Quantum Critical Scaling and the Origin of Non-Fermi-Liquid Behavior in $Sc_{1-x}U_xPd_3$

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We used inelastic neutron scattering to study magnetic excitations of $Sc_{1-x}U_xPd_3$ for U concentrations ($x = 0.25, 0.35$) near the spin glass quantum critical point (QCP). The excitations are spatially incoherent, broad in energy ($E = \hbar\omega$), and follow ω/T scaling at all wave vectors investigated. Since similar ω/T scaling has been observed for $UCu_{5-x}Pd_x$ and $CeCu_{6-x}Au_x$ near the antiferromagnetic QCP, we argue that the observed non-Fermi-liquid behavior in these f-electron materials arises from the critical phenomena near a $T = 0$ K phase transition, irrespective of the nature of the transition.

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The breakdown of Fermi-liquid theory has been observed in a class of strongly correlated f-electron materials, following the original discovery of this so-called non-Fermi-liquid (NFL) behavior in the $Y_{1-x}U_xPd_3$ pseudobinary alloy in 1991 [1–4]. In spite of intensive theoretical and experimental efforts over the past decade [5–13], it is still unclear whether the observed NFL behavior is an intrinsic property [14,15] or extrinsic property associated with metallurgical inhomogeneity in these materials [16]. Models describing this anomalous NFL behavior include single-ion physics of noninteracting local magnetic moments [5,6], close proximity to a $T = 0$ K second-order phase transition or quantum critical point (QCP) [7–10], and disorder induced effects [11–13].

We studied $Sc_{1-x}U_xPd_3$ because this system has a phase diagram and NFL properties similar to $Y_{1-x}U_xPd_3$, but with a nearly homogeneous U distribution in the $ScPd_3$ matrix [14]. While the U inhomogeneity in $Y_{1-x}U_xPd_3$ is unlikely the main cause of the NFL behavior [15], neutron-scattering experiments seeking to provide constraints on various microscopic models have reached different conclusions. According to Lea, Leask, and Wolf [17], the cubic crystalline electric field (CEF) of $ScPd_3$ splits the U^{4+} $J = 4$ multiplet into Γ_4 and Γ_5 triplets, a Γ_1 singlet, and a Γ_3 doublet. If the single-ion based two-channel quadrupolar Kondo effect (QKE) is responsible for the NFL behavior in $Y_{0.8}U_{0.2}Pd_3$ [1], the U^{4+} ground state should be a nonmagnetic Γ_3 with magnetic Γ_5 and Γ_4 excited states [5]. In contrast, polarized triple-axis neutron-scattering experiments on $Y_{1-x}U_xPd_3$ reveal a magnetic ground state for $x = 0.45$ and possibly for $x = 0.2$, thus precluding the possibility of a QKE [18]. However, based on subsequent neutron time-of-flight measurements, Bull *et al.* [19] argue that the Γ_3 doublet ground state is more consistent with the $x = 0.45$ data and the $x = 0.2$ compound has a degenerate Γ_3 and Γ_5 ground state. In this case, the QKE could be the predominant cause of NFL behavior [15].

In this Letter, we report neutron-scattering experiments on $Sc_{1-x}U_xPd_3$ ($x = 0.0, 0.25, 0.35$). We show that magnetic excitations at the NFL concentration ($Sc_{0.65}U_{0.35}Pd_3$) do not form the distinct CEF excitations seen in $Y_{0.55}U_{0.45}Pd_3$. Instead, the susceptibility $\chi''(q, \omega, T)$ at all probed wave vectors (q), temperatures (T), and energies ($\hbar\omega$) obeys ω/T scaling indicative of a $T = 0$ K second-order phase transition. While such behavior is also observed in the NFL compounds $CeCu_{6-x}Au_x$ [20], $UCu_{5-x}Pd_x$ [21,22], and $Ce(Rh_{0.8}Pd_{0.2})Sb$ [23] near antiferromagnetic (AF) QCP, $\chi''(q, \omega, T)$ in ($x = 0.35$) is wave vector independent with no spatial correlations and obeys ω/T scaling over a much wider energy range with a different critical exponent. Therefore, the dynamics of isolated U ions are responsible for the temperature and energy scaling, suggesting that the NFL behavior originates from the spin-glass phase transition suppressed to near zero temperature.

