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1996 J. Phys.: Condens. Matter 8 9815

(http://iopscience.iop.org/0953-8984/8/48/011)

J. Phys.: Condens. Matter **8** (1996) 9815–9823. Printed in the UK

Non-Fermi-liquid scaling in $UCu_{5-x}Pd_x$ ($x = 1$, 1.5)

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Received 6 August 1996

Abstract. We review the compiled measurements of the imaginary part of the dynamical magnetic susceptibility $\chi''(\omega, T)$, static susceptibility $\chi(T)$, electrical resistivity $\rho(T)$ and specific heat $C(T)$ in the uranium intermetallics $UCu_{5-x}Pd_x$ ($x = 1$, 1.5). We assess the temperature- and energy-dependences predicted by single-ion and disorder-dominated models and compare these results to experiments. For temperatures T and excitation energies ω in the range 12 K $< \omega$, $T <$ 150 K, our analysis suggests that the dynamics of isolated uranium ions are responsible for the observed temperature and frequency scaling, although inter-ion interactions may become important at lower temperatures and frequencies.

Fermi liquid theory has long been the paradigm for our phenomenological understanding of interacting metals, linking temperature- and energy-dependence of experimental quantities to the intrinsic energy scale of the electronic excitations. However, the discovery of qualitatively different 'non-Fermi liquid' behaviour in a number of different classes of non-ordering metals suggests that the scaling properties of electronic excitations are more varied than originally thought. The experimental task required to broaden the phenomenology of metals to include non-Fermi liquids is to identify the intrinsic scale for the fundamental excitations. This is accomplished on a case by case basis by assembling a compendium of experimental results on the temperature- and energy-dependences of quantities such as the dynamic and static susceptibilities, specific heat and electrical resistivity. If these temperature- and energy-dependences are consistent with a constant energy scale for the excitations, the material is a Fermi liquid, while a variable energy scale leads to 'non-Fermi liquid' behaviour. Among the heavy-fermion-derived materials proposed to have this non-Fermi liquid behaviour, the system $UCu_{5-x}Pd_x$ ($1 \leqslant x \leqslant 1.5$) has one of the most complete data sets [1–5]. The purpose of this paper is to review these experimental results, assess their internal consistency and compare several theoretical views which have been proposed to provide the microscopic mechanism for the non-Fermi liquid phenomenology.

The system $UCu_{5-x}Pd_x$ is isostructural, forming in the face-centred cubic $AuBe_5$ structure [6]. Pd doping rapidly suppresses the antiferromagnetic order of the parent compound UCu_5 and no order is observed in UCu_4Pd or $UCu_{3.5}Pd_{1.5}$ down to very low temperatures. Spin glass order is found for the compounds $UCu_{5-x}Pd_x$ ($x \geqslant 2$) [1, 2]. Much interest has focused on the intermediate compounds UCu_4Pd and $UCu_{3.5}Pd_{1.5}$, in

0953-8984/96/489815+09$19.50

which unusual divergences in specific heat, magnetic susceptibility and electrical resistivity are observed at low temperatures [1–3].

Inelastic neutron scattering measurements [4] have been used to study the energy and temperature scaling properties of the magnetic excitations in polycrystalline UCu_4Pd and $UCu_{3.5}Pd_{1.5}$ for excitation energies in the range 2–200 meV and temperatures in the range 10–300 K. The magnetic spectrum $S(\omega)$ depicted in figure 1 for UCu_4Pd is broad and quasi-elastic, with no wavevector-dependence other than that of the uranium magnetic form factor. Identical results were obtained for $UCu_{3.5}Pd_{1.5}$. The lack of any temperature-dependence for $S(\omega)$ for $\Delta\omega > 0$ argues convincingly that the imaginary part of the dynamic susceptibility $\chi''(\omega, T)$ is a function of ω/T, since

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

where $n(\omega) + 1$ is the thermal occupation factor. This result is remarkable, insofar as it indicates that the characteristic energy scale for the magnetic excitations in this system is not constant, as is found in many cerium and uranium intermetallics, including the parent compound UCu_5 [7], but rather is variable. The magnetic spectra for ω less than 25 meV and for temperatures in the range 10–300 K both for UCu_4Pd and for $UCu_{3.5}Pd_{1.5}$ can be described by a universal function of ω/T, as demonstrated in figure2:

$$\chi''(\omega/T)T^{1/3} = (\omega/T)^{-1/3}\mathcal{Z}(\omega/T) \qquad \mathcal{Z}(\omega/T) \rightarrow \text{ constant as } \omega/T \rightarrow \infty. \tag{2}$$

