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Non-Fermi liquid behavior and quantum criticality in $Sc_{1-x}U_xPd_3$ and $URu_{2-x}Re_xSi_2$

M.B. Maple[a,*], N.P. Butch[a], E.D. Bauer[a,1], V.S. Zapf[a,1], P.-C. Ho[a], S.D. Wilson[b], Pengcheng Dai[b,c], D.T. Adroja[d], S.-H. Lee[e], J.-H. Chung[e,f], J.W. Lynn[e]

[a]*Department of Physics and Institute for Pure and Applied Physical Sciences, University of California, San Diego, La Jolla, CA 92093, USA*
[b]*Department of Physics and Astronomy, The University of Tennessee, Knoxville, TN 37996-1200, USA*
[c]*Condensed Matter Sciences Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, USA*
[d]*ISIS Facility, Rutherford Appleton Laboratory, Chilton, Didcot, Oxon OX11 0QX, UK*
[e]*NIST Center for Neutron Research, National Institute of Standards and Technology, Gaithersberg, MD 20899, USA*
[f]*Department of Materials Science and Engineering, University of Maryland, College Park, MD 20742, USA*

Abstract

The compounds $Sc_{1-x}U_xPd_3$ and $URu_{2-x}Re_xSi_2$ exhibit non-Fermi liquid behavior in the vicinity of zero-temperature magnetic phase transitions accessed by chemical substitution. Notably, the quantum phase transitions in these pseudobinary systems are associated with spin glass and ferromagnetic ordering, respectively. Recent studies have uncovered new details of the unconventional physical phenomena occurring in these materials.

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1. Introduction

Quantum criticality is one of the most challenging problems in the physics of f-electron materials. Many experimental investigations on f-electron materials have revealed non-Fermi liquid (NFL) behavior in physical properties in the vicinity of a critical pressure, magnetic field, or composition at which a second-order magnetic phase transition is suppressed to 0 K (quantum critical point, QCP). Most of the studies have been made on f-electron systems that undergo antiferromagnetic (AFM) transitions, while relatively few studies have been made on f-electron systems that display ferromagnetic (FM) or spin glass (SG) transitions. Furthermore, the FM transitions that have been studied appear to become first order near the QCP, at least for stoichiometric compounds (e.g., UGe_2, and the d-electron systems MnSi and $ZrZn_2$). In this paper, we summarize our recent investigations of NFL behavior near QCPs accessed by chemical substitution in the U-based f-electron systems $Sc_{1-x}U_xPd_3$ and $URu_{2-x}Re_xSi_2$, which have SG and FM QCPs, respectively.

2. $Sc_{1-x}U_xPd_3$

Motivated by evidence for the occurrence of the quadrupolar Kondo effect (QKE) in $Y_{1-x}U_xPd_3$ [1,2], studies of other $M_{1-x}U_xPd_3$ systems, where M is tri- or tetravalent, were attempted [3]. The most rewarding of these investigations, in terms of NFL research, has been the study of $Sc_{1-x}U_xPd_3$ [4]. Remarkably, the phase diagram of this system (Fig. 1) closely resembles that of $Y_{1-x}U_xPd_3$, with the Sc-based compounds displaying SG and AFM ordering at slightly higher values of x than the Y compounds, in addition to having a higher Kondo

*Corresponding author. Tel.: +1 858 534 3969; fax: +1 858 534 1241.
E-mail address: mbmaple@physics.ucsd.edu (M.B. Maple).
[1]Present address: Los Alamos National Laboratory, Los Alamos, NM 87545, USA.

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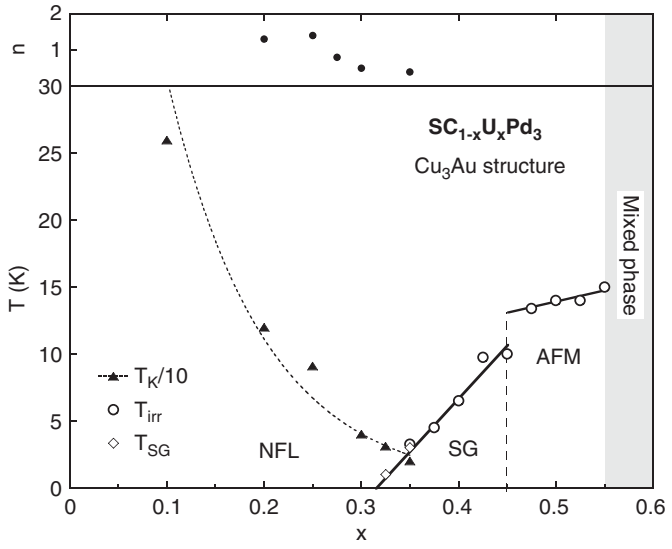