Our experiments were performed on the HET time-of-flight spectrometer at the UK ISIS spallation neutron source [19], and on the BT-2 and cold neutron SPINS triple-axis spectrometers at the NIST Center for Neutron Research (NCNR). The HET data were collected with incident beam energies (E_i) of both 18 and 65 meV for a range of temperatures, and a vanadium standard was used to normalize the scattering intensity to absolute units. The magnetic scattering in U-doped materials was determined by comparing the scattering intensity with that of the nonmagnetic $ScPd_3$ parent compound for 18 meV data, and by subtraction of a parent-compound-generated mapping

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background for 65 meV data [19,24]. To study the low-energy spin dynamics, we used SPINS with final neutron energy fixed at $E_f = 5$ meV. An incident beam collimation of $80'$ was used followed by a cold Be filter and a radial collimator after the sample. We also collected data using polarized neutrons on BT-2 to separate the magnetic signal from nuclear spin incoherent scattering. For the experiment, we prepared 18 g polycrystalline samples of $Sc_{1-x}U_xPd_3$ ($x = 0$, 0.25, 0.35) through arc-melting techniques [14]. The lattice parameters of these cubic Cu_3Au structure materials are $a = b = c = 4.01$ Å for $x = 0.35$ and 3.99 Å for $x = 0.25$.

Figure 1 summarizes HET results with $E_i = 18$ meV for $Sc_{1-x}U_xPd_3$ at $x = 0$, 0.25, 0.35, where we probed excitations in the energy range between 3 and 13 meV. The scattering for $x = 0.35$ in the energy-momentum (E-q) space probed [Fig. 1(a)] shows a broad continuum of intensity with no peak at the expected AF ordering wave vector for $Y_{0.55}U_{0.45}Pd_3$ marked as the vertical dashed line [18]. Different energy-integrated cuts at 5 K [see inset of Fig. 1(a)] show no modulation at any wave vector, different from that of $Y_{1-x}U_xPd_3$ [Fig. 10 of Ref. [19]). To see if the scattering in Fig. 1(a) is magnetic, we compare q-integrated energy cuts for all three concentrations at 5 K [Fig. 1(b)]. While the outcome shows clear magnetic response for the two doped systems, the scattering is broad and featureless with no evidence for localized CEF states. In addition, the magnetic scattering does not follow the U-concentration scaling. Assuming that the magnetic fluctuations in $Sc_{1-x}U_xPd_3$ scale linearly with the U solute concentration, one would expect scattering for $x = 0.25$ as the dashed line in Fig. 1(b). The actual scattering from the $x = 0.25$ concentration instead almost lies directly on top of the nonmagnetic parent background with much less magnetic signal. We note that similar behavior has also been observed in $Y_{1-x}U_xPd_3$ [19].

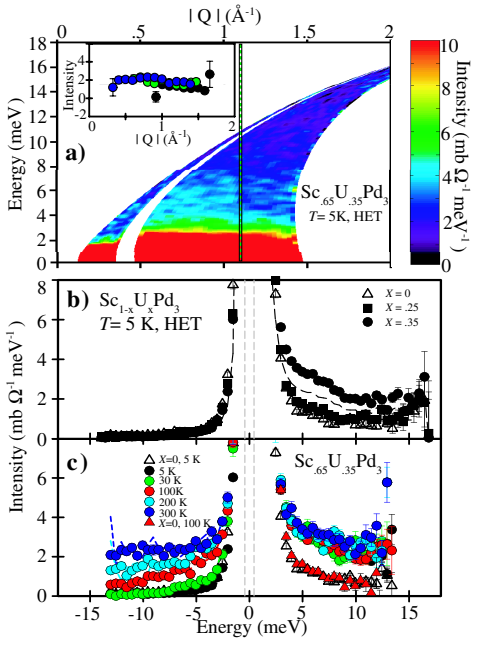

FIG. 1 (color online). (a) E-q image of $Sc_{1-x}U_xPd_3$ ($x = 0.35$) at 5 K for $E_i = 18$ meV. The inset shows E-integrated [$E =$ 3–6 meV (top circles); 6–9 meV (middle circles); 9–12 meV (bottom filled circles)] q cuts at 5 K. (b) The q-integrated ($0 < q < 2.5$ Å^{-1}) $S(\omega, T)$ for $x = 0.0$, 0.25, and 0.35. The dashed line shows the expected magnetic scattering for $x = 0.25$ assuming simple U-concentration scaling. (c) The q-integrated ($0 < q < 1.5$ Å^{-1}) $S(\omega, T)$ for $x = 0.35$ at various temperatures. Dashed lines on the $E < 0$ side reflect detailed balance expectations calculated from the magnetic scattering on the $E > 0$ side. Grey, vertical dashed lines show the resolution half-width of 0.433 meV.

