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Non-Fermi Liquid and Spin-Glass Behavior of the $Sc_{1-x}U_xPd_3$ System

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Abstract

Previous electrical resistivity $\rho(T)$, magnetic susceptibility $\chi(T)$, and specific heat C(T) measurements on the $Y_{1-x}U_xPd_3$ system have revealed Kondo behavior for $0 < x \leq 0.2$ and spin-glass-like behavior for $0.3 \leq x \leq 0.5$. The Kondo behavior is unusual and characterized by non-Fermi liquid behavior at low temperatures $T << T_K$, where T_K is the Kondo temperature: $\rho(T)/\rho(0) \approx 1 - T/(aT_K)$, and $C(T)/T \approx -(1/T_K)\ln T$ with evidence for a finite T = 0 residual entropy $S(0) = (R/2)\ln(2)$. We report measurements of $\rho(T)$, $\chi(T)$, and C(T) on the $Sc_{1-x}U_xPd_3$ system which reveal similar Kondo, non-Fermi liquid, and spin-glass behaviors.

Running title: Non-Fermi Liquid and Spin-Glass Behavior of $Sc_{1-x}U_xPd_3$
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Previous measurements of the $Y_{1-x}U_xPd_3$ system have revealed interesting magnetic, transport, and thermodynamic properties [1]. This system exhibits Kondo behavior for $x \leq 0.2$ and spin-glass behavior for $0.3 \leq x \leq 0.5$. The Kondo behavior is unusual and characterized by non-Fermi liquid behavior at low temperatures $T << T_K$, where T_K is the Kondo temperature. The electrical resistivity varies nearly linearly with temperature $\rho(T)/\rho(0) \approx 1 - T/(aT_K)$, and the electronic specific heat diverges logarithmically $\Delta C(T)/T \approx -(1/T_K)\ln T$ with evidence for a finite $T = 0$ residual entropy $S(0) = (R/2)\ln(2)$.

In this paper, we present results for the related system $Sc_{1-x}U_xPd_3$, where Sc is trivalent like Y, but has a smaller ionic radius. The results for $Sc_{1-x}U_xPd_3$ are similar to $Y_{1-x}U_xPd_3$; Kondo behavior is observed with a T_K that decreases with increasing x, but $Sc_{1-x}U_xPd_3$ has a larger value of T_K than $Y_{1-x}U_xPd_3$ for a given x. This result can be understood using the simple $U \rightarrow \infty$ expression for T_K,

$$k_BT_K \sim E_F \exp\left(\frac{-\varepsilon_f}{<V_{kf}^2> N(E_F)}\right) \qquad (1)$$

First, in Fermi-level tuning, the substitution of tetravalent U for trivalent Sc increases the number of conduction electrons, which increases the binding energy of the 5f electrons $\varepsilon_f = |E_F - E_{5f}|$, where E_F is the Fermi level, and thus decreases T_K. Second, the smaller size of Sc^{3+} compared to Y^{3+} can cause $Sc_{1-x}U_xPd_3$ to have a larger conduction electron – U 5f electron hybridization characterized by the matrix element V_{kf}, and/or a larger host density of states at the Fermi level $N(E_F)$ than $Y_{1-x}U_xPd_3$, yielding a larger T_K for a given x. Indeed, specific heat measurements [2] and band structure calculations [3] find that $N(E_F)$ of $ScPd_3$ is approximately 2.5 times larger than that of YPd_3. Behavior that is generally consistent with the two-channel quadrupolar Kondo effect is observed for U concentrations as high as 0.3. For $0.4 \leq x \leq 0.5$., $Sc_{1-x}U_xPd_3$ exhibits cooperative spin-glass behavior.

Samples of $Sc_{1-x}U_xPd_3$, where x = 0, 0.1, 0.2, 0.3, 0.4, and 0.5, were prepared by arc-melting the elemental metals on a copper hearth. Measurements of $\rho(T)$ were performed using a standard ac four-wire technique to temperatures as low as 1.2K; the magnetic susceptibility was measured using a commercial SQUID magnetometer down to 1.8K. The specific heat C(T) was measured using a semi-adiabatic heat-pulse technique for temperatures $0.5K \leq T \leq 20K$. X-ray powder diffraction measurements revealed that each sample consisted of a single phase with the cubic Cu_3Au crystal structure, except for the sample with x = 0.6, whose X-ray pattern showed mixed cubic and hexagonal phases. The cubic lattice parameter increases linearly with increasing x: $a(\text{Å}) = (3.955\pm0.002) + (0.150\pm0.005)x$.

