Non-Fermi-Liquid Scaling of the Magnetic Response in $UCu_{5-x}Pd_x$ ($x = 1, 1.5$)

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We have determined the magnetic response $S(\omega)$ of $UCu_{5-x}Pd_x$ ($x = 1, 1.5$) for temperatures T ranging from 12 to 300 K and energy transfers ω from 0.5 to 200 meV. $S(\omega)$ is virtually identical in the two compounds, displaying localized moment dynamics for $\omega \gtrsim 25$ meV. For $\omega < 25$ meV, temperature provides the only energy scale for the magnetic excitations with the dynamical susceptibility described by a universal scaling function $\chi''(\omega, T) \sim \omega^{-1/3} Z(\omega/T)$. We argue that $S(\omega)$ represents critical scattering associated with a $T = 0$ phase transition, whose origin lies with the magnetic screening of individual uranium ions.

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Fermi liquid theory (FLT) is fundamental to our understanding of electronic excitations in metals, as it establishes a one-to-one correspondence between the excitations of a free electron gas and those of interacting conduction electrons in metals. One of the central premises of FLT is the existence of a single energy scale, the Fermi temperature E_F, and for energies $E \ll E_F$ and temperatures $k_B T \ll E_F$ the electronic properties display universal behavior. Perhaps in no other solid system have the predictions of FLT been more strikingly realized than in the heavy-fermion intermetallics. In a number of these f-electron-based compounds, analyses of the low-temperature specific heat, magnetization, electrical resistivity, and dynamical susceptibility [1,2] display a dependence on a single energy scale consistent with FLT, although unprecedented mass enhancements of 10^2–10^3 imply strong electronic interactions. However, a new class of compositionally related compounds, typified by $UCu_{5-x}Pd_x$, has recently been identified that exhibits [3,4] low-temperature properties inconsistent with FLT. Weak power law and logarithmic divergences in the low-temperature properties of these materials suggest the presence of a zero-temperature phase transition, whose origin is thought to lie either with an unconventional moment screening mechanism as in $U_{0.2}Y_{0.8}Pd_3$ [5] or $U_{0.01}Th_{0.99}Ru_2Si_2$ [6], or with long-range magnetic or spin-glass order suppressed to $T = 0$ as in $CeCu_{5.9}Au_{0.1}$ [7] or $UCu_{3.5}Pd_{1.5}$ [4]. The results of inelastic scattering experiments presented here reveal a qualitatively new scale-invariant spectrum of magnetic excitations for this class of materials, in which temperature replaces a fixed E_F in dictating the electronic properties.

We focus here on one member of this class, $UCu_{5-x}Pd_x$. For $x \leq 2.5$, this intermetallic system crystallizes in the $AuBe_5$ structure, characterized by a periodic fcc uranium lattice with two inequivalent copper sites [8]. The parent compound UCu_5 is a prototypical Kondo lattice antiferromagnet with a Néel temperature $T_N = 15$ K [9] and a Kondo temperature of ~80 K, inferred from the quasielastic linewidth observed in inelastic neutron scattering experiments [10]. Upon substitution of Pd for Cu, the antiferromagnetic order is quickly suppressed, vanishing for Pd concentration x between 0.5 and 1 [4,8], and a spin-glass regime is observed for $x \geq 2$. Compounds with intermediate values $x = 1, 1.5$ display no long-range magnetic order of any kind at the lowest temperatures, while low-temperature thermal and transport measurements exhibit remarkable temperature T and magnetic field H scaling properties [3,4]. These properties are manifestly non-Fermi-liquid-like, with the electrical resistivity $\rho(T) \sim a - bT$, the static magnetic susceptibility $\chi_0 \sim T^{-1/3}$, and the specific heat $C \sim T \log T$. These low-temperature divergences suggest the existence of unusual magnetic excitations, whose scaling properties are distinct from those of the more familiar Fermi liquid which has $\rho(T) \sim T^2$, $\chi_0 \sim$ const, and $C \sim \gamma T$. It is the object of the neutron scattering experiments reported here to identify these excitations and to describe their underlying frequency and temperature scaling properties.