Since the $Sc_{1-x}U_xPd_3$ $x = 0.25$ compound is nearly nonmagnetic and does not exhibit strong NFL features [15], we focus on the NFL $x = 0.35$ compound and study the temperature evolution of the magnetic scattering. The most striking feature of the data is the temperature independence on the neutron energy loss side of the spectra, while the neutron energy gain side obeys detailed balance as shown in Fig. 1(c). To confirm that such behavior indeed arises from the U magnetic moment, we performed a careful study of the temperature dependence of the nonmagnetic $ScPd_3$ and found that the nonmagnetic scattering is temperature independent below 100 K and increases only slightly at 300 K [Fig. 1(c)].

To determine the magnetic excitations of the $x = 0.35$ compound above 13 meV, we increased $E_i = 65$ meV at the HET. Figure 2(a) shows the scattering at $T = 5$ K for

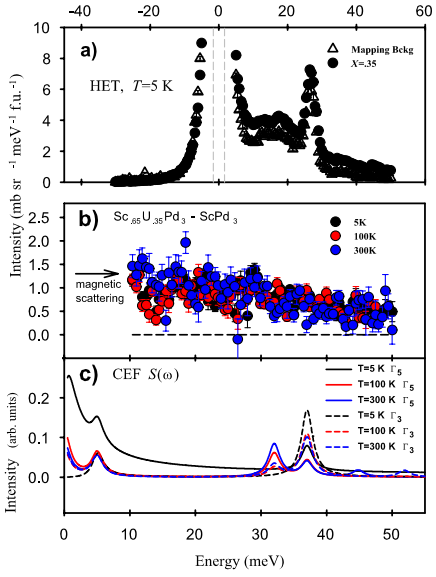

FIG. 2 (color online). (a) q-integrated ($0 < q < 2.5$ Å^{-1}) $S(\omega, T)$ for $x = 0.35$ with $E_i = 65$ meV. Open triangles show the mapping background, and vertical dashed lines are the resolution half-width of 1.65 meV. (b) Net magnetic scattering at various temperatures. (c) Calculated CEF $S(\omega, T)$ for two possible ground states: magnetic Γ_5 and nonmagnetic Γ_3.

both the $x = 0.35$ compound and the nonmagnetic background [19,24]. The resulting difference spectra at several temperatures are shown in Fig. 2(b). Similar to Fig. 1(c), the magnetic excitations are broad, temperature independent, and extend up to 50 meV. If excitations from the U moments in the $x = 0.35$ compound have localized states at ~6 meV and ~36 meV as in the AF ordered $Y_{0.55}U_{0.45}Pd_3$ [18,19], one can calculate the expected temperature dependence of the CEF levels assuming either Γ_5 [18] or Γ_3 [19] as the zero-energy ground state [Fig. 2(c)]. The comparison of Figs. 2(b) and 2(c) reveals that both CEF models are incompatible with the data.

If excitations in the NFL $x = 0.35$ are indeed nonlocalized, one would expect to find magnetic scattering at energies much less than 3 meV. Figure 3(a) shows energy scans at $q = 1.3$ Å^{-1} for $x = 0.35$ and $x = 0.0$ using SPINS at NCNR. Consistent with results at higher energies (Figs. 1 and 2), magnetic excitations between 0.4 and 8 meV are broad, featureless, and temperature independent from 1.4 to 300 K. To see if magnetic scattering in $x = 0.35$ peaks at the same AF wave vector as $Y_{0.55}U_{0.45}Pd_3$ [18], we carried out a series of energy scans at different wave vectors at $T = 1.4$ K. The outcome shows no enhancement along any wave vectors probed [Fig. 3(b)]. To see if there is magnetic scattering at an arbitrary ($q = 1.95$ Å^{-1}) elastic position, we performed polarized neutron beam measurements on $x = 0.35$ at $T = 5$ K using BT-2 at NCNR. The flipping ratios for both horizontal and vertical guide fields are ~20. By subtracting vertical field intensity from that in horizontal field, we confirmed the presence of elastic magnetic scattering [18,25]. To further prove that the observed excitations are from U moments, we show in Fig. 3(c) the wave vector dependence of the magnetic scattering from both HET and SPINS experiments normalized to the expected U^{4+} and U^{3+} magnetic form factors. The data are clearly consistent with U magnetic scattering.

