Evidence for a Common Physical Description of Non-Fermi-Liquid Behavior in Chemically Substituted f-Electron Systems

M. C. de Andrade, R. Chau, R. P. Dickey, N. R. Dilley, E. J. Freeman, D. A. Gajewski, and M. B. Maple

Department of Physics and Institute for Pure and Applied Physical Sciences, University of California, San Diego, La Jolla, California 92093

R. Movshovich

Los Alamos National Laboratory, Los Alamos, New Mexico 87545

A. H. Castro Neto and G. Castilla

Department of Physics, University of California, Riverside, California 92521

B. A. Jones

IBM Almaden Research Center, San Jose, California 95120-6099

The non-Fermi-liquid (NFL) behavior observed in the low temperature specific heat $C(T)$ and magnetic susceptibility $\chi(T)$ of many chemically substituted f-electron systems is analyzed within the context of a recently developed theory based on Griffiths' singularities. Measurements of $C(T)$ and $\chi(T)$ in the systems $Th_{1-x}U_xPd_2Al_3$, $Y_{1-x}U_xPd_3$, and $UCu_{5-x}M_x$ ($M = Pd, Pt$) are found to be consistent with $C(T)/T \propto \chi(T) \propto T^{-1+\lambda}$ predicted by this model with $\lambda < 1$ in the NFL regime. These results suggest that the NFL properties observed in a wide variety of disordered f-electron systems can be described within the context of a common physical picture. [S0031-9007(98)07863-6]

Transport, thermal, and magnetic measurements on a number of chemically substituted (i.e., disordered) rare earth and actinide compounds have revealed low temperature physical properties that show striking departures from the predictions of Fermi-liquid theory [1]. Several theoretical models have been developed to account for the non-Fermi-liquid (NFL) behavior observed in f-electron materials. These models include a multichannel Kondo effect of magnetic or electric origin [2–4], fluctuations of an order parameter in the vicinity of a second order phase transition at $T = 0$ K [5–7], a disordered distribution of Kondo temperatures [8,9], and an electron polaron model for heavy fermion systems [10]. However, none of these models have been able to account for all of the NFL characteristics observed in the wide variety of systems that belong to this new class of strongly correlated f-electron materials. Three of us (A. H. C. N., G. E. C., and B. A. J.) have recently proposed a model where NFL behavior is associated with the proximity to a quantum critical point and the formation of magnetic clusters in the paramagnetic phase due to the competition between the Kondo effect and the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction in the presence of magnetic anisotropy and disorder inherent in alloyed materials [11]. This model predicts that various physical properties diverge with decreasing temperature as weak power laws of temperature and that this behavior persists over appreciable ranges of substituent concentration, similar to what has been observed in a number of f-electron materials.

In this Letter, we compare low temperature specific heat $C(T)$ and magnetic susceptibility $\chi(T)$ data derived from our own measurements on the ordered f-electron sublattice systems $UCu_{5-x}M_x$ ($M = Pd, Pt$) (Ref. [12,13]) and the disordered f-electron sublattice systems $Th_{1-x}U_xPd_2Al_3$ (Ref. [14]) and $Y_{1-x}U_xPd_3$ (Ref. [15]) with the predictions of the new model proposed in Ref. [11]. Some of the data are new and reported here for the first time; the rest of the data have been published previously [13–17]. We find that the NFL behavior of $C(T)$ and $\chi(T)$ in these compounds is consistent with the predictions of the model proposed in Ref. [11]. Specifically, $C(T)/T$ and $\chi(T)$ can be described by divergent power laws in temperature at the lowest temperatures for all of the compounds investigated.

Details of the procedures used to prepare the polycrystalline uranium compounds studied in this work are described elsewhere [13,15]. Magnetization $M(T)$ measurements were performed after field cooling a sample to the lowest temperature using a Quantum Design SQUID magnetometer in fields of 0.5 and 1 tesla and at temperatures in the range $1.8 \leq T \leq 300$ K. The low temperature $M(T)$ data ($0.4 \leq T \leq 2$ K) were acquired with a ^{3}He Faraday magnetometer (FM). Heat capacity measurements were made using a standard heat-pulse technique in ^{3}He-^{4}He dilution and ^{3}He refrigerators.

