (5)$$

Eq. (4) has the same form as the two-channel spin-$\frac{1}{2}$ Kondo formula in which $b = 0.25$ and $b' = 2.4$ [10]. We have used these values of b and b' in Eq. (4) to analyze the $C(T)/T$ data for a sample with $x = 0.2$ since it yielded a value of 42 K for T_K which is close to the value inferred from the electrical resistivity at higher temperatures according to the criterion $\Delta\rho(T_K)/\Delta\rho(0) \equiv 0.8$ (T_K is the temperature where $\Delta\rho(T)$ starts to deviate from a $\ln T$ dependence). The values $a = 0.23$ and $c = 0.36$ in Eqs. (3) and (5) were determined from the low temperature $\Delta\rho(T)$ and $\Delta\chi(T)$ data for the specimen with $x = 0.2$ by setting T_K in Eqs. (3) and (5) equal to 42 K.

In the range $100 \leqslant T \leqslant 300$ K, $\Delta\chi(T)$ can be described by a Curie–Weiss law

$$\Delta\chi(T) = \chi(T) - \chi_0 = N\mu_{eff}^2/3k_B(T - \theta_p) \quad (6)$$

where χ_0 is a temperature-independent contribution, N is the number of U ions, μ_{eff} is the effective magnetic moment per U ion, and θ_p is the Curie–Weiss temperature. The effective moment $\mu_{eff} \approx 3.1\mu_B$ is smaller than the free ion value of $3.58\mu_B$ for tetravalent U and the Curie–Weiss temperature θ_p is large and negative. The reduced value of μ_{eff} and the large value of $|\theta_p|$ are both characteristic of systems which exhibit the Kondo effect where $-\theta_p = \beta T_K$ with $\beta \approx 3$–4. Since $|\theta_p|$ tends to saturate to a constant value of ~ 100 K for large values of $x \approx 0.5$, we assume that this value represents a non-Kondo contribution that is due to CEF effects and the spin glass and/or antiferromagnetic ordering that occurs for $x \geqslant 0.2$. Therefore, we set $T_K = (-\theta_p - 100\,\text{K})/3$, which gives a value consistent with the values of T_K inferred from the other measurements described above. The behavior of T_K as a function of x, as determined from the $\rho(T)$, $C(T)$, and $\chi(T)$ measurements at low and high temperatures, is shown in Fig. 2. The decrease of T_K with x is consistent with the "Fermi-level tuning" scenario and the expected exponential form given by Eq. (2). We reached similar conclusions in a previous investigation of the

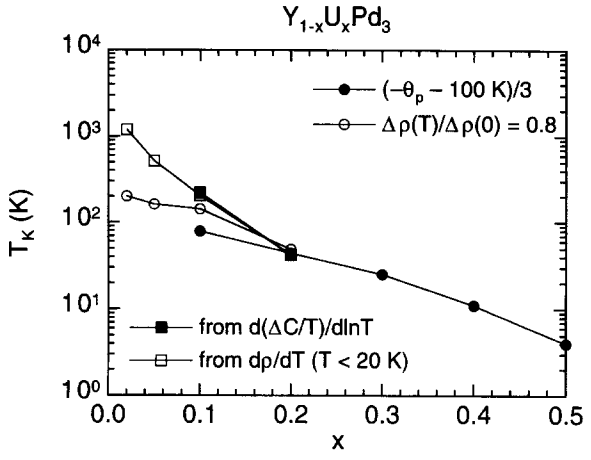

Fig. 2. Log T_K versus x for $Y_{1-x}U_xPd_3$ where T_K was inferred from $\rho(T)$ and $\chi(T)$ data at high temperatures $T \geq T_K$ (circles) and $\rho(T)$ and $C(T)$ data at lower temperatures $T \ll T_K$, (squares), as described in text.

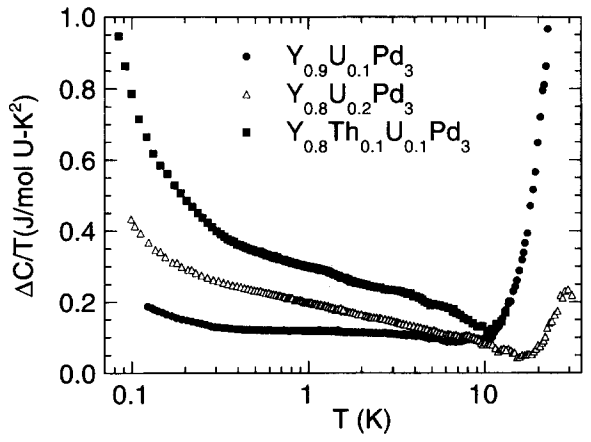

Fig. 3. U contribution to the specific heat ΔC divided by temperature T, $\Delta C(T)/T$, versus log T to temperatures as low as 80 mK for $Y_{0.9}U_{0.1}Pd_3$, $Y_{0.8}Th_{0.1}U_{0.1}Pd_3$, and $Y_{0.8}U_{0.2}Pd_3$.

