Magnetic Correlations and the Quantum Critical Point of $UCu_{5-x}Pd_x (x = 1, 1.5)$

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(Received 7 May 2001; published 23 October 2001)

We have used inelastic neutron scattering to determine the magnetic susceptibility $\chi(q, \omega, T)$ of the non-Fermi-liquid compounds $UCu_{5-x}Pd_x (x = 1, 1.5)$ for energies ω between 0.2 and 2 meV, and for temperatures T between 1.6 and 250 K. Spatial correlations in both UCu_4Pd and $UCu_{3.5}Pd_{1.5}$ extend over length scales comparable to the unit cell, and display very little temperature dependence. In contrast, the wave vector independent susceptibility diverges as $T \rightarrow 0$. We find that the excitations at all q, and for all T and ω accessed display the same type of non-Fermi-liquid ω/T scaling.

DOI: 10.1103/PhysRevLett.87.197205 PACS numbers: 75.40.Gb, 71.10.Hf, 75.30.Mb

It is well known [1] that magnetic order in heavy fermions can be suppressed by pressure and composition, giving way to strongly correlated metals with Fermi liquid ground states [2]. In the past few years, a novel metallic state with non-Fermi-liquid (nFL) scaling properties has been discovered in a number of such systems near the zero temperature magnetic instability which separates the paramagnetic and magnetically ordered phases. Here unusually weak power law and logarithmic temperature divergences are found in the specific heat, electrical resistivity, and magnetic susceptibility, implying excitations with unusual energy and temperature dependences [3]. Inelastic neutron scattering measurements have directly probed these excitations, finding the underlying imaginary part of the dynamical susceptibility χ'' to be a function of excitation energy ω scaled by temperature T [4–7].

Despite the generic nature of these observations, there is increasing evidence that there are two broad classes of heavy fermion nFL hosts. The first is exemplified by $CeCu_{6-x}Au_x$, where the magnetic susceptibility $\chi(q, \omega, T)$ is strongly enhanced at the propagation wave vectors q^* of the magnetic structure in the parent antiferromagnet [5–7]. Further, $\chi(q^*)$ increases strongly as the temperature approaches zero, suggesting that it may be possible to define a spatial correlation length much as for a conventional second order phase transition [8].

$UCu_{5-x}Pd_x$ $(x = 1, 1.5)$ is the best characterized example of the second type of nFL system. The specific heat divided by temperature and the static susceptibility weakly diverge with decreasing temperature, as is found in all nFL systems [9]. Inelastic neutron scattering experiments [4] find that χ is independent of the wave vector, while displaying the ω/T scaling which is the hallmark of nFL excitations: $\chi''(\omega, T) = T^{-1/3} Z(\omega/T)$. The scaling properties of these excitations have been shown to be consistent with the observed temperature dependences in the static susceptibility and the electrical resistivity [9–11], at least for temperatures comparable to the excitation energies probed in this experiment [4]. Both nuclear magnetic resonance (NMR) [12] and muon spin rotation (μSR) [13] experiments found that the quasistatic correlations at temperatures where nFL phenomena are observed extend over length scales comparable to the unit cell, results which were thought to be consistent with the degree of Pd-Cu site disorder revealed in extended x-ray absorption fine structure experiments [14] and structural refinements using neutron diffraction [15]. Similar experiments found much lower levels of chemical disorder in $CeCu_{6-x}Au_x (x \simeq x_c)$ [16], leading to proposals that non-Fermi liquid behavior may have different origins in systems with large and small amounts of disorder [12,17,18].

The absence of critical spatial correlations in $UCu_{5-x}Pd_x$ is a strong indication that the origins of non-Fermi liquid behavior in $UCu_{5-x}Pd_x$ and $CeCu_{6-x}Au_x$ are different. So far, experiments carried out on $UCu_{5-x}Pd_x$ have not found any wave vector modulation of $\chi(q, \omega, T)$. It is still unclear whether this is because disorder limits the growth of spatial correlations or because experiments have not yet been carried out at sufficiently low energies and temperatures to access the free development of critical spatial correlations. The goal of the inelastic neutron scattering experiment reported here is to explore these two possibilities by extending our previous experiments on $UCu_{5-x}Pd_x$ to lower excitation energies and temperatures. Our primary result is that long range correlations never develop on the range of temperatures for which non-Fermi liquid phenomena are observed, and—surprisingly—that it is the local susceptibility which is critical in $UCu_{5-x}Pd_x (x = 1, 1.5)$. The previously observed ω/T scaling is found to be robust at the lowest energies and temperatures.