Log-log plots of C vs T in the NFL regime are shown in Fig. 1 for several samples in the $Y_{1-x}U_xPd_3$ ($x = 0.2$) and $UCu_{5-x}M_x$ ($M = Pt, x = 1$; $M = Pd, x = 1, 1.5$) systems and in Fig. 2 for the $Th_{1-x}U_xPd_2Al_3$ ($x = 0, 0.2, 0.4, 0.6$) system. In these figures, the error
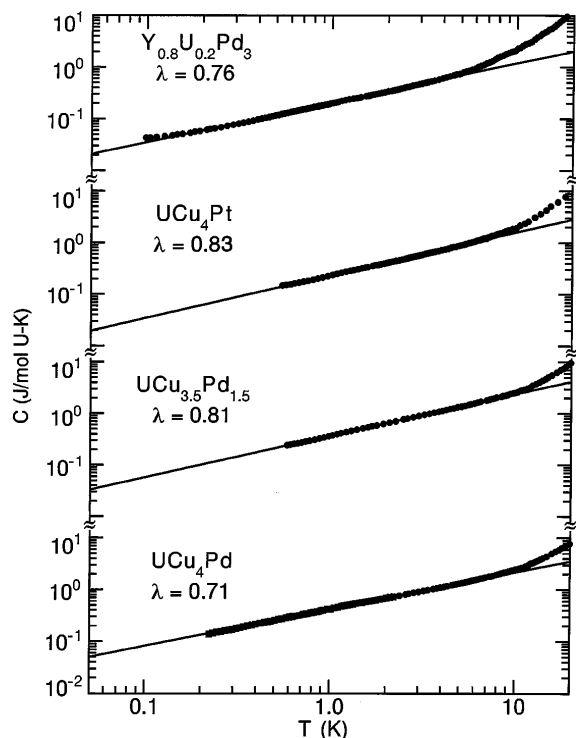
FIG. 1.   Log-log plots of specific heat C vs temperature T for several U-based NFL systems. Solid lines are fits of the data to Eq. (1). Square symbols represent data on UCu_4Pd from Ref. 19 for the temperature range $0.2 \leq T \leq 2$ K.

bars are smaller than the size of the symbols. The solid lines represent least squares fits of the expression relating the specific heat C to the magnetic susceptibility χ, given by

$$C(T)/T \propto \chi(T) \propto T^{-1+\lambda} \qquad (1)$$

at low temperatures, where λ is a parameter determined by the best fit. The values of λ for different compounds and/or different compositions x of the chemical substituent are indicated in Figs. 1 and 2 and are given in Table I. To test the quality of the fits, the reduced chi square χ_ν^2 was calculated [18] and will be discussed below. The log-log plots of C vs T (Figs. 1 and 2) reveal that a power law with $\lambda < 1$ provides an excellent description of the data for all of the systems considered. For the $Th_{1-x}U_xPd_2Al_3$ system, the power law describes the data from 0.08 K up to 8.5 K. In the $Y_{1-x}U_xPd_3$ ($UCu_{5-x}M_x$) system, the best fit was achieved for $0.1 \leq T \leq 3.6$ K ($0.2 \leq T \leq 4.3$ K). For $ThPd_2Al_3$ we obtain $\lambda = 1$ as expected for a Fermi liquid. For the UCu_4Pd system, the data from Ref. [19] were added (square symbols) in the temperature range $0.2 \leq T \leq 2$ K. The data were normalized to our data to account for the small (6%) experimental difference between the two experiments.