$Y_{0.9-y}Th_yU_{0.1}Pd_3$ system where the "Fermi level tuning" was accomplished by varying the composition y of the tetravalent Th with fixed U concentration [11].

The low temperature $\Delta\rho(T)$ data for the $Y_{1-x}U_xPd_3$ system can be fitted by the more general relation $\Delta\rho(T)/\Delta\rho(0) = 1 - a(T/T_K)^n$, where $\rho(0)$ and n are adjustable parameters, in the range $0.02 \leq x \leq 0.2$. The best fits yield the value $n = 1.0 \pm 0.1$, suggesting that the linear T-dependence of $\Delta\rho$ is a single ion effect. The $\Delta C(T)/T$ data can be described by Eq. (4) in the range $0.3 \leq T \leq 10$ K, but deviate from this logarithmic T-dependence below 0.3 K as shown in Fig. 3 for $Y_{0.9}U_{0.1}Pd_3$, $Y_{0.8}Th_{0.1}U_{0.1}Pd_3$, and $Y_{0.8}U_{0.2}Pd_3$ where the measurements extend down to ~80 mK. A similar deviation of $\Delta C(T)/T$ from a logarithmic T-dependence at low temperatures was reported by Ott et al. [12]. Within the context of a two-channel spin-$\frac{1}{2}$ Kondo model, this upturn in $\Delta C(T)/T$ could be due to a lifting of the degeneracy of a U^{4+} doublet ground state by magnetic or quadrupolar interactions between U ions which would remove the residual $(R/2)\ln(2)$ entropy. The $\Delta\chi(T)$ data can be described by Eq. (5) between 0.6 and 40 K with parameter values $\chi(0) = 5.9 \times 10^{-3}$ emu/mol U, $c = 0.36$, and $T_K = 42$ K for $x = 0.2$. The $\chi(T)$ data were extracted from magnetization $M(H, T)$ data by decomposing the nonlinear M versus H isotherm at 0.6 K into a linear contribution $M_l(H, T) = \chi(T)H$, assumed to be intrinsic, and a nonlinear contribution $M_{nl}(H, T)$, assumed to be due to magnetic impurities, which scales with $H/(T + \theta)$, where $\theta = 0.235$ K, and can be approximated by a Brillouin function. The nonlinear contribution, which could be accounted for by magnetic impurities equivalent to 0.1% Ho, was subtracted from the $M(H, T)$ data to determine $\chi(T)$.

The development of a model that can account for the NFL behavior in the low temperature physical properties of $Y_{1-x}U_xPd_3$ requires information about the ground state of U^{4+} in the cubic CEF. In a cubic CEF, the nine-fold degenerate $J = 4$ Hund's rule multiplet of U^{4+} is split into Γ_4 and Γ_5 triplets, a Γ_1 singlet, and a Γ_3 nonmagnetic doublet that carries an electric quadrupole moment. If Γ_3 were the ground state, then the NFL characteristics in the physical properties at low temperature could be associated with a quadrupolar Kondo effect [12]. The quadrupolar Kondo model maps onto the two-channel spin-$\frac{1}{2}$ Kondo model and is based on an exchange interaction between an $S = \frac{1}{2}$ pseudo spin associated with the electric quadrupole moment of the Γ_3 ground state, and $\sigma = \frac{1}{2}$ pseudo spins of two time-reversed channels of conduction electrons, one channel corresponding to magnetic spin up and the other to magnetic spin down. According to the quadrupolar Kondo model, the electrical resistivity should vary as $\Delta\rho(T)/\Delta\rho(0) = 1 - a(T/T_K)^{1/2}$ [14], in disagreement with the linear T-dependence of Eq. (1) that is observed experimentally, while the specific heat should be given by Eq. (4) [10] and the magnetic susceptibility by Eq. (5) (where $\chi(0)$ is the van Vleck susceptibility for the Γ_3 ground state and the first CEF excited state) [15], both of which have T-dependences that are consistent with experiment. When the NFL behavior of $Y_{1-x}U_xPd_3$ was first reported, the $T^{1/2}$ temperature dependence of $\Delta\rho$ had not yet been calculated and arguments were advanced that $\Delta\rho(T)$ should have a linear T-dependence. Also, the value of $\Delta\chi(0)$ in Eq. (5) was consistent with the calculated van Vleck susceptibility between a Γ_3 ground state and the Γ_5 first excited state at 7 meV, the CEF