The results reported here were obtained on the IN6 time of flight spectrometer at the Institut Laue Langevin, using a neutron wavelength of 5.1 Å. The samples were slabs, cut from the arc melted, polycrystalline samples used in our previous inelastic neutron scattering experiment [4].

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The sample dimensions were selected to minimize multiple scattering, and to allow accurate absorption corrections. The scattering is expressed in absolute units through normalization to a vanadium standard and has been corrected for the uranium ion magnetic form factor. It is not possible to subtract the nonmagnetic scattering, since for the energies of interest we do not access sufficiently large wave vectors at this neutron wavelength. However, Monte Carlo calculations indicate this is a small part of the total scattering at the energies, wave vectors, and temperatures of the current experiment.

The most dramatic result of this experiment is the identification of a distinct wave vector dependence of the scattered intensity S, which was not detected in our initial experiment at higher energies and wave vectors [4]. A contour plot of $\chi''(q,\omega,T) = S(q,\omega,T)[1 - \exp(-\hbar\omega/k_BT)]$ in UCu_4Pd at 1.6 K is presented in Fig. 1, with neutron energy transfers ω ranging from -0.5 to 1.75 meV, and wave vector magnitudes q as large as 2 Å^{-1}. The most prominent feature of $\chi''(\omega,q)$ is the broad ridge of scattering centered in both UCu_4Pd and $UCu_{3.5}Pd_{1.5}$ at $q_{\max} = 0.9$ Å$^{-1} \simeq \frac{1}{2}(1,1,1)$. In order to study these correlations quantitatively, we have obtained $\chi'(q)$ from these data by means of the Kramers-Kronig relation. $\chi'(q,T)$ is plotted for fixed temperatures from 1.6–250 K for UCu_4Pd and for $UCu_{3.5}Pd_{1.5}$ at 1.7 K in Fig. 2a.

We have obtained a satisfactory description of $\chi'(q,T)$ at each temperature using a liquidlike structure factor similar to those used in systems like frustrated magnets [19]

$$\chi'(q,T) = \chi'_{\mathrm{loc}}(T)\left[1 + \sum_{i=1}^{4} a_i(T)\sin(qr_i)/qr_i\right], \quad (1)$$

which expresses short range correlations between a spin at the origin and other spins at distances r_i within the unit cell [20]. The best fits obtained from this model are compared to the $\chi'(q,T)$ data in Fig. 1. The suppression of scattering at small q is typical of liquid structures, and is well reproduced by the fit. The accurate position of the peak at $q_{\max}$ is mainly determined by the nearest neighbor

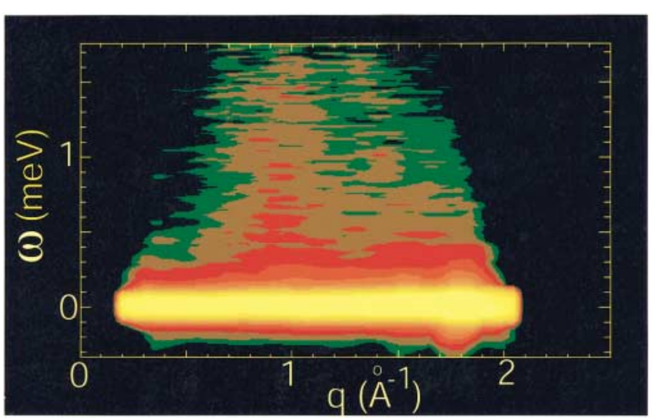

FIG. 1 (color). A contour plot of $\chi''(q,\omega)$ of UCu_4Pd at 1.6 K.

($i = 1$) term, and depends only on known moment separations r_i which are input to the fit. Figure 2a indicates that there is little difference between the low temperature correlations in UCu_4Pd and $UCu_{3.5}Pd_{1.5}$, indicating that the short ranged correlations are insensitive to the much larger degree of disorder present in $UCu_{3.5}Pd_{1.5}$ [15]. In both compositions, the maximum in $\chi'(q)$ occurs near the magnetic zone center $q \simeq |\frac{1}{2}(1,1,1)|$, as in the parent antiferromagnet UCu_5 [21]. Finally, $\chi'(q)$ approaches a constant value at large q, $\chi'_{\mathrm{loc}}(T)$, which represents the local susceptibility.

