ELSEVIER

Physica B 199&200 (1994) 423–425

PHYSICA B

Fermi level tuning in $(Y_{1-y}M_y)_{1-x}U_xPd_3$ (M = Th, La)

M.B. Maple[a,b,*], D.A. Gajewski[a], C.L. Seaman[a], J.W. Allen[c]

[a] *Department of Physics and Institute for Pure and Applied Physical Sciences, University of California, San Diego, La Jolla, CA 92093, USA*

[b] *Center for Materials Science, Los Alamos National Laboratory, Los Alamos, NM 87545, USA*

[c] *Randall Laboratory, University of Michigan, Ann Arbor, MI 48109-1120, USA*

Abstract

The effects of the increase of the U 5f binding energy and the decrease in the localized U 5f electron–conduction electron hybridization on the Kondo temperature, with U intersite interactions held constant to first approximation, are under investigation by means of electrical resistivity, magnetic susceptibility, and specific heat as a function of y for fixed values of x in the $(Y_{1-y}M_y)_{1-x}U_xPd_3$ (M = Th, La) systems.

Photoemission spectroscopy measurements on the $Y_{1-x}U_xPd_3$ system [1] have revealed a "Fermi level tuning" effect in which the separation δE between the Fermi level E_F and the energy E_{5f} of the U 5f states, $\delta E \equiv E_F - E_{5f}$, increases upon substitution of tetravalent U for trivalent Y by ~ 1 eV as x varies from 0 to 1. This occurs through an increase in the conduction electron density with x, and, in turn, E_F. The increase of δE with increasing x is also manifested in a corresponding decrease of the Kondo temperature T_K, as inferred from features in the electrical resistivity $\rho(T)$, magnetic susceptibility $\chi(T)$, and the low temperature specific heat $C(T)$, where $C(T)$ was analyzed in terms of a two channel Kondo effect, presumed to be due to the interaction of the conduction electrons with the electric quadrupole moments of the Γ_3 nonmagnetic doublets of U^{4+} in the cubic $Y_{1-x}U_xPd_3$ system [2–4]. However, the substitution of U^{4+} for Y^{3+} in the $Y_{1-x}U_xPd_3$ system has the additional effect of increasing the U intersite interaction strength as x is increased which leads to spin-glass freezing below a characteristic temperature T_{SG} that increases with x. In order to study the effects of the increase of δE and the decrease in the localized U 5f electron–conduction electron hybridization V_{kf} on T_K, with U intersite interactions held constant to first approximation, we have been performing measurements of $\rho(T)$, $\chi(T)$, and $C(T)$ as functions of y for a fixed value $x = 0.1$ in the $(Y_{1-y}M_y)_{1-x}U_xPd_3$ (M = Th, La) systems. The results for M = Th, where the primary effect should be to increase δE, are reported herein. The experiments in which M = La, where the major effect should be to decrease V_{kf} due to the expansion of the YPd_3 lattice (La^{3+} has a larger metallic radius that Y^{3+}), are still in progress.

Shown in Fig. 1(a) are plots of ρ versus T between 1.2 K and 300 K for $Y_{0.9-y}Th_yU_{0.1}Pd_3$ samples with $y = 0, 0.1, 0.2, 0.3$, and 0.4. The $\rho(T)$ data for $0 \leqslant y \leqslant 0.2$ reveal that ρ increases with decreasing T, indicative of a Kondo effect. The increase of the temperature dependence of ρ with y for $0 \leqslant y \leqslant 0.2$ suggests that T_K decreases with y, consistent with Fermi level tuning. In order to estimate values of T_K from the $\rho(T)$ data for

* Corresponding author.