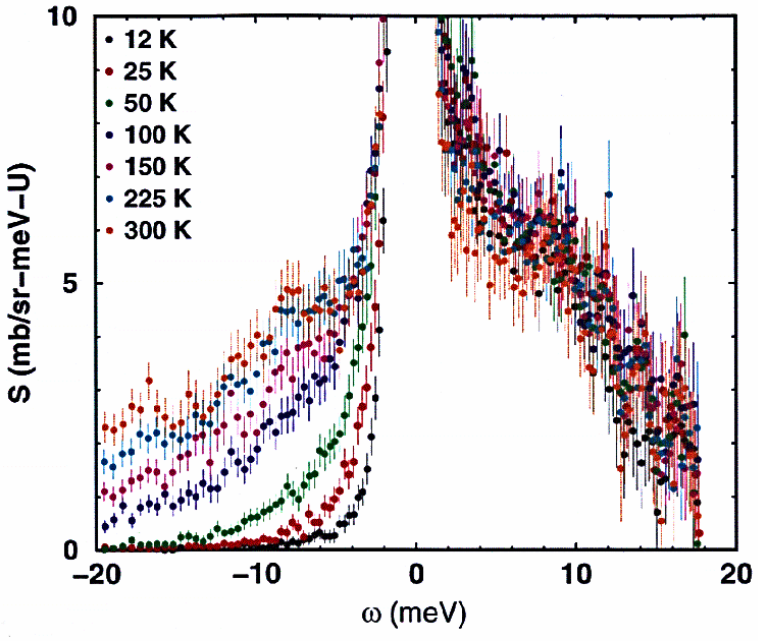

Figure 1. $S(\omega)$ of UCu_4Pd at fixed temperatures in the range 12–300 K. The incident neutron energy is 20 meV.

The results summarized in figures 1 and 2 argue that temperature provides the characteristic energy scale for the magnetic excitations in UCu_4Pd and $UCu_{3.5}Pd_{1.5}$, and is the most direct evidence that these compounds are non-Fermi liquids. It is also important to demonstrate that these excitations are responsible for the anomalous temperature-dependences observed in bulk properties such as the specific heat, electrical resistivity

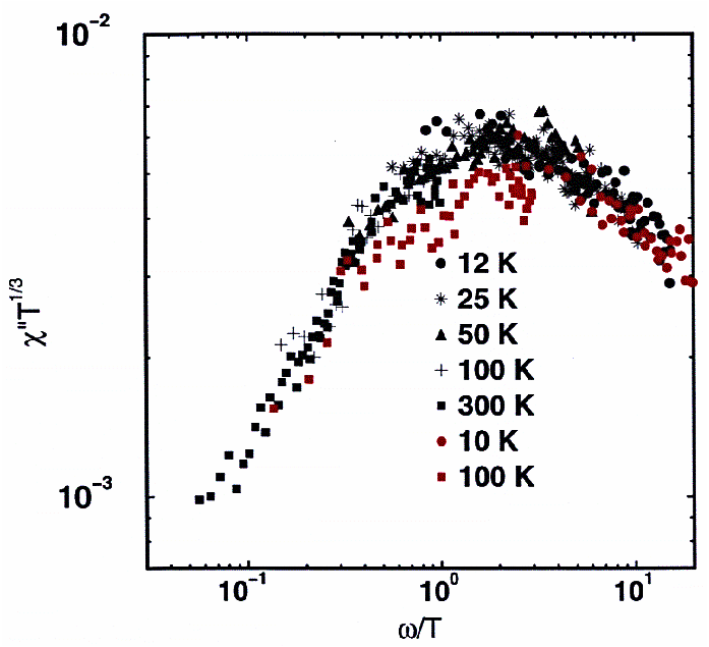

Figure 2. Universal scaling of $\chi''(\omega/T)$. Black data points represent different temperatures for UCu_4Pd; red points are for $UCu_{3.5}Pd_{1.5}$. Excitation energies greater than 25 meV are omitted.

and magnetic susceptibility. This link is made by recalling that the temperature-dependence of the static susceptibility $\chi(T)$ is related to the wavevector averaged $\chi''(\omega, \overline{q})$ by the Kramers–Kronig relation

$$\chi(T) = \frac{1}{\pi}\int \mathrm{d}\omega \frac{\chi''(\omega, \overline{q})}{\omega}. \tag{3}$$

As demonstrated in figure 3, integrating the observed magnetic spectrum from 2 to 200 meV reproduces the measured static susceptibility in the range 10–300 K in UCu_4Pd. However, excitations with energies larger than 25 meV provide only a small and nearly temperature-independent contribution to the static susceptibility. Virtually all of the diverging temperature-dependence found for $\chi(T)$ in the temperature range 10–300 K originates from the excitations having $\omega \leqslant 25$ meV, displaying ω/T scaling. Since equation (2) argues that $\chi''(\omega/T) \sim \omega^{-1/3}$ in the low-temperature limit, the Kramers–Kronig relation constrains the temperature-dependence of $\chi \sim T^{-1/3}$. Accordingly, we have plotted $(\chi(T) - \chi_0)^{-3}$ as a function of T in figure 4 to show that, for temperatures from approximately 30 K in UCu_4Pd and 10 K in $UCu_{3.5}Pd_{1.5}$ up to room temperature,