Despite the rapid suppression of $\chi'(q)$ with increased temperature depicted in Fig. 2a, the range of the spatial correlations themselves changes very little with temperature. Instead, the temperature dependence of the uniform susceptibility χ'_{loc} dominates the overall temperature dependence of $\chi'(q,T)$, as shown in Fig. 2b. Here we have plotted $\chi'(q,T)$ of UCu_4Pd normalized by $\chi'_{\mathrm{loc}}(T)$ at 1.6 and 250 K. We observe a modest suppression or broadening of the peak at $q_{\max}$ as the temperature is raised to 250 K, but overall the temperature dependence of the summed spin pair correlations $\chi'(q,T)/\chi'_{\mathrm{loc}}(T) = [1 + \sum a_i(T)\sin(qr_i)/qr_i]$ is surprisingly weak. The inset of Fig. 2b shows that $\int_{0.2A^{-1}}^{2A^{-1}} dq\, \chi'(q,T)$ is almost constant for 1.6 K $\leq T \leq$ 250 K, indicating that virtually none of the response moves out of the energy and wave vector windows of the experiment as the temperature is reduced from 250 to 1.6 K.

The temperature dependences of the parameters deduced from the liquid structure fits are presented in Fig. 3. As shown in Fig. 3a and its inset, the local susceptibility $\chi'_{\mathrm{loc}}(T)$ diverges with lowered temperature as $T^{-0.67\pm0.09}$.

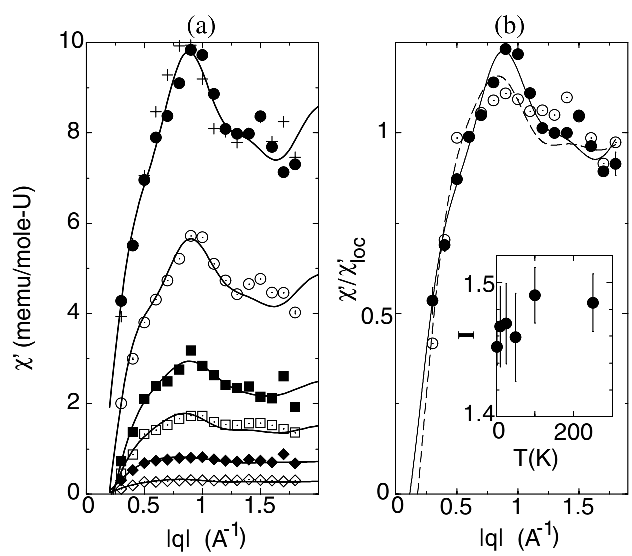

FIG. 2. (a) $\chi'(q)$ of UCu_4Pd at 1.6 K (•), 10 K (○), 25 K (■), 50 K (□), 100 K (◆), and 250 K (◇). $\chi'(q)$ of $UCu_{3.5}Pd_{1.5}$ at 1.7 K (+) is included for comparison. As described in text, solid lines are fits to a liquid structure factor [Eq. (1)]. (b) $\chi'(q)$, normalized by the wave vector independent $\chi'_{\mathrm{loc}}(T)$, at 1.6 K (•) and 250 K (○). Inset: The integral I of $\chi'(q)$ over wave vectors is nearly temperature independent.

The temperature dependences of the four spin-spin expectation values $a_i = \langle S_0 \cdot S_i \rangle$ $(i = 1, 4)$ are plotted in Fig. 3b. For $T \leq 10$ K, all of the spin correlations except a_3 are negative or antiferromagnetic. The low temperature magnetic structure is shown in the inset of Fig. 3b, with the first and fourth neighbor spins, located at the face centered positions, antiparallel to the spins on the cube corners. This is the same magnetic cell found in UCu_5 [22], and in the absence of longer range interactions leads to magnetic frustration. In UCu_4Pd, increasing the temperature changes the sign of a_3 and replaces frustration with increasingly short ranged correlations. Ultimately, all the a_i converge to small and temperature independent values above ~100 K. The a_i are all negative at every temperature in $UCu_{3.5}Pd_{1.5}$.

The fundamental property of excitations with non-Fermi liquid character is their lack of a characteristic energy scale, other than temperature itself. Our results show that this is true for excitations in UCu_4Pd which have very different wave vectors. We begin by considering excitations with wave vectors in the range 1.2–1.4 Å^{-1}, where Fig. 2a shows $\chi'(q)$ is largely independent of wave vector. Figure 4a shows that here $\chi''(q, \omega, T)T^{1/3}$, obtained at temperatures between 1.6 and 250 K, can be expressed as a single function of ω/T. Similar data, obtained previously [4] using the HET spectrometer with excitation energies between 2 and 25 meV, are also plotted in Fig. 4a, displaying identical scaling properties. Only at the highest temperatures (250 K for the IN6 experiment and 300 K for the HET experiment) do we observe any difference between the two data sets. This may simply be the result of oversubtraction of the nonmagnetic scattering in the HET experiment, but it is also possible that there is a high temperature cutoff for wave vector independent nFL scaling.