Neutron time-of-flight measurements were used to determine the inelastic scattering intensity of arc-melted, polycrystalline buttons of UCu_4Pd and $UCu_{3.5}Pd_{1.5}$, as well as the nonmagnetic isostructural analog compound $ZrCu_5$. We have used the HET spectrometer at the ISIS Pulsed Neutron Facility for energy transfers from ~3 to 400 meV, while the medium resolution time-of-flight spectrometer at the National Institute of Standards and Technology [11] was used to extend the energy window down to ~0.5 meV. In every case, vanadium normalization was used to provide an absolute intensity calibration. The magnetic part of the response $S(\omega)$ was obtained from the measured scattering by subtracting

single and multiple phonon contributions obtained from Monte Carlo calculations [12].

A comparison of $S(\omega)$ at 12 K for UCu_4Pd and $UCu_{3.5}Pd_{1.5}$ is presented in Fig. 1. As demonstrated in the inset of Fig. 1, $S(\omega)$ in UCu_4Pd has no appreciable dependence on the wave-vector magnitude other than that of the U^{3+} and U^{4+} magnetic form factor [13]. Consequently $S(\omega)$ has been summed over experimental wave vectors. $S(\omega)$ is virtually identical for the two compounds, broad and quasielastic with no evidence for distinct crystal field levels. $S(\omega)$ cannot be described by a single Lorentzian over the complete range of energy transfers, instead displaying qualitatively different behavior above and below $\omega^* \simeq 25$ meV. For $\omega \geq \omega^*$, $S(\omega) \sim \omega^{-1}$, which we hypothesize is the high-energy tail of a Lorentzian response similar to that found in the parent compound UCu_5 [10]. The characteristic linewidth, taken to be the single uranium ion Kondo temperature, is found to be 8.8 meV, similar to the value of 8 meV found in UCu_5. However, the proposed Lorentzian response is cut off at a characteristic energy $\omega^* \sim 25$ meV, and at smaller energy transfers we find a weak power law divergence $S(\omega) \sim \omega^{1/3}$.

The most striking property of $S(\omega)$ is its temperature dependence, demonstrated for UCu_4Pd in Fig. 2. Similar results were obtained for $UCu_{3.5}Pd_{1.5}$. While the detailed balance requirement results in an accumulation of scattering intensity on the energy gain ($\omega < 0$) side of the elastic line with increasing temperature, $S(\omega)$ is *completely temperature independent* for neutron energy loss. This remarkable result requires that the underlying dynamical susceptibility $\chi(\omega, T)$ have a very special form. $S(\omega)$ and the imaginary part of the dynamical susceptibility $\chi''(\omega, T)$ are related by

$$S(\omega) = [n(\omega) + 1]\chi''(\omega, T), \tag{1}$$

where $n(\omega) + 1$ is the thermal occupation factor. We infer from the temperature independence of $S(\omega)$ that

$$T \gg \omega, \quad [n(\omega) + 1] \sim T/\omega, \quad \chi''(\omega, T) \sim (\omega/T)\mathcal{G}(\omega), \tag{2}$$

$$T \ll \omega, \quad [n(\omega) + 1] \sim 1, \quad \chi''(\omega, T) \sim \mathcal{G}(\omega), \tag{3}$$

where $\mathcal{G}(\omega) \sim \omega^{-1} (\omega \geq \omega^*)$ and $\mathcal{G}(\omega) \sim \omega^{-1/3} (\omega \leq \omega^*)$. Like the marginal Fermi liquid which has $\mathcal{G}(\omega)$ constant [14,15], the excitations in UCu_4Pd and $UCu_{3.5}Pd_{1.5}$ are distinguished by their scale-invariant properties, with the nature of the response for $\omega < \omega^*$ depending entirely on whether the energy transfer is larger or smaller than the experimental temperature. Since the characteristic energy scale for the excitations represented in this response is not constant, as for a Fermi liquid, but instead variable temperature, we refer to this generic type of scaling as non-Fermi-liquid-like.