SSDI 0921-4526(93)E0302-W

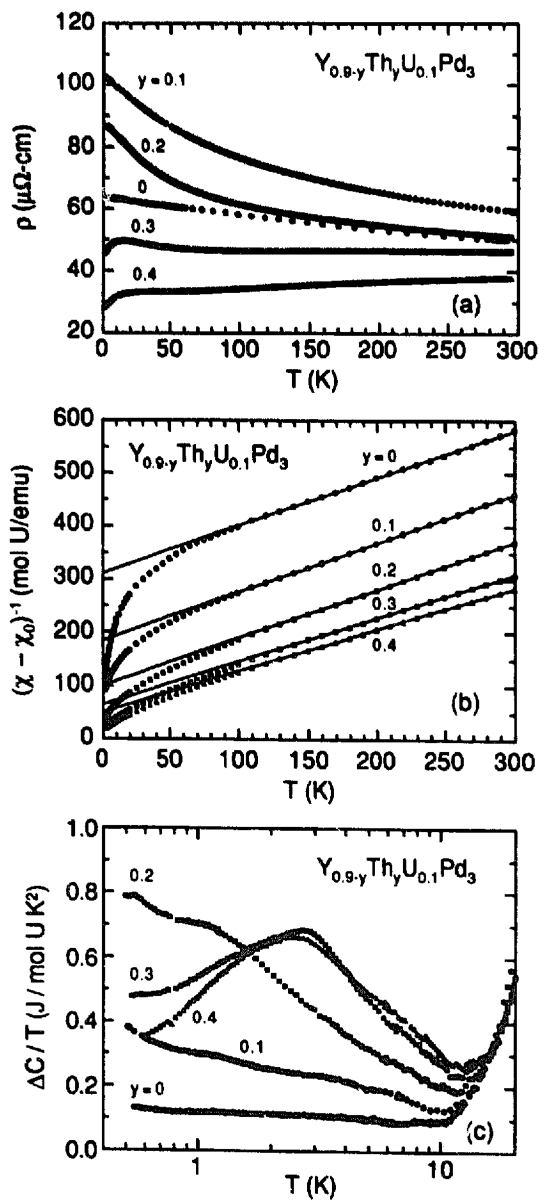

Fig. 1. Temperature T dependence of the electrical resistivity ρ, magnetic susceptibility χ, and electronic specific heat ΔC of the $Y_{0.9-y}Th_yU_{0.1}Pd_3$ (y = 0, 0.1, 0.2, 0.3, 0.4) system: (a) ρ versus T; (b) $(\chi - \chi_0)^{-1}$ versus T, where χ_0 is defined in the text; (c) $\Delta C/T$ versus ln T.

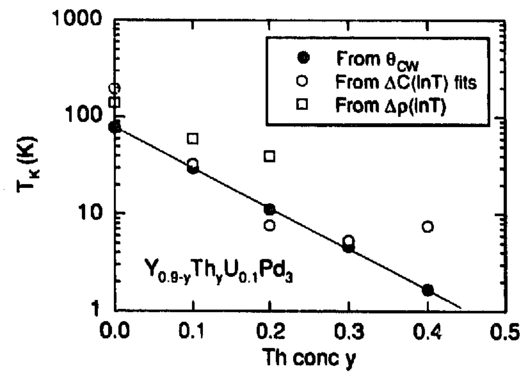

Fig. 2. Kondo temperature T_K on a logarithmic scale versus Th concentration y for the $Y_{0.9-y}Th_yU_{0.1}Pd_3$ system. T_K was inferred from ρ versus T (open squares), χ versus T (solid circles) and $\Delta C/T$ versus ln T (open circles) data shown in Figs. 1(a), (b) and (c), respectively.

$Y_{0.9-y}Th_yU_{0.1}Pd_3$ with y = 0.1 and 0.2, we have subtracted the $\rho(T)$ data for YPd_3 to obtain the U contribution $\Delta\rho(T)$ and defined T_K as the temperature at which $\Delta\rho(T)/\Delta\rho(0) = 0.8$; the values of T_K obtained from this procedure are plotted in Fig. 2 (open squares).

Displayed in Fig. 1(b) are plots of $(\chi - \chi_0)^{-1}$ versus T for $Y_{0.9-y}Th_yU_{0.1}Pd_3$ samples with y = 0, 0.1, 0.2, 0.3, and 0.4 between 1.8 K and 300 K, where χ_0 was determined by fitting the $\chi(T)$ data to the sum of a constant χ_0 and a Curie–Weiss law $N\mu_{eff}^2/3k_B(T - \theta_{CW})$, where N is the number of U ions, μ_{eff} is the U effective moment, and θ_{CW} is the Curie–Weiss temperature. The $(\chi - \chi_0)^{-1}$ versus T data are linear in the range $100\ K < T \leqslant 300\ K$ and the values of μ_{eff}, χ_0, and θ_{CW} derived from the fits depend on y: i.e., $\mu_{eff}(\mu_B) = (2.92 \pm 0.05) + (0.6 \pm 0.2)y$, $\chi_0(10^{-5}\ \text{emu/mol U}) = (-4.0 \pm 0.6) - (25 \pm 3)y$, and $-\theta_{CW}(K) = [371 \exp(-9.56y)] + 57$. Below ~ 100 K, the $(\chi - \chi_0)^{-1}$ versus T data fall below the linear fits to the higher T data and approach a finite value as $T \to 0$, indicative of a nonmagnetic ground state, which is presumably the U Γ_3 nonmagnetic doublet. The exponential decrease of $|\theta_{CW}|$ with y is consistent with the decrease of T_K with y inferred from the $\rho(T)$ data of Fig. 1(a). For Kondo systems, $\theta_{CW} \approx 3$–$5\ T_K$. The value of θ_{CW} appears to saturate to a value ≈ -57 K for large y. A comparable x-independent value of θ_{CW} was observed for the related system $La_{1-x}U_xPd_3$ which does not show a Kondo effect. Shown in Fig. 2 is a plot of T_K versus Th concentration y (solid circles), inferred from the values of θ_{CW} according to the relation $T_K \equiv [-\theta_{CW} - 57]/4.8$. The scaling factor 4.8 was derived by defining T_K of the y = 0.1 sample to be the value obtained from the specific heat as described below.