In order to provide a more direct comparison between power law and logarithmic behavior, semilog plots of C/T vs T are shown in Fig. 3 for three selected compounds,

UCu_4Pd, $Th_{0.4}U_{0.6}Pd_2Al_3$, and $Y_{0.8}U_{0.2}Pd_3$. The upturns in $C(T)/T$ at high temperatures are due to the phonon contributions and Schottky anomalies arising from crystalline electric field splitting of the U $5f$ Hund's rule ground state multiplet and are excluded from the fitting range. These contributions were not subtracted because they could not be estimated with sufficient accuracy. In this figure, both power law (solid lines) and logarithmic (dashed lines) temperature dependences were fit to the data over the same temperature range, and χ_ν^2 was then calculated to assess the quality of each fit. Table II shows the calculated values of χ_ν^2 for each of the compounds in Fig. 3. Recall that for an optimum fit, χ_ν^2 is close to 1 [18]. Both the logarithmic and power law descriptions of the specific heat agree with the experimental data within the experimental resolution, and the χ_ν^2 values in Table II indicate that a power law provides a comparable (e.g., $Th_{0.4}U_{0.6}Pd_2Al_3$) and, perhaps, even slightly better description (e.g., $Y_{0.8}U_{0.2}Pd_3$ and UCu_4Pd) of the data than a logarithmic relation over the same temperature range.

Log-log plots of the magnetic susceptibility $\chi(T)$ vs T for UCu_4Pt, $Th_{0.4}U_{0.6}Pd_2Al_3$, and $Y_{0.8}U_{0.2}Pd_3$ are shown in Fig. 4. The data have been fitted with power laws [Eq. (1)] and give values of λ, listed in Table I, that are close to those obtained from the power law fits to the $C(T)$ data. The magnetic susceptibility was obtained from the linear part of the magnetization M vs magnetic field H isotherm at low fields $\mu H/kT \ll 1$, where μ is the
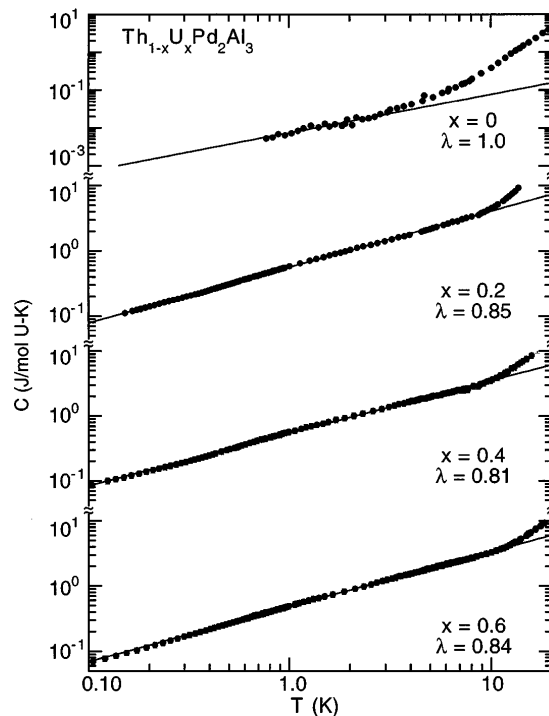

FIG. 2.   Log-log plots of specific heat C vs temperature T for the $Th_{1-x}U_xPd_2Al_3$ system for several values of x. Solid lines are fits of the data to Eq. (1).

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TABLE I. Exponent λ obtained from fits of Eq. (1) to specific heat (λ_C) and magnetic susceptibility (λ_χ) data for the f-electron systems shown in Figs. 1 and 2.

	Th$_{1-x}$U$_x$Pd$_2$Al$_3$						UCu$_{5-x}$Pd$_x$			
x	0	0.2	0.4	0.6	0.8	0.9	1.0	1.5	UCu$_4$Pt	Y$_{0.8}$U$_{0.2}$Pd$_3$
λ_C	1	0.85	0.81	0.84	0.81	1	0.71	0.81	0.83	0.76
λ_χ	~1	0.6	0.63	0.63	0.6	$\cdots$	0.72	0.78	0.77	0.70