energy level scheme that Mook et al. [16] inferred from inelastic neutron scattering measurements on $Y_{0.8}U_{0.2}Pd_3$. The conclusion that the ground state is the Γ_3 nonmagnetic doublet was based on the small quasielastic line width $\Delta/2 < 0.1$ meV which is significantly smaller than the $k_B T_K \sim 4$ meV value expected for a Kondo effect of magnetic origin. Thus, it appeared that the NFL behavior of $Y_{1-x}U_xPd_3$ could be explained in terms of a quadrupolar Kondo effect.

In subsequent inelastic neutron scattering studies on samples with $x = 0.2$ and 0.45, McEwen et al. [17] also concluded that the Γ_3 nonmagnetic doublet was the probable ground state, but with the Γ_4 and Γ_5 excited levels at 2.5 and 36 meV, respectively. They attributed the features at 16 me V to phonons and noted that there was dispersion of the Γ_3–Γ_4 excitation from which they inferred the existence of nearest neighbor AFM correlations.

Recently, Dai et al. [7] performed polarized inelastic neutron scattering (INS) as well as elastic neutron scattering measurements on samples with $x = 0.2$ and 0.45. The polarized INS measurements indicated that the ground state was the Γ_5 triplet with an excited state Γ_3 nonmagnetic doublet at 5 meV and a Γ_4 triplet at 39 meV. The quasielastic line width $\Delta/2$ was estimated to be $\leqslant 1$ meV, still much smaller than the value $k_B T_K \sim$ 4 meV expected for a magnetic Kondo effect. However, it is possible that the Γ_5 ground state is split due to local deviations from cubic symmetry at the U sites by an amount that is smaller than the resolution of the polarized INS measurements (~ 5 meV). Elastic scattering measurements on the sample with $x = 0.45$ revealed the occurrence of long-range AFM order with a Néel temperature $T_N = 21$ K and an ordered moment $\mu = 0.7\mu_B/U$. The AFM structure is the same as for the compound UPd_4, which also has the cubic Cu_3Au structure, in which there is a doubling of the chemical unit cell in two directions [7]. The magnetization $M(T)$ was found to be reversible and to vary as $[M(T)/M(0)]^2 = (1 - T/T_N)^{0.7}$. The AFM ordering in the $x = 0.45$ sample is rather surprising since the magnetization of this sample exhibits irreversible behavior reminiscent of spin glass freezing with T_{irr} equal to the Néel temperature T_N. Neutron scattering measurements on the sample with $x = 0.2$ did not indicate any magnetic order above 0.2 K. However, critical fluctuations associated with AFM ordering with the same wave vector as the $x = 0.45$ sample were observed on cooling from 77 to 0.2 K.

A metallurgical study of selected $Y_{1-x}U_xPd_3$ samples in the range $0 \leqslant x \leqslant 0.2$ by means of electron probe microanalysis by Süllow et al. [18] revealed local variations of the composition x on a scale of 10 μm for arc-melted and unannealed samples, such as those used in the present investigation. We have also observed U concentration fluctuations and changes in microstructure as x is varied in the $Y_{1-x}U_xPd_3$ system in our ongoing investigation which will be reported on at a later date.

Thus, it would appear that the situation in the $Y_{1-x}U_xPd_3$ system is considerably more complex than originally envisaged, and no single model presently available seems to be able to account for all of the properties of this rich and interesting pseudobinary alloy system. It is possible that a combination of factors including a multichannel Kondo effect, fluctuations of an order parameter above a 0 K second-order phase transition, and chemical disorder will be required to explain the behavior of this system.