This observation is detailed in Fig. 3, in which the energy dependence of $\chi''(\omega, T)$ is presented for temperatures ranging from 12 to 300 K. As schematically indicated in the inset, there are three qualitatively different regimes of behavior. As previously noted, a Lorentzian regime (I) is observed at every temperature for energy transfers greater than ω^*, where ω^* increases slightly with temperature. For energy transfers less than ω^*, the form of the response depends on whether the energy transfer is greater or less than the experimental temperature. For $\omega \ll T$, $\chi''(\omega, T) \sim \omega/T^{\eta}$, with $\eta = 1 \pm 0.3$ (region II). With increasing temperature, a crossover is observed at $\omega \sim T$ to the temperature independent form $\chi''(\omega, T) \sim \omega^{-1/3}$ (region III). Note that the width of the intermediate region III is narrow at high temperature, resulting here in a smooth crossover from region II to region I with increasing energy transfer.

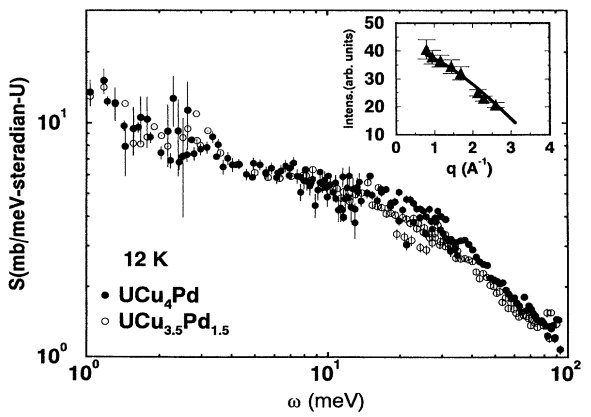

FIG. 1. The magnetic scattering intensities $S(\omega)$ for UCu_4Pd (filled circles) and $UCu_{3.5}Pd_{1.5}$ (open circles) at 12 K. Inset: $S(\omega)$ integrated over ω, as a function of wave-vector magnitude q. Solid line is the magnetic form factor for U^{3+}/U^{4+} ions [13].

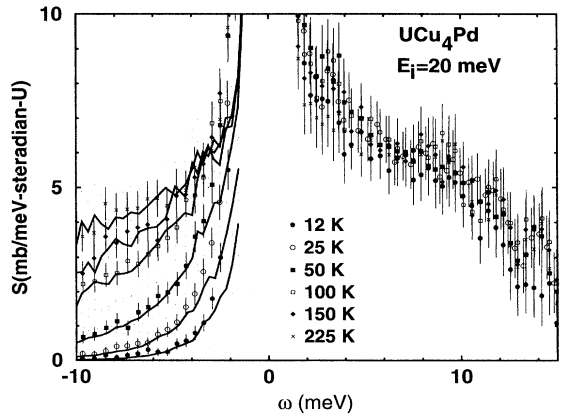

FIG. 2. $S(\omega)$ of UCu_4Pd at fixed temperatures ranging from 12 to 225 K. The incident energy is 20 meV. Solid lines for energy gain are calculated from the energy loss part of the neutron spectrum by $S(\omega > 0)\exp(-\omega/T) = S(\omega < 0)$, demonstrating that the magnetic scattering obeys detailed balance.