The resistivity for $ScPd_3$ exhibits normal metallic behavior. For samples with finite x, the magnetic impurity ion contribution was estimated by subtracting $\rho(T)$ for $ScPd_3$: $\Delta\rho(T) = \rho(T, x) - \rho(T, x = 0)$. For $0.1 \leq x \leq 0.3$ and high temperatures $T > T_K$, $\Delta\rho(T)$ increases logarithmically with decreasing temperature (Fig. 1), consistent with Kondo scattering of the conduction electrons by local U ion moments. A rough estimate of T_K was obtained from the temperature at which $\Delta\rho(T) / \Delta\rho(0) = 0.8$, which is approximately the point at which the data begin to deviate from logarithmic behavior; this gives values of T_K that agree fairly well with those obtained from measurements of C(T). We find that T_K decreases with increasing x, consistent with Fermi-level tuning: $T_K \approx$ 260K, 120K, and 60K for x = 0.1, 0.2, and 0.3, respectively. These values are larger than similar estimates for $Y_{1-x}U_xPd_3$: $T_K \approx$ 140K and 50K for x = 0.1 and 0.2, respectively [4], which we attribute to the smaller ionic radius of Sc, as described earlier. We note that the Kondo temperature for $Sc_{0.7}U_{0.3}Pd_3$ is comparable to that for $Y_{0.8}U_{0.2}Pd_3$. At lower temperatures, the $\Delta\rho(T)$ data were fit to a power law, $1 - \Delta\rho(T)/\Delta\rho(0) = (T / T_o)^n$ from 1.2K to 20K, shown in the inset of Fig. 1. Solid lines represent the best fits with $n = 0.9\pm0.2$, 1.4 ± 0.2, and 1.0 ± 0.2 for x = 0.1, 0.2, and 0.3, respectively. This result is in contrast to Fermi liquid theory which predicts n = 2, and to the two-channel quadrupolar Kondo model which predicts n = 0.5 in the low temperature limit [5].

The resistivity of the sample with x = 0.4 increases with decreasing temperature, indicative of a Kondo effect, but exhibits a maximum near 15K, and then decreases down to 1.2K. For the sample with $x = 0.5$, $\Delta\rho$ decreases slowly with temperature down to ~ 30K, and then decreases more rapidly at lower temperatures. This deviation from the Kondo behavior of samples with smaller x could be due to spin-glass cooperative behavior of the U ions, indicated by measurements of the magnetic susceptibility in low fields (1 Oe) which revealed irreversible behavior for $x = 0.4$ and 0.5. The spin-glass freezing temperature T_{SG}, defined as the onset of irreversibility, is approximately equal to 7K for $x = 0.4$ and 13K for $x = 0.5$. As the concentration of U ions is increased, the characteristic Kondo energy scale decreases, while interimpurity interactions increase, resulting in spin-glass freezing with freezing temperatures T_{SG} that increase with x. The values of T_{SG} are smaller than those for $Y_{1-x}U_xPd_3$ in which $T_{SG} \approx 10K$ for $x = 0.4$ and $\approx 20K$ for $x = 0.5$. No irreversibility was observed down to 1.8K for $x = 0.3$, similar to $Y_{0.8}U_{0.2}Pd_3$.

Figure 2 shows the temperature dependence of the inverse magnetic susceptibility $\chi(T)$ between $1.8K \leq T \leq 300K$ for $x = 0.1$, 0.2, 0.3, 0.4, and 0.5. The data were fit to a Curie-Weiss law plus a constant, $\chi = \chi_o + C / (T - \theta_{CW})$, in the range $150K \leq T \leq 300K$. Below 150K, $\chi(T)$ departs from Curie-Weiss behavior and approaches a finite value as $T \rightarrow 0$, indicative of a nonmagnetic ground state. The effective moment μ_{eff} appears to increase slightly with increasing x, from approximately $3.0\mu_B$ for $x = 0.1$ to $3.6\mu_B$ for $x = 0.5$. These values are comparable to the free ion values of $3.58\mu_B$ and $3.62\mu_B$ for tetravalent and trivalent U, respectively. The constant χ_o shows a decreasing trend with increasing x, varying from approximately $+0.25 \cdot 10^{-4}$ emu/mole formula for $x = 0$ [6], passing through zero for $x \approx 0.1$ to $-3.79 \cdot 10^{-4}$ emu/mole formula for $x = 0.5$.