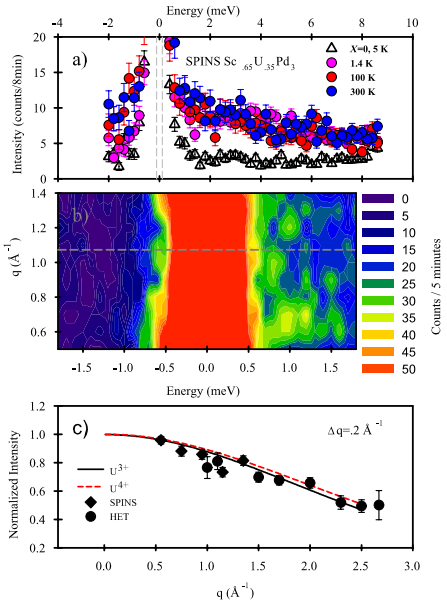

FIG. 3 (color online). (a) Energy scans at fixed $q = 1.3$ Å^{-1} for $x = 0.35$ and 0.0 on SPINS. Dashed vertical lines show the resolution half-width of 0.12 meV. (b) (q, E) map of magnetic fluctuations for $x = 0.35$ at 1.4 K. The dashed grey line shows the AF ordering wave vector of $(0.5, 0.5, 0)$ for $Y_{0.55}U_{0.45}Pd_3$ [18]. (c) q dependence of magnetic excitations. Solid lines show calculated magnetic form factors for U^{3+} and U^{4+} ions.

The absence of any characteristic q and E scale in the magnetic excitations of the $x = 0.35$ compound suggests that isolated U ions are responsible for the observed spin dynamical behavior. The unique temperature independent form of the magnetic scattering $S(q, \omega, T)$ bears a remarkable resemblance to that of $UCu_{5-x}Pd_x$, where the excitations at all q, and for limited temperatures and energies (< 25 meV) accessed display the same type of NFL ω/T scaling [21,22]. The measured $S(q, \omega, T)$ is related to the imaginary part of the dynamical susceptibility, $\chi''(q, \omega, T)$, via $S(q, \omega, T) = \chi''(q, \omega, T)/[1 - \exp(-\hbar\omega/k_BT)]$, where $[n(\omega) + 1] = 1/[1 - \exp(-\hbar\omega/k_BT)]$ is the Bose population factor. In calculating $\chi''(q, \omega, T)$ for the various temperatures, we find that χ'' multiplied by $T^{1/5}$ collapses onto a single curve for all data sets as a function of ω/T.

Figure 4 shows the outcome of our analysis, where the SPINS data have been scaled to the absolute scale of the HET data through normalizing the elastic incoherent scattering of $x = 0.0$ and 0.35. In the final plot, all data have been corrected for their magnetic form factor dependence, which is critical for the time-of-flight data because of the coupled E-q values. The obtained scaling exponent of 1/5 represents a purely empirical analysis; however, slight deviations from this value induce substantial discontinuities in the resulting ω/T scaling plot. For comparison, the scaling exponent of $UCu_{5-x}Pd_x$ is 1/3 [21,22].