The same type of ω/T scaling is found for excitations at every wave vector. We have selected three different but representative wave vector intervals—0.5 ± 0.05, 0.9 ± 0.05, and 1.2 ± 0.05 Å^{-1}—a range of wave vectors over which there is a substantial modulation of χ'. Figure 4b shows that on each interval $\chi''(\omega, T)T^{1/3} = \mathcal{Z}(\omega/T)$, which is the same scaling behavior described in Fig. 4a. In the top panel of Fig. 4b, we demonstrate that the scaled data at each q collapse onto a single curve if we divide $\chi''(\omega, T)T^{1/3}$ through by a q-dependent scaling factor with the same qualitative features as $\chi'(q)$. The success of this procedure indicates that the wave vector and energy dependences of $\chi(q, \omega, T)$ are separable:

$$\chi''(q, \omega, T) = [T^{-1/3}\mathcal{Z}(\omega/T)] \times \left[\sum_{i=1}^{4}(1 + a_i(T)\sin qr_i/qr_i)\right]. \quad (2)$$

In short, we find that the nFL dynamics in UCu_4Pd are the same at all wave vectors [23]. This is in complete contrast to the case of $CeCu_{6-x}Au_x$ $(x = x_c = 0.1)$ [5,7], where the ω/T scaling occurs for only certain wave vectors.

This experiment shows that the ω/T scaling found in the earlier HET experiment [4] is present on an expanded range of temperatures between 1.6 and 300 K, and for all energies between 0.2 and 25 meV. In particular, there is no indication of a crossover to the spatially critical dynamics expected in the vicinity of a second order phase transition

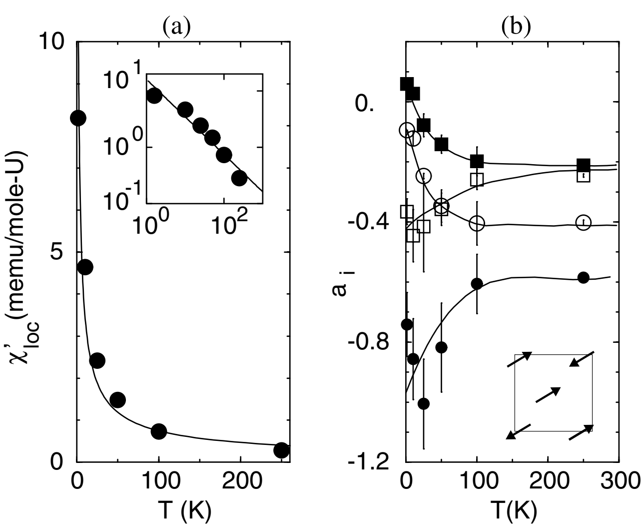

FIG. 3. (a) The temperature dependence of χ'_{loc} in UCu_4Pd. Solid line is $\chi'_{loc}(T) \propto T^{-0.67\pm0.09}$. Inset: same quantities in a double log plot. (b) Temperature dependences of parameters a_i of the liquid structure factor model for UCu_4Pd. The first four shell parameters are represented by the filled circles $(i = 1)$, open circles $(i = 2)$, open squares $(i = 3)$, and filled squares $(i = 4)$. Solid lines are guides for the eye. Inset: The deduced low temperature magnetic structure of UCu_4Pd.