The most interesting result, however, is the behavior of the Curie-Weiss temperature θ_{CW} which is shown in the inset of Fig. 2 (the solid line is a guide to the eye). The Curie-Weiss temperature has an enormous negative value of -689K for $x = 0.1$, and increases monotonically with increasing x, reaching -164K for $x = 0.5$, and apparently saturating to a small negative value at higher x. Considering that $-\theta_{CW} \approx (3\text{–}4)T_K$ for Kondo systems, these data are consistent with

Fermi-level tuning, further illustrating the decrease of the Kondo temperature with increasing x. Such a trend was observed for $Y_{1-x}U_xPd_3$, but with smaller values of $|\theta_{CW}|$ for a given x. The Kondo temperature T_K inferred from $|\theta_{CW}|$ is associated with the antiferromagnetic exchange interaction between the conduction electrons and the Γ_4 and Γ_5 excited magnetic states which are thermally populated at higher temperatures. This value of T_K should be close to that which is related to the quadrupolar interaction of the conduction electrons with the Γ_3 nonmagnetic doublet ground state, since the coupling strengths arising from the U localized 5f – conduction electron hybridization are comparable for the magnetic and quadrupolar Kondo mechanisms.

Specific heat data for $Sc_{0.7}U_{0.3}Pd_3$ and $Y_{0.8}U_{0.2}Pd_3$ [1] are presented in Fig. 3(a) as $\Delta C/T$ versus T on a semilogarithmic scale. The contribution of the magnetic U ions, ΔC, was obtained by subtracting an estimated phonon term βT^3 from C(T), where β was obtained by scaling the value for $ScPd_3$ [2] by the molecular weight of $Sc_{0.7}U_{0.3}Pd_3$. The data display a T = 0 logarithmic divergence over a decade of temperature, in agreement with the two-channel Kondo model with $T_K \approx 40K$ [1]. The upturn at higher temperature is attributed to a Schottky anomaly arising from excited crystalline electric field levels and/or incorrect subtraction of the phonon contribution. The striking similarity between the two samples, with different U ion concentrations, is in agreement with the increase in the Kondo energy scale when Sc is used instead of Y, which is ascribed to the smaller ionic radius of Sc. Figure 3(b) shows that the entropy ΔS associated with the U ions saturates to approximately (R/2)ln(2), which implies a T = 0 entropy of the same value, in agreement with the prediction of the two-channel Kondo model.

Measurements of $\rho(T)$, $\chi(T)$, and C(T) on $Sc_{1-x}U_xPd_3$ exhibit Kondo behavior for x = 0.1, 0.2, and 0.3, and spin-glass behavior for x = 0.4 and 0.5. The results are similar to $Y_{1-x}U_xPd_3$ in that the Kondo behavior is characterized by non-Fermi liquid behavior, and appears to be two-channel in nature and mediated by a quadrupolar interaction. We observe Fermi-level tuning upon the substitution of tetravalent U for trivalent Sc; however, the Kondo energy scale is higher than that for $Y_{1-x}U_xPd_3$ for a given x due to the smaller ionic radius of Sc which results in stronger hybridization of the U 5f states with the conduction band states and a higher host density of states at the Fermi level.

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References

[1] C. L. Seaman, M. B. Maple, B. W. Lee, S. Ghamaty, M. S. Torikachvili, J.-S. Kang, L. Z. Liu, J. W. Allen, and D. L. Cox, Phys. Rev. Lett. **67** (1991) 2882; J. Alloys and Compounds **181** (1992) 327.

[2] M. J. Besnus, J. P. Kappler, and A. Meyer, J. Phys. F. **13** (1983) 597.

[3] C. Koenig, Z. Phys. B **50** (1983) 33.

[4] C. L. Seaman and M. B. Maple, SCES Proceedings (1993).

[5] A. W. W. Ludwig and I. Affleck, Phys. Rev. Lett. **67** (1991) 3160.

[6] W. E. Gardner, J. Penfold, T. F. Smith, and I. R. Harris, J. Phys. F. **2** (1972) 133.