Fig. 1. $Sc_{1-x}U_xPd_3$ T–x phase diagram, including the compositional dependence of the $\rho(T)$ exponent $n(x)$. After Ref. [5].

temperature T_K at any value of x in the paramagnetic state. Given that Sc and Y are isoelectronic, this similarity may not seem surprising, but it should be noted that $La_{1-x}U_xPd_3$ does not display Kondo or NFL behavior [3,5]. Practically, the significance of the correspondence between the Sc and Y compounds lies in the microscopic compositional inhomogeneity in $Y_{1-x}U_xPd_3$, which has complicated the interpretation of its physical properties [5,6]. In contrast, $Sc_{1-x}U_xPd_3$ is metallurgically well-behaved, making it possible to study the same NFL behavior in a homogeneous system [5]. This also underscores the robustness of the NFL properties in these materials, which persist in the Y-based compositions despite microstructural issues.

For low values of x, the physical phenomena are understood in terms of single-ion Kondo behavior. Because Sc^{3+} is smaller than Y^{3+}, the magnitude of the local-itinerant electron exchange interaction is greater in $Sc_{1-x}U_xPd_3$, and thus T_K is larger for a given value of x. Furthermore, as x increases, trivalent Sc is replaced by tetravalent U, increasing the number of conduction electrons, which raises the Fermi energy E_F and thus the local 5f electron binding energy on the U sites. This phenomenon, called Fermi level tuning, leads to the observed decrease in T_K [2,7].

For low values of x, the physical properties of $Sc_{1-x}U_xPd_3$ deviate significantly from the predictions of Fermi liquid theory. As with $Y_{1-x}U_xPd_3$, the U contribution to the electrical resistivity, electronic specific heat (Fig. 2), and magnetic susceptibility can be described by

$$\Delta\rho(T)\sim 1 - a(T/T_K), \tag{1}$$

$$\Delta C(T)/T\sim -(1/bT_K)\ln(T/T_K), \tag{2}$$

$$\Delta\chi(T)\sim 1 - c(T/T_K)^{1/2}. \tag{3}$$

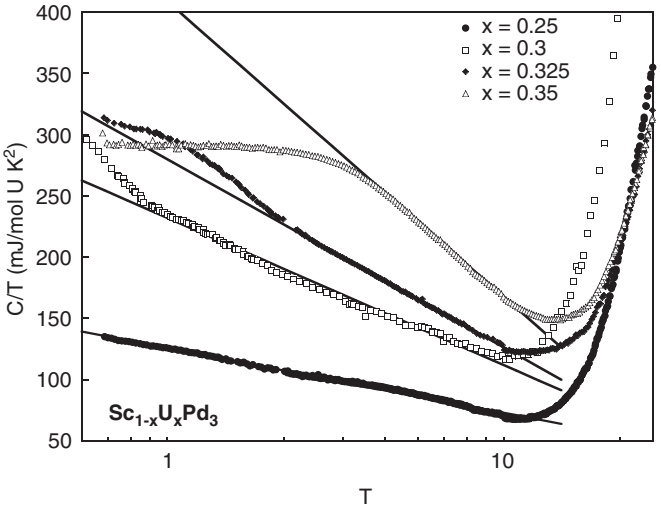

Fig. 2. Electronic specific heat $C(T)/T$ of $Sc_{1-x}U_xPd_3$. The lines are logarithmic fits to the data. After Ref. [8].

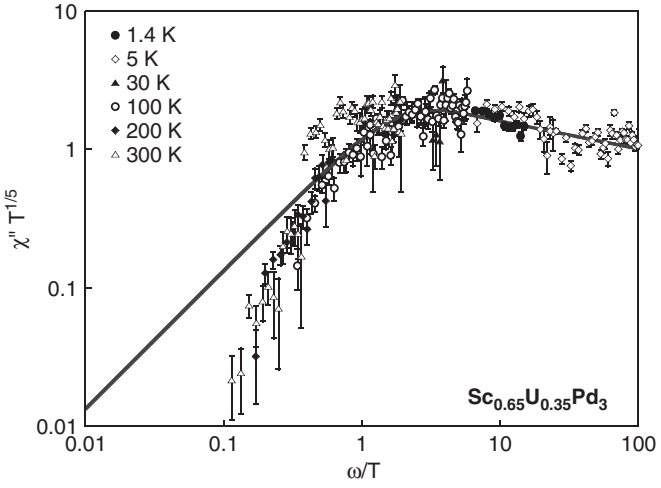

Fig. 3. Scaling plot for $Sc_{0.65}U_{0.35}Pd_3$. The curve is the scaling function for an AFM QCP proposed by Si et al. [33]. After Ref. [11].