magnetic moment and k is Boltzmann's constant. This procedure was especially important at low temperatures where the M vs H curves display negative curvature which is more pronounced at lower temperatures. Assuming the negative curvature of $M(H)$ is intrinsic, this method of extracting $\chi(T)$ suggests that the origin of the NFL behavior is magnetic in nature, since $C(T)/T$ and $\chi(T)$ have the same temperature dependence. We also note that the values of λ obtained from magnetization measurements on the Th$_{1-x}$U$_x$Pd$_2$Al$_3$ system reflect the average of the magnetization over crystallites with a hexagonal structure oriented in different directions, which may explain the difference between the values of λ from specific heat and susceptibility in this system. In a single crystal, we expect better agreement between the specific heat and the magnetization [20].
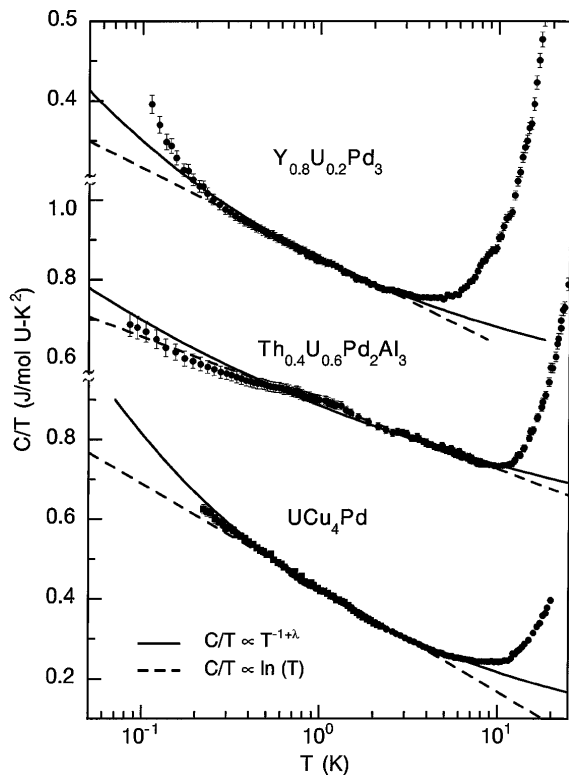

FIG. 3. Semilog plots of specific heat C/T vs temperature T for selected U-based NFL compounds. Square symbols represent data on UCu$_4$Pd from Ref. 19 in the temperature range $0.2 \leq T \leq 2$ K. Solid and dashed lines are fits to the expressions given in the figure.

We have shown from our experimental data that the NFL behavior observed in $C(T)$ and $\chi(T)$ given in Eq. (1) is consistent with the existence of Griffiths' singularities. The values of λ obtained from the $C(T)$ and $\chi(T)$ data agree with one another within experimental resolution, suggesting that the NFL behavior originates from magnetic interactions. The U-based systems investigated in this work have all of the prerequisites for the Griffiths' phase scenario: the Kondo effect, RKKY interactions between the U magnetic moments, magnetic anisotropy due to spin-orbit interactions, and disorder associated with the chemical substitutions. Furthermore, the imaginary part of the frequency dependent susceptibility $\chi''(\omega)$ of UCu$_{3.5}$Pd$_{1.5}$ and UCu$_4$Pd, derived from neutron scattering measurements on these materials [21], is described well by the Griffiths' phase result $\chi''(\omega) \propto \omega^{-1+\lambda} \tanh(\omega/T)$ [11] with a value $\lambda \approx 0.7$, in good agreement with the values of λ determined from the above analysis of the $C(T)$ and $\chi(T)$ measurements. Nuclear magnetic resonance (NMR) and muon spin rotation (μSR) experiments [8] on these same compounds reveal a distribution of susceptibilities given by $\delta\chi/\chi \propto T^{-\lambda/2}$ which is also consistent with the presence of a Griffiths' phase at low temperatures [11]. Finally, recent NMR and μSR experiments on UCu$_{5-x}$Pd$_x$ [8] and pressure experiments on the NFL systems CeRh$_2$Si$_2$ and CeRh$_{2-x}$Ru$_x$Si$_2$ [22] indicate that disorder plays an especially important role in NFL behavior. In addition, the relatively large range of substituent concentration x over which the NFL behavior extends in certain NFL systems is consistent with the notion of a Griffiths' phase since these systems have rather large values of T_K ($\lesssim 10^2$ K) and the range of x over which the Griffiths' phase extends is predicted to be larger for larger values of T_K [11]. Thus, there is sufficient evidence to suggest that the Griffiths' phase model is a viable candidate for the NFL properties of the f-electron systems investigated herein. Moreover, recent calculations yield a linear temperature dependence of the resistivity in these systems due to electron scattering by magnetic clusters [23] in agreement with transport data [1].