Figure Captions

Fig. 1. Uranium contribution to the electrical resistivity $\Delta\rho(T)$ due to the presence of magnetic U impurity ions of $Sc_{1-x}U_xPd_3$ (x = 0.1, 0.2, 0.3), normalized to the extrapolated zero temperature value, $\Delta\rho(0)$, on a semilogarithmic scale. Inset: Log–log plot of $1 - \Delta\rho(T)/\Delta\rho(0)$ vs T at low temperatures. Solid lines are least-squares fits to a power law $(T/T_o)^n$, where n is given in the text.

Fig. 2. Magnetic susceptibility $(\chi - \chi_o)^{-1}$ vs T for $Sc_{1-x}U_xPd_3$ (x = 0.1, 0.2, 0.3, 0.4, 0.5), where χ_o is defined in the text. Solid lines represent linear fits to data for $150K \leq T \leq 300K$. Inset: Curie-Weiss temperature θ_{CW} vs x. The solid line is a guide to the eye.

Fig. 3. (a) U ion contribution to the specific heat $\Delta C/T$ vs T on a semilogarithmic scale for $Y_{0.8}U_{0.2}Pd_3$ and $Sc_{0.7}U_{0.3}Pd_3$. (b) U ion contribution to the entropy, ΔS vs T.

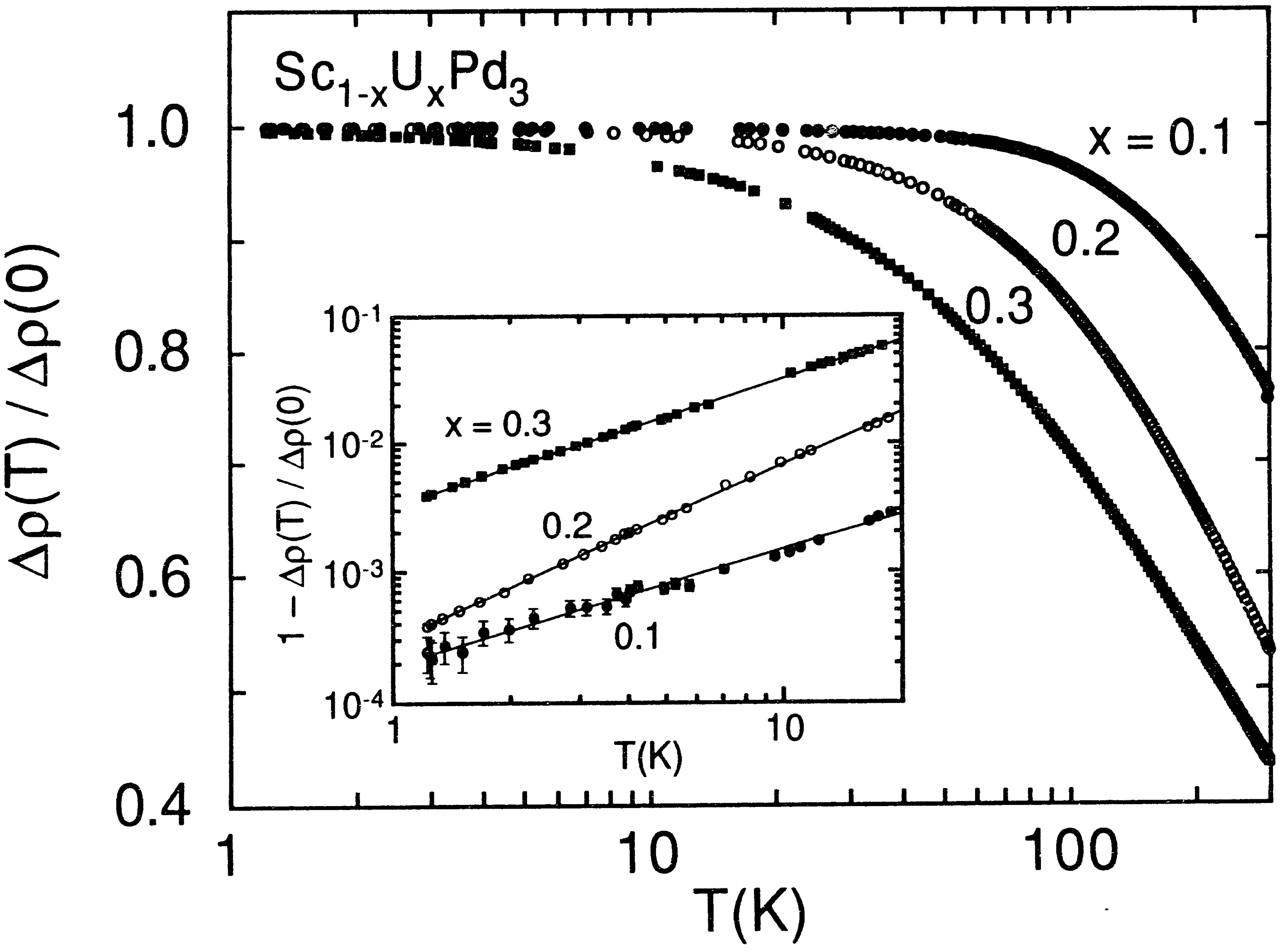

Fig. 1

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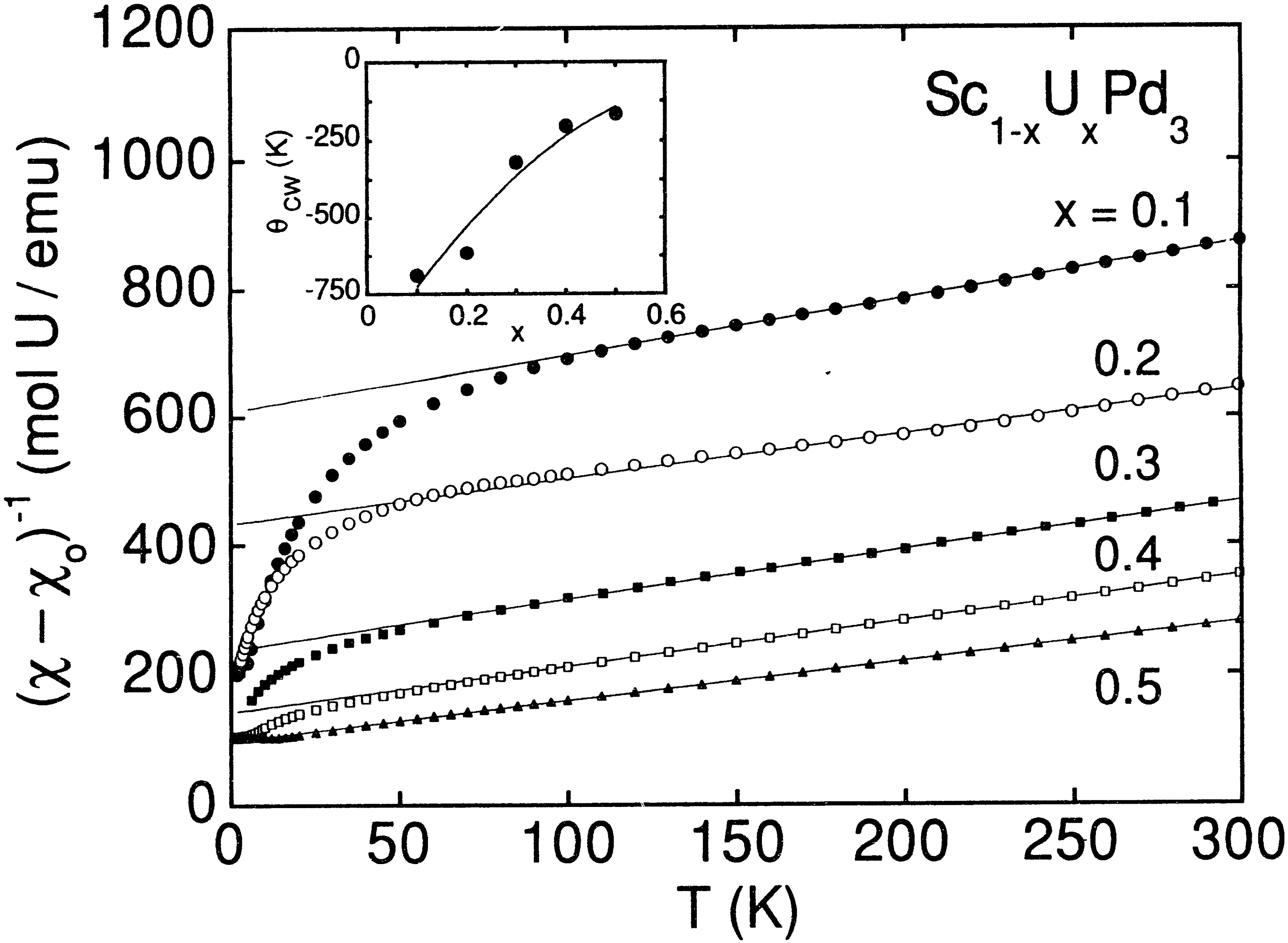