Thus, it was found that the Kondo, or a similar, energy scale is still relevant for compounds near and in the SG phase. Electronic specific heat was found to follow the form of Eq. (2) for T above the SG transition, while for $0.1\,K < T < 2.5\,K$, the electrical resistivity obeys

$$\Delta\rho(T)\sim 1 - a(T/T_K)^n, \tag{4}$$

where n ranges from 1.3 to 0.5 (Fig. 1) [8]. Eqs. (2) and (3) agree with the QKE model, which predicts $\rho(T)\sim -\frac{1}{2}(T/T_K)^{1/2}$, $C(T)/T\sim -\ln(T/T_K)$, and $\chi(T)\sim -(T/T_K)^{\frac{1}{2}}$ [9,10]. Although most measurements of $\rho(T)$ clearly disagree with the QKE predictions, surprisingly, for concentrations in the vicinity of the onset of SG, the low-T resistivity can indeed be fit by Eq. (4) with $n = 0.5$.

Recent inelastic neutron scattering measurements have shed new light on the curious behavior of $Sc_{1-x}U_xPd_3$ [11]. Magnetic excitations in the $x = 0.35$ compound are broad in energy $\hbar\omega$, T-independent, and wave vector q-independent, consistent with the single-ion scenario. However,

even more compelling is the discovery that the imaginary part of the dynamical susceptibility $\chi''(q,\omega,T)$ scales with ω/T for all studied q, ω, and T with a scaling exponent $\alpha=\frac{1}{5}$ (Fig. 3). This scaling indicates that temperature is the only relevant energy scale and reflects the influence of the SG QCP. Because similar scaling has been observed in $UCu_{5-x}Pd_x$ ($\alpha=\frac{1}{3}$) [12,13] and $CeCu_{6-x}Au_x$ ($\alpha=0.75$) [14], which have AFM QCPs, it is likely that the NFL behavior in all three systems is tied to quantum criticality, specifically related to local moment fluctuations, regardless of the nature of the ordered phase. This assertion is underscored by the dominant role played by Kondo impurity-type physics in $Sc_{1-x}U_xPd_3$, even at high values of x.

3. $URu_{2-x}Re_xSi_2$

Although the compound URu_2Si_2 has been intensely studied for two decades, a complete understanding of its physical properties has proven elusive. The observation of heavy fermion superconductivity (SC) below ~1 K in URu_2Si_2 [15–17] sparked a period of research that quickly focused on the nature of a phase transition with a large $C(T)$ jump that occurs at ~17 K, initially believed to be associated with AFM or spin-density wave (SDW) ordering [17]. Since those early studies, the compound has been examined by all manner of physical probes and subjected to high magnetic fields, high pressures, and chemical substitution.

Undoubtedly, URu_2Si_2 maintains its popularity due to its mysterious ordered (hidden order, HO) phase below 17 K. Studies performed under pressure have focused on discovering the nature of this ordered phase. For instance, the first resistivity measurements under pressure [18] supported the idea of SDW order competing with SC for the Fermi surface, which was originally inferred from specific heat measurements of the respective energy gaps. More recently, neutron-scattering measurements under pressure identified a phase transition from a small-moment HO phase to a true AFM state with a larger staggered moment [19]. Subsequent NMR measurements under pressure suggest that the AFM staggered moment is not changed by pressure, but that the macroscopic volume fraction occupied by the true AFM phase increases with pressure [20]. However, this scenario may be inconsistent with studies of thermal expansion [21] and the deHaas–van Alphen effect under pressure [22], as well as recent neutron scattering at ambient pressure [23]. Complementing these investigations, at ambient pressure and in high magnetic fields, neutron scattering, transport, and magnetization measurements have revealed a complex phase diagram, with several novel phases occurring in the vicinity of the QCP (~35 T) where the HO phase is suppressed, including what could be reentrant hidden order at even higher fields [24–26].

Attempts to gain insight into the unusual physical properties of URu_2Si_2 by substituting transition metal elements for Ru have led to interesting discoveries. Rhodium substitution has yielded many insights into the nature of the URu_2Si_2 HO phase [27]. Neutron scattering studies of $URu_{2-x}Rh_xSi_2$ have identified a rich phase diagram, and recent low-doping investigations lend support to the small-volume fraction AFM explanation of the $x=0$ HO phase [28]. At low Rh concentrations and high fields, it was found that even in the absence of the HO phase, indications of a QCP at high field still persist, including the ordered phase that forms in its vicinity to 'protect' the system from entering a highly-degenerate ground state [29].

Doping with Re, Tc, or Mn results in suppression of the HO and SC phases, and leads to FM order at intermediate concentrations [30]. The FM is itinerant in nature, with a maximum ordered moment of $\sim 0.45\mu_B$, while the paramagnetic effective moment is $\sim 1.5\mu_B$ [31]. Notably, these investigations constitute the first observation of a FM instability in a heavy fermion system (Fig. 4). Moreover, these experiments suggest the existence of a sharp electronic density of states at E_F in URu_2Si_2, and, along with studies of substitution of Os, Rh, and Ir, corroborate the idea of competition for electronic states between the HO and SC phases [27].