3. $M_{1-x}U_xPd_3$ systems based on other M-elements

We have investigated the properties of systems with the formula $M_{1-x}U_xPd_3$ for M = Sc, La, Pr, Zr, and Th, in addition to Y [8, 19]. The systems based on trivalent M-elements (Sc, La, Pr) crystallize in the cubic Cu_3Au structure for $0 \leqslant x < 0.5$, while those based on the tetravalent M-elements (Zr, Th) crystallize in the hexagonal Ni_3Ti structure, like pure UPd_3. No Kondo effect is observed for $M_{1-x}U_xPd_3$ with M = La, Pr (very low T_K), while a Kondo effect is observed for $M_{1-x}U_xPd_3$ with M = Sc, Y with values of T_K that exhibit Fermi level tuning and are larger for Sc than the corresponding T_K values for Y. This follows from Eq. (1) if $N(E_F)$ and/or $\langle V_{kf}^2 \rangle$ increases with decreasing unit cell volume v_c of the MPd_3 host compounds of trivalent M-elements. The values of v_c are related in the following way: $v_c(La) > v_c(Pr) > v_c(Y) > v_c(Sc)$, yielding the following ordering of T_K: $T_K(La) < T_K(Pr) < T_K(Y) < T_K(Sc)$. Electrical resistivity ρ versus T data between ~ 2 K and room temperature for $M_{1-x}U_xPd_3$ systems with M = Sc, Y, Pr, La are shown in Fig. 4. At low temperatures, the physical properties of $M_{1-x}U_xPd_3$ exhibit non-Fermi-liquid characteristics for M = Sc and Y and Fermi-liquid behavior for M = La and Pr. The $\Delta C/T$ versus T^2 data in the range $0.4 \leqslant T \leqslant 5$ K for $La_{1-x}U_xPd_3$ shown in Fig. 5 reveal a large temperature independent enhancement of γ of ~ 500 mJ/mol U K^2 for the samples with $x = 0.025$ and 0.050, indicative of local Fermi-liquid behavior. This large enhancement of γ implies that there is appreciable hybridization of the localized U 5f states with the conduction electron states, but the physics is apparently different than in the $M_{1-x}U_xPd_3$ systems with M = Sc and Y that display local non-Fermi-liquid behavior at low temperatures. Features are observed in the specific heat of the samples with $x = 0.075$ and 0.10 that are probably associated with spin-glass or AFM ordering of the U ions, consistent with the irreversibility observed in low field $M(T)$ measurements on these