Fig. 2

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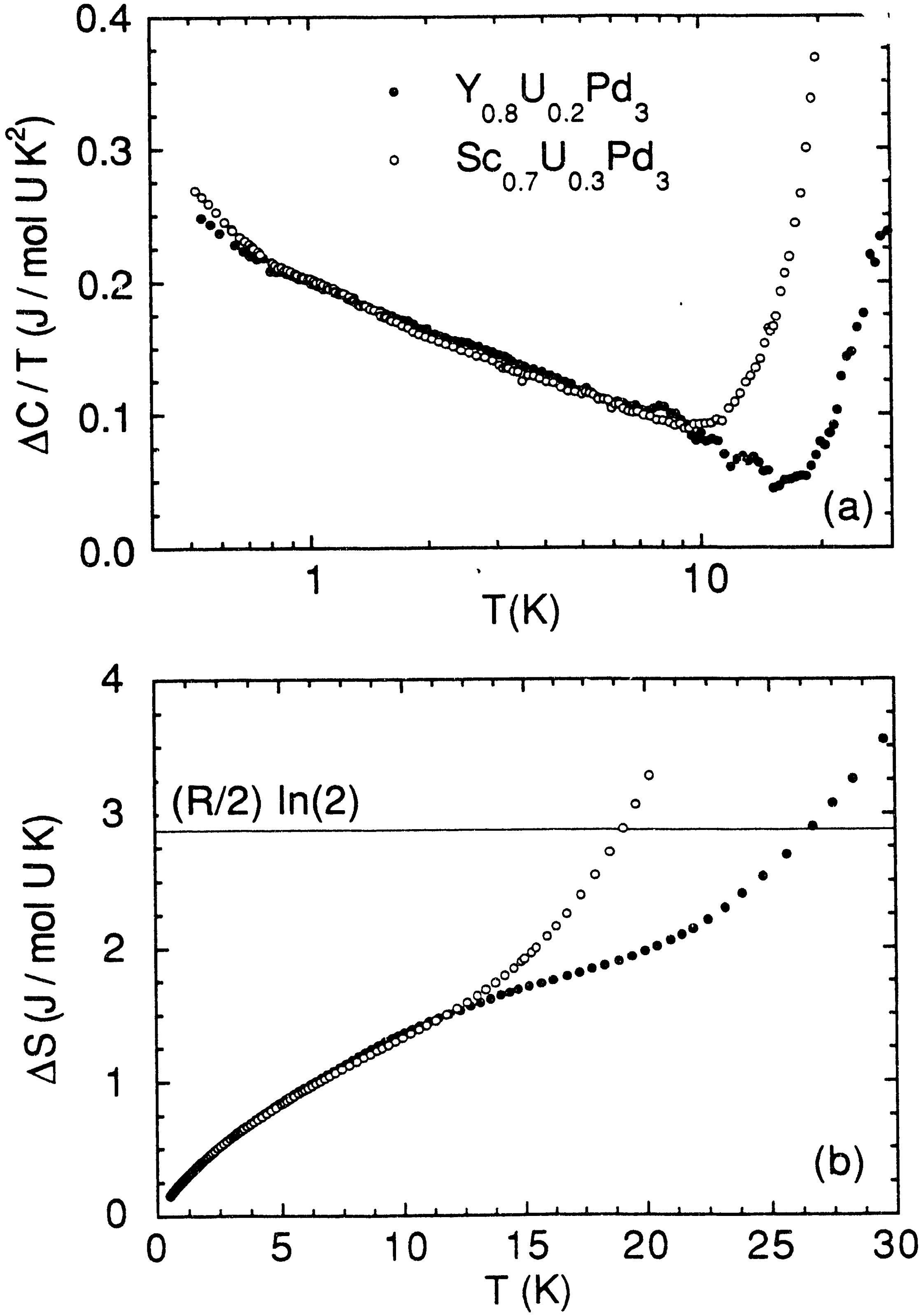

Fig. 3

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