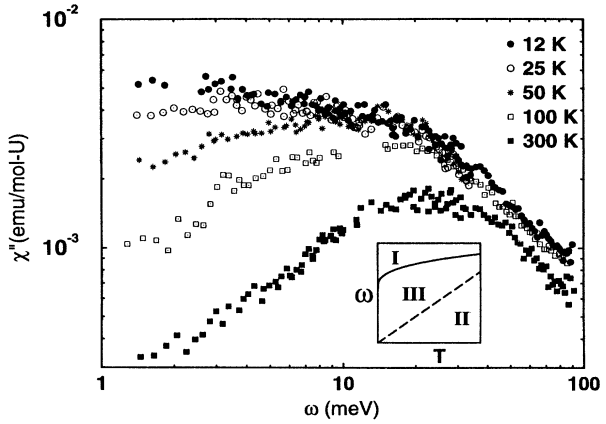

FIG. 3. $\chi''(\omega)$ for UCu_4Pd at constant temperatures ranging from 12 to 300 K. Inset: three qualitatively different regimes are observed: I ($\omega \geq \omega^*$): $\chi''(\omega,T) \sim (\omega/T_K)/[1 + (\omega/T_K)^2]$, $T_K \sim 8$ meV II ($\omega \ll T$): $\chi''(\omega,T) \sim \omega/T^{1\pm1/3}$ III ($\omega \gg T$): $\chi''(\omega,T) \sim \omega^{-1/3}$. Solid line: $\omega^*(T)$, dashed line: $\omega = T$.

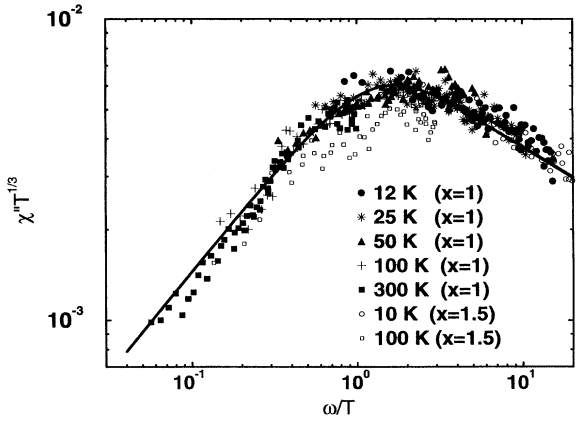

FIG. 4. $\chi''(\omega,T)T^{1/3}$ has almost identical universal scaling properties for both compounds $UCu_{5-x}Pd_x$ ($x = 1, 1.5$). Data with energy transfers $\omega \geq 25$ meV are not included in this plot. Solid line: $\chi''(\omega,T)T^{1/3} \sim (T/\omega)^{1/3}\tanh(\omega/1.2T)$.

We have found a universal scaling function which relates the limiting behaviors for large and small ω/T outlined in Fig. 3. As demonstrated in Fig. 4, the data in regions II and III for both UCu_4Pd and $UCu_{3.5}Pd_{1.5}$ collapse onto a single curve when plotted as functions of ω/T, with a weakly divergent prefactor $T^{-1/3}$. That is, the dynamical susceptibility can be written very generally as

$$\chi''(\omega,T)T^{1/3} \sim (T/\omega)^{1/3}\mathcal{Z}(\omega/T), \qquad (4)$$

a scaling relation which indicates that $S(\omega)$ represents critical scattering. A Kramers-Kronig analysis of $\chi''(\omega,T)$ reveals that the high-energy excitations with $\omega > \omega^*$ provide a small and almost temperature-independent contribution to the static susceptibility $\chi(T)$ for $T \leq 300$ K, while the excitations described by scaling relation (4) are wholly responsible for the weak power law divergence $\chi(T) \sim T^{-1/3}$ (85 K $\lesssim T \lesssim$ 300 K) [16]. The lack of a finite transition temperature in Eq. (4), as well as the diverging low-temperature static susceptibility $\chi(T) \sim T^{-1/3}$ argues that the phase transition associated with this critical scaling occurs at $T = 0$. Our results suggest that the existence of long-range, long-lived excitations associated with this quantum phase transition leads to the breakdown of Fermi-liquid behavior in UCu_4Pd and $UCu_{3.5}Pd_{1.5}$. However, further neutron scattering studies at smaller energy transfers are required to allow a detailed comparison to the scaling exponents deduced from the magnetization and specific heat measurements for $T \leq 20$ K [17].