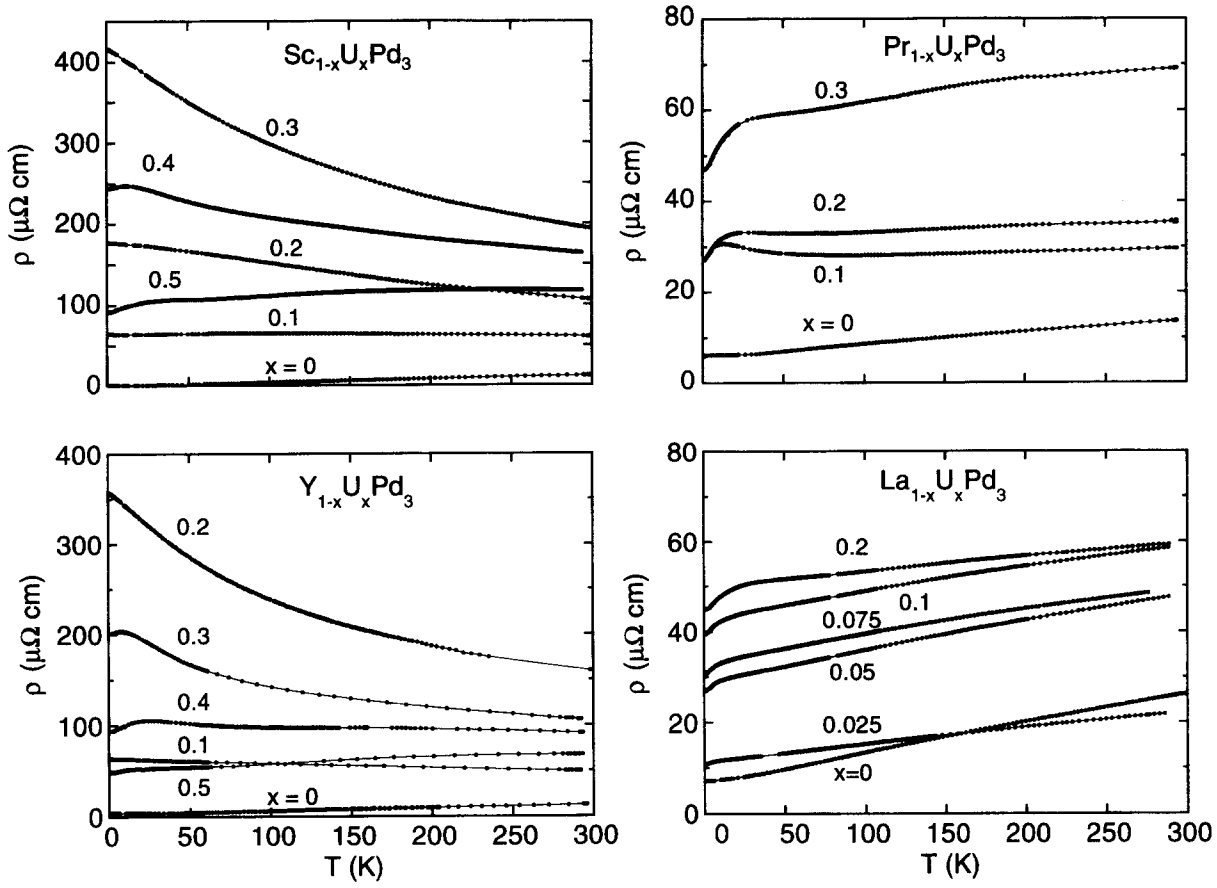

Fig. 4. Electrical resistivity ρ versus temperature T for $M_{1-x}U_xPd_3$ (M = Sc, Y, Pr, La).

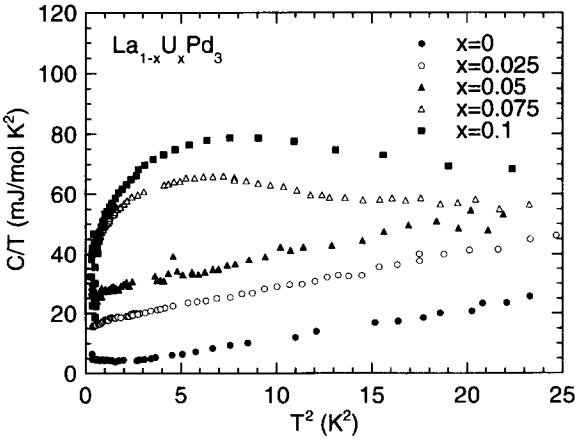

Fig. 5. Low temperature specific heat C divided by temperature T versus T^2 for $La_{1-x}U_xPd_3$ samples with $x = 0$, 0.025, 0.050, 0.075, and 0.10.

samples. For the $M_{1-x}U_xPd_3$ systems with M = Zr and Th, no Kondo effect is observed, presumably because these tetravalent ions result in large values of ε_{5f} and correspond to small values of T_K.

4. Concluding remarks

The $Y_{1-x}U_xPd_3$ system exhibits a rich variety of behavior in the cubic Cu_3Au phase ($0 < x \leqslant 0.55$) which includes an unconventional Kondo effect, Fermi level tuning of the Kondo temperature, NFL behavior of the low temperature physical properties that scale with T_K ($0 < x \leqslant 0.2$), and spin-glass and/or long-range AFM order ($0.2 \leqslant x \leqslant 0.55$). No Kondo effect is observed for $M_{1-x}U_xPd_3$ with M = La, Pr (very low T_K), while a Kondo effect is observed for $M_{1-x}U_xPd_3$ with M = Sc, Y with values of T_K that exhibit Fermi level tuning and are larger for Sc than the corresponding T_K values for Y. Possible mechanisms for this behavior include a multichannel Kondo effect, of either magnetic or

electric (quadrupolar) origin, and fluctuations of an order parameter above a 0 K second-order phase transition. Chemical disorder may also play an important role. The NFL behavior seems to be a general phenomenon of f-electron systems; more than ten f-electron systems have been found in which the T-dependences of $\Delta\rho(T)$, $\Delta C(T)$, and $\Delta\chi(T)$ at low temperatures $T \ll T_0$, where T_0 is a characteristic temperature (which can be identified with T_K in many several systems), scale with T_0 and are the same as those exhibited by $Y_{1-x}U_xPd_3$, although in some cases the parameter a that appears in the electrical resistivity is negative [1,2]. As we suggested several years ago [1,2], these systematics of NFL behavior appear to be general characteristics of a new class of strongly correlated f-electron materials. Much more work remains to be done to find new f-electron systems that exhibit NFL behavior, characterize the NFL behavior, and establish the underlying microscopic mechanisms that are responsible for the NFL behavior. Further work also needs to be done on the related $M_{1-x}U_xPd_3$ (M = Sc, La, Pr, Zr, Th) systems to determine how their physical properties compare to those of the $Y_{1-x}U_xPd_3$ system and explore new physics of these materials.

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