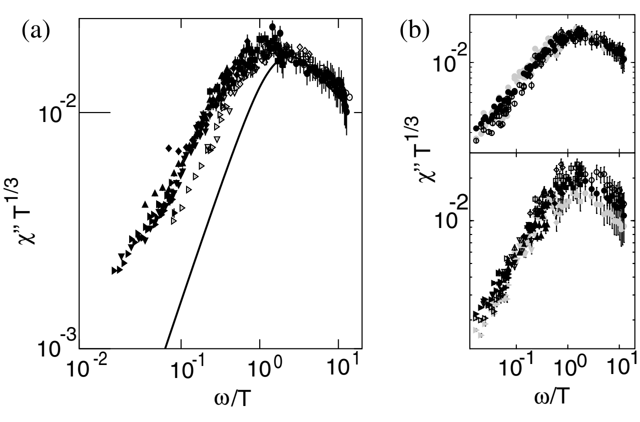

FIG. 4. (a) $\chi''(q, \omega, T)\,T^{1/3}$ in UCu_4Pd scales with ω/T for 1.2 Å$^{-1} \leq q \leq$ 1.4 Å^{-1}. Units for χ'' are emu/mol-U. Filled symbols are the current experiment ($\omega \leq 2$ meV); open symbols are from a previous [4] experiment (2 meV $\leq \omega \leq$ 25 meV). IN6: (•) 1.6 K, (■) 10 K, (◆) 25 K, (▲) 50 K, (▼) 100 K, (▶) 250 K. HET: (○) 12 K, (□) 25 K, (◇) 50 K, (▽) 100 K, (▷) 300 K. Solid line is a generic impurity model [11] with $\Delta = 1/3$. (b) Lower panel: Same quantities for $q = 0.5 \pm 0.05$ Å^{-1} (grey filled circles), $q = 0.9 \pm 0.05$ Å^{-1} (open circles), and $q = 1.2 \pm 0.05$ Å^{-1} (black filled circles). In the upper panel, these data have been normalized to the $q = 0.9 \pm 0.05$ Å^{-1} data.

in a system like $CeCu_{6-x}Au_x$ ($x = x_c = 0.1$). We have previously suggested that the unusual dynamics of U moments in UCu_4Pd and $UCu_{3.5}Pd_{1.5}$ were consistent with a generic type of moment compensation mechanism, characterized by a power-law time dependence of the local spin correlation function [11]. The expanded range of ω/T of the current experiment shows (Fig. 4a) that the agreement between the theoretical expression and the data is increasingly poor for $\omega/T \ll 1$.

Our work provides direct experimental evidence for short ranged magnetic correlations near the $x = 1, T = 0$ quantum critical point in $UCu_{5-x}Pd_x$. The similarity of results in UCu_4Pd and $UCu_{3.5}Pd_{1.5}$ suggests that the critical regime is extended. Our neutron scattering experiments agree with the general conclusions of NMR [12] and μSR experiments [13], which found that magnetic correlations extend over length scales comparable to the unit cell. Further, we have demonstrated that the magnetic structure of the parent antiferromagnet UCu_5 is preserved in UCu_4Pd only within the unit cell, and that excitations involving these small groups of correlated spins are largely uncoupled from those of neighboring cells over the temperature range of our experiments.

We believe that there are three attributes of $UCu_{5-x}Pd_x$ which together limit the growth of magnetic correlations in the nFL compositions UCu_4Pd and $UCu_{3.5}Pd_{1.5}$. Compositional or structural disorder cannot be the only factor which determines the magnetic texture: UCu_4Pd has a substantial degree of Pd-Cu site disorder, but more is found at other compositions [15]. Frustration is a natural attribute of the magnetic unit cells of both UCu_5 and UCu_4Pd, unique among known nFL systems. Finally, the local character of the magnetic excitations is consistent with the breakdown of coupling among moments, and may be considered a harbinger of the massive Fermi liquid state which appears at the $T = 0, x = 1$ critical point [7]. This localization may be especially pronounced in $UCu_{5-x}Pd_x$, since it is thought on chemical grounds that substitution of Cu by Pd reduces the conduction electron density of states [24], potentially enhancing the strength of many body correlations.

Our experiments reveal $UCu_{5-x}Pd_x(x = 1, 1.5)$ to be systems in which spatially extended magnetic interactions are unimportant for the singular dynamics. We find that the local magnetic susceptibility diverges with lowered temperature, while nascent magnetic correlations remain confined to the unit cell at all temperatures. ω/T scaling of $\chi''(q, \omega, T)T^{1/3}$ has been observed to the lowest energies and temperatures. The scaling function is the same for a wide range of wave vectors, indicating that the energy and wave vector dependences of χ'' are uncoupled. None of these observations can be accommodated within the phenomenological framework of magnetic quantum phase transitions which has been successful in describing $\chi(q, \omega, T)$ in $CeCu_{6-x}Au_x$ [7]. This comparison lends new strength to the possibility that there are different classes of quantum critical behaviors, even among $T = 0$ antiferromagnets, making it all the more urgent to extend these types of neutron scattering studies to new systems.

We are grateful to A. M. Tsvelik, P. Coleman, A. Castro Neto, J. Mydosh, B. A. Jones, C. M. Varma, and D. E. MacLaughlin for stimulating discussions. Work at the University of Michigan was supported by the National Science Foundation under Grant No. DMR-99-77300. Research at UCSD was supported by the National Science Foundation under Grant No. DMR-97-0545. Work at Argonne was carried out under the auspices of the U.S. DOE.

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