TABLE 2. Calculated values of the reduced chi square χ_ν^2 from fits to specific heat data shown in Fig. 3.

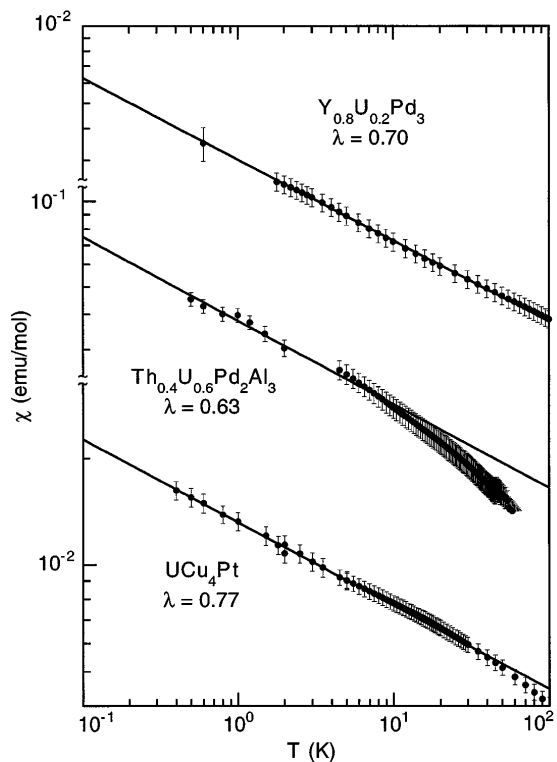|	Th$_{0.4}$U$_{0.6}$Pd$_2$Al$_3$	UCu$_4$Pt	Y$_{0.8}$U$_{0.2}$Pd$_3$
$C/T \propto \ln T$	0.31	1.70	3.48
$C/T \propto T^{-1+\lambda}$	0.94	1.07	0.97

FIG. 4. Log-log plots of magnetic susceptibility χ vs temperature T for selected U-based NFL compounds. Solid lines are fits of the data to Eq. (1).

We point out that in some of these systems it is also possible that a quadrupolar Kondo effect, due to the exchange interaction between the quadrupolar moment of a Γ_3 nonmagnetic doublet ground state of U and the conduction electrons, may occur and lead to NFL behavior [16]. However, it is not clear that the multichannel Kondo and Kondo disorder models, which are single ion models, are applicable at the relatively large substituent concentrations where NFL behavior is often observed in f-electron materials. Models based on the existence of quantum critical points are known to produce power laws in thermodynamic properties [5]. The NFL behavior can be interpreted as a generic feature in the vicinity of a quantum critical point [7]. However, in the absence of disorder and at zero temperature, the quantum critical regions reduce to points in the phase diagram and are not general enough to explain the broad range of substituent concentrations over which NFL behavior is observed. While each of these models has varying degrees of success in describing the temperature dependence of the physical properties in these materials, none have the general applicability to a wide range of

f-electron systems, nor the apparent internal consistency found in our analysis in terms of Griffiths' singularities.

[1] See, for instance, *Proceedings of the Conference on Non-Fermi Liquid Behavior in Metals, Santa Barbara, 1996,* edited by P. Coleman, M. B. Maple, and A. Millis [J. Phys. Condens. Matter **8** (1996)].

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