In $URu_{2-x}Re_xSi_2$ at low temperatures, NFL behavior is observed in the specific heat and electrical resistivity for $0.15\leqslant x\leqslant 0.6$ [32]. The resistivity and specific heat can be described by

$$\rho(T)\sim\rho_0+aT^n, \tag{5}$$

over 1–2 decades in T, and

$$C(T)/T\sim\gamma_0-c_0\ln T, \tag{6}$$

over ~1 decade in T. For $0.15\leqslant x\leqslant 6.2$, below 5 K the magnetic susceptibility (not shown) can be fit by a

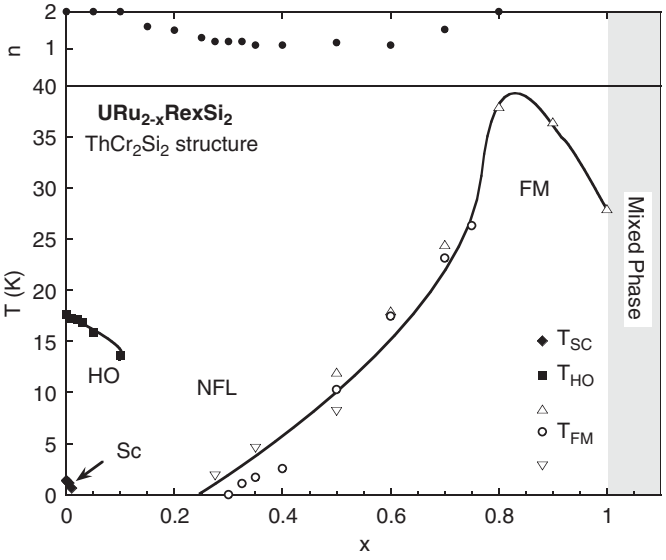

Fig. 4. $URu_{2-x}Re_xSi_2$ T–x phase diagram, including the compositional dependence of the $\rho(T)$ exponent $n(x)$. After Ref. [32].

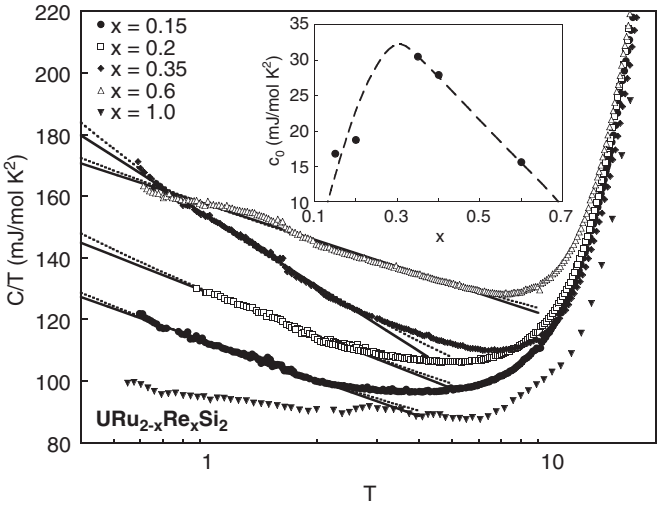

Fig. 5. Electronic specific heat $C(T)/T$ of $URu_{2-x}Re_xSi_2$. The solid lines are logarithmic fits to the data. The inset shows the compositional dependence of the coefficient of the logarithmic term $c_0(x)$. After Ref. [32].

power law

$$\chi(T) \propto T^{-b}, \tag{7}$$

where b increases from ~0.2 to ~0.4 as x increases. The dependence on x of the resistivity exponent n is displayed in Fig. 4, while $C(T)/T$ is shown in Fig. 5, along with a plot of the dependence of the magnitude c_0 of the logarithmic contribution to $C(T)$. Note that $n(x)$ and $c_0(x)$ deviate maximally from their Fermi liquid values ($n = 2$, $c_0 = 0$) at $x \approx 0.3$, in the vicinity of the FM quantum phase transition, strongly suggesting that the NFL behavior is associated with quantum critical fluctuations. More importantly, these results provide the first strong evidence for the occurrence of NFL behavior over at least a decade in temperature well within a FM ordered state, at $x = 0.6$.

4. Summary

The compounds $Sc_{1-x}U_xPd_3$ and $URu_{2-x}Re_xSi_2$ exhibit unconventional power law and logarithmic T-dependences in their physical properties, which are linked to the influence of quantum phase transitions in their T–x phase diagrams. These results are arguably even more interesting because the relevant ordered phases are SG and FM, respectively, in contrast to most known systems exhibiting AFM QCPs.

Acknowledgment

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