While the temperature independence of $S(\omega)$ suggests [Eq. (1)] that $\mathcal{Z} \sim 1/[n(\omega) + 1]$, systematic deviations from the data for both UCu_4Pd and $UCu_{3.5}Pd_{1.5}$ are found for this $\mathcal{Z}$ at small ω/T. A more successful scaling function is demonstrated in Fig. 4 with $\mathcal{Z} \sim \tanh(\omega/1.2T)$. This choice was prompted by previous experimental findings in the normal state of the high-T_c copper oxides [15], but we stress that the overall responses are not identical and do not necessarily imply similar microscopic physics in the two systems.

Our major result is that we have observed a qualitatively new type of magnetic response for an f-electron based system. Our measurements are consistent with an ordinary localized moment response for $\omega \simeq 25$ meV, while at lower energies

$$\chi'' \sim \omega^{-1/3}\mathcal{Z}(\omega/T), \qquad (5)$$

a scale-invariant form reminiscent of that found in some metallic copper oxide compounds [15]. The lack of any characteristic energy scale for the response, beyond that of temperature itself, provides a dramatic distinction from either the Lindhard-like $\chi'' \sim \omega/T_K^2$ or localized moment Lorentzian $\chi'' \sim (\omega/T_K)/(1 + \omega^2/T_K^2)$ [10,18] previously observed in f-electron systems, as both are characterized by a single energy scale, the Kondo temperature T_K. In particular, our observations rule out single channel Kondo impurity or normal metallic Kondo lattice models as the underlying Hamiltonian, since both are known to have Fermi-liquid ground states [19,20].

Although there is both experimental [7] and theoretical [21] evidence for non-Fermi-liquid scaling near both magnetic ordering and spin-glass transitions, the observation of a similar response in both UCu_4Pd and $UCu_{3.5}Pd_{1.5}$ compounds known from the $UCu_{5-x}Pd_x$ phase diagram to have differing stabilities to anitferromagnetic or spin-glass order, supports the view that the dynamic scale invariance is instead a property of isolated uranium ions. The lack of wave-vector dependence on the response is consistent with this conclusion, although we caution that independent of the interaction range, even ordinary mean field critical scattering [22] becomes largely wave-vector independent for wave vectors large compared to $2k_F$, a situation realized for the 0.5 to $\sim 4k_F$ wave-vector range probed in our experiment.

Several theoretical proposals exist for f-electron systems which feature non-Fermi-liquid ground states. Cox [23] has proposed that tetravalent uranium ions in a cubic crystal field experience the electrical equivalent of the magnetic Kondo effect, the quadrupolar Kondo effect. However, this model requires a nonmagnetic doublet ground state, which is ruled out in UCu_4Pd and $UCu_{3.5}Pd_{1.5}$ by our observation of magnetic quasielastic scattering and the absence of distinct crystal field levels. The related magnetic multichannel Kondo effect [24–26] remains a possible explanation for our results. However, stringent conditions must be satisfied by the channel degeneracy and coupling strengths to realize a non-Fermi-liquid ground state. What is more, no models have yet been presented for either trivalent or tetravalent magnetic ground states of uranium ions in cubic crystal field which would motivate a description for UCu_4Pd and $UCu_{3.5}Pd_{1.5}$ in terms of the multichannel Kondo effect.

To conclude, we have identified a novel magnetic response in both UCu_4Pd and $UCu_{3.5}Pd_{1.5}$ whose scale invariance suggests an origin in critical scattering associated with a zero-temperature phase transition. While the origin of this quantum phase transition remains obscure at present, our results suggest a novel dynamics for isolated uranium ions, perhaps consistent with a multichannel Kondo effect.

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