The discovery of ω/T scaling in the NFL $x = 0.35$ compound strongly suggests that the magnetic fluctuations in this system arise from the close proximity to a $T = 0$ K phase transition. Similar ω/T scaling was first identified in the NFL $UCu_{5-x}Pd_x$ system, but with much smaller energy

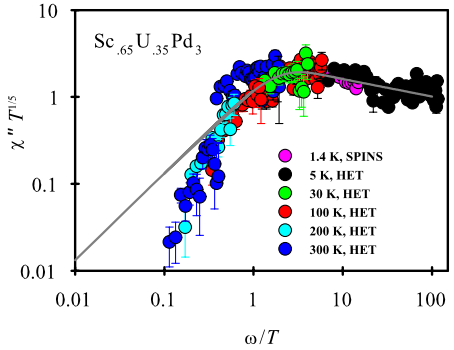

FIG. 4 (color online). Scaling plot for $Sc_{0.65}U_{0.35}Pd_3$. The 300 K HET data exhibit a slight deviation due possibly to underestimation of phonon contributions.

range [21,22]. The key difference, however, is that $Sc_{0.65}U_{0.35}Pd_3$ does not have any enhancement in the magnetic scattering around the expected AF ordering vector of higher U concentrations [18]. While the antiferromagnetism in $Y_{1-x}U_xPd_3$ compounds with $x \geq 0.41$ may not control the spin dynamics for $Sc_{0.65}U_{0.35}Pd_3$, our results are consistent with the observation that the spin-glass transition temperatures of the NFL $x = 0.35$ and 0.3 compounds are suppressed close to $T = 0$ K [15]. Since NFL behavior has previously been attributed to the proximity of an AF QCP at $T = 0$ K in $CeCu_{5.9}Au_{0.1}$ [20], our results suggest that details of the $T = 0$ K phase transition are unimportant for the NFL behavior.

Theoretically, the NFL behavior may arise from the proximity to a $T = 0$ K spin-glass quantum phase transition, although models in their present forms do not predict the observed ω/T scaling [7–9]. Recent experiments on $Ce(Ru_{0.5}Rh_{0.5})Si_2$ [26] have attributed the NFL behavior to the disorder near a spin-glass QCP [11–13]. On the other hand, disorder was found not to be the main cause for the NFL behavior in quantum spin glasses $UCu_{5-x}Pd_x$ at $x = 1.0$ and 1.5 [27,28]. Assuming that disorder does not play a major role [14,15], one can envision three different microscopic scenarios for the NFL behavior in $(Y, Sc)_{1-x}U_xPd_3$. The first is the QKE [5], where one would expect localized spin excitations with nonmagnetic Γ_3 as the ground state. Inspection of previous data for $Y_{0.8}U_{0.2}Pd_3$ [18,19] as well as Figs. 1–3 for $Sc_{0.65}U_{0.35}Pd_3$ reveals no convincing evidence for localized states. In addition, there is clear magnetic scattering at $E = 0$ meV, and the temperature dependence of magnetic excitations does not follow the expectations of a simple CEF scheme (Figs. 1–3). The second is the $T = 0$ K AF phase transition [2,10]. However, $\chi''(q, \omega, T)$ displays localized moment dynamics with no evidence for U-U correlations [Fig. 1(a)] [29]. Instead, the data are consistent with ω/T scaling analogous to $UCu_{5-x}Pd_x$, and therefore can be understood as manifestations of single-impurity critical scaling associated with a spin-glass phase transition suppressed to near 0 K [30]. The solid line in Fig. 4 shows the theoretically proposed spin susceptibility scaling function $\chi''(q, \omega, T) = 1/[AT^{\alpha}F(\omega/T)]$ with $\alpha = 1/5$ and $F(\omega/T) = \exp[\alpha\Psi(1/2 - i\omega/2\pi T)]$ [10,31]. Although notable deviations with the opposite sign from $UCu_{5-x}Pd_x$ are seen for small ω/T [22], the model accurately describes the data over a remarkable ω/T range (Fig. 4).

In summary, we have used inelastic neutron scattering to show that magnetic excitations in the NFL $Sc_{1-x}U_xPd_3$ ($x = 0.35$) compound are broad and featureless in wave vector and energy. The absence of any characteristic energy scale, other than the temperature itself, suggests that the microscopic origin of the NFL behavior lies with individual U ions near a $T = 0$ K spin-glass phase transition. Therefore, the NFL properties in a wide variety of f-electron systems including $(Y, Sc)_{1-x}U_xPd_3$, $UCu_{5-x}Pd_x$, $CeCu_{6-x}Au_x$, and $Ce(Rh_{0.8}Pd_{0.2})Sb$ can be described by a common physical picture, being near a $T = 0$ K quantum phase transition. Although the intrinsic disorder in these systems is essential for establishing the spin-glass ground state [11–13], it cannot be the main cause of the NFL behavior.

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