Presented in Fig. 1(c) are plots of $\Delta C/T$ versus ln T for $Y_{0.9-y}Th_yU_{0.1}Pd_3$ between ~ 0.5 K and 20 K. The increase of the magnitude of the slope $|d(\Delta C/T)/d\ln T|$ with y is consistent with a decrease of T_K with y. The features in the $\Delta C/T$ versus ln T data at ~ 1 K (a shoulder) for the sample with y = 0.2 and at ~ 2.4 K (a maximum) for the samples with x = 0.3 and 0.4 may be due to the splitting of the Γ_3 nonmagnetic doublet ground state, which is analogous to Zeeman splitting of the magnetic doublet for a magnetic two-channel Kondo effect. It is interesting that the evolution of the shape of the $\Delta C(T)$ curves with y resembles that of the calculated $\Delta C(T)$ curves for a two channel magnetic Kondo effect in the presence of an applied magnetic field [5]. Such a splitting of the U Γ_3 doublet could arise from a local charge asymmetry or distortion of the lattice from cubic symmetry about a U site due to the substitution of Th. An analysis of the slope $|d(\Delta C/T)/d\ln T|$ within the context of the two channel Kondo model yields a decrease of T_K with y, similar to that deduced from the $\chi(T)$ data, as shown in Fig. 2 (open circles). A plot of the entropy $\Delta S(T)$, estimated in the same manner as described in Refs. [2, 3], yields a value of $R \ln 2$ near 15 K for the samples with $y = 0.3$ and 0.4, and substantially reduced values at $\sim$ 15 K for the samples with $y = 0, 0.1$, and 0.2, consistent with the existence of a residual zero temperature entropy of $(1/2)R \ln 2$. In addition, low field $\chi(T)$ measurements on $Y_{0.5}Th_{0.4}U_{0.1}Pd_3$ reveal a slight irreversibility, indicating spin-glass freezing, below $T_{SG} \approx 2.4$ K. This temperature coincides with a peak in $\Delta C(T)/T$.

Specific heat measurements on a $La_{0.9}U_{0.1}Pd_3$ sample reveal a peak in $\Delta C(T)$ near 5 K that can be described by a 2-level Schottky anomaly with a Gaussian distribution of splittings centered at 0.8 meV and a FWHM of 1.6 meV. The peak position and width of the Schottky anomaly are similar to those for the $Y_{0.5}Th_{0.4}U_{0.1}Pd_3$ sample shown in Fig. 1(c), suggesting a similar mean and width of the splitting between the U Γ_3 nonmagnetic doublet ground state for the two systems.

The electrical resistivity, magnetic susceptibility, and specific heat measurements on the $Y_{0.9-y}Th_yU_{0.1}Pd_3$ system, in which the U concentration is held constant to minimize changes in the U intersite interactions, clearly reveal the scaling of the physical properties with T_K and the phenomenon of Fermi level tuning in this system. The self consistency of the y dependence of T_K determined from the data support the interpretation of the Kondo and non-Fermi-liquid behavior in terms of a two channel quadrupolar Kondo effect.

Acknowledgements

This research was supported by the National Science Foundation under Grants No. DMR 91-07698 (UCSD) and No. DMR 91-08015 (UM) and the US Department of Energy under Grant No. DE-FG03-86ER-45230 (UCSD). Work at Los Alamos was performed under the auspices of the US Department of Energy. The SQUID magnetometer used in these studies was provided by the Center for Interface and Materials Science and funded by the W.A. Keck Foundation.

References

[1] J.-S. Kang, J.W. Allen, M.B. Maple, M.S. Torikachvili, W.P. Ellis, B.B. Pate, Z.-X. Shen, J.J. Yeh and I. Lindau, Phys. Rev. B 39 (1989) 13529.

[2] 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.

[3] 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, J. Alloys and Compounds 181 (1992) 327.

[4] H.A. Mook, C.L. Seaman, M.B. Maple, M.A. Lopez de la Torre, D.L. Cox and M. Makivic, Physica B 188 (1993) 341.

[5] P.D. Sacramento and P. Schlottmann, Phys. Rev. B 43 (1991) 13294.