$$\chi(T) \sim T^{-\Delta} \qquad \Delta = \tfrac{1}{3}. \tag{4}$$

The divergences of χ at low temperature and χ'' at low energies argue for the existence of a low- or zero-temperature critical point. The lack of wavevector-dependence for $S(\omega)$ as well as the similarity of $S(\omega)$ in UCu_4Pd and $UCu_{3.5}Pd_{1.5}$, although the former is closer in composition to antiferromagnetic order and the latter to spin glass order, indicates that, for temperatures in the range 10–300 K and excitation energies in the range 2–25 meV, the divergences are dominated by incoherent fluctuations of individual uranium ions. The ability to regard the uranium ions as independent simplifies the theoretical analysis considerably,

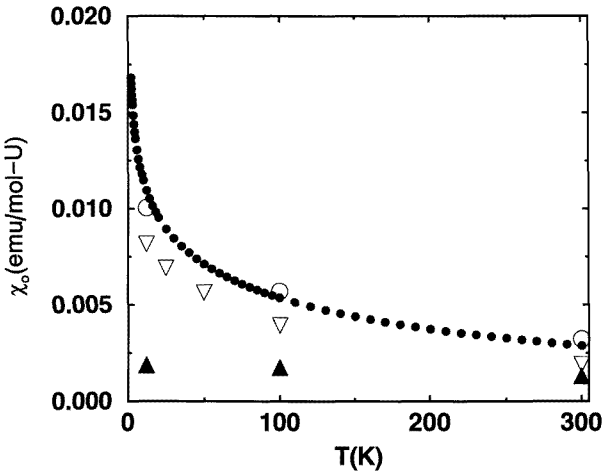

Figure 3. A comparison of the temperature-dependence of the measured static susceptibility $\chi_0(T)$ (filled circles) to those calculated by integrating $\chi''(\omega)$ from 25 meV to 200 meV (filled triangles) and from 2 meV to 25 meV (open triangles). Integrating $\chi''(\omega)$ over the entire experimental range, 2–200 meV (open circles) accounts for virtually the entire measures static susceptibility $\chi_0(T)$.

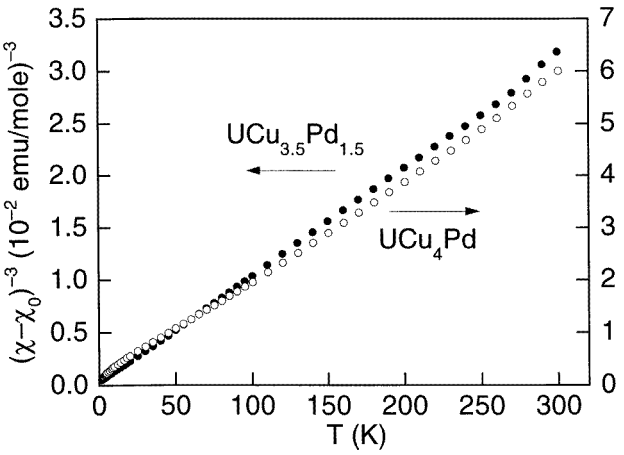

Figure 4. The temperature-dependences of the static susceptibilities $\chi_0(T)$ for both UCu_4Pd and $UCu_{3.5}Pd_{1.5}$. The measuring field is 1 T.

because single-ion critical modes have a special symmetry which may be exploited to obtain universal expressions for all properties which depend on the dynamics near a critical point [8]. Conformal field theory [9] finds that

$$\chi''(\omega,\ T) = \frac{A\mu_B^2}{T}\left(\frac{T}{T_0}\right)^{2\Delta} \mathrm{Im}\left(\frac{\Gamma\{\Delta - \mathrm{i}[\omega/(2\pi T)]\}}{\Gamma\{1-\Delta-\mathrm{i}[\omega/(2\pi T)]\}}\right) \tag{5}$$

where A is a dimensionless constant, Γ the gamma function and T_0 is a characteristic energy below which the scaling occurs. With the exception of $\Delta = 0$, which is a Fermi liquid, an intrinsic feature of impurity models is the scaling variable, ω/T. Equation (5) is compared

to the measured $\chi''(\omega/T)$ in figure 5. The best fit is obtained for $\Delta = \frac{1}{3}$. The agreement between the experimental data and equation (5) is excellent at large ω/T and the maximum is accurately predicted. Some deviation between the data and equation (5) is found at small ω/T. This is expected since the scaling form is only valid for ω, $T \ll T_0$, which we estimate to be approximately 150 K. As we will argue below, there is experimental evidence for a cross over to a qualitatively new type of scaling for ω, $T \leqslant$ 1–2 meV, and this too will compromise the quality of the fit for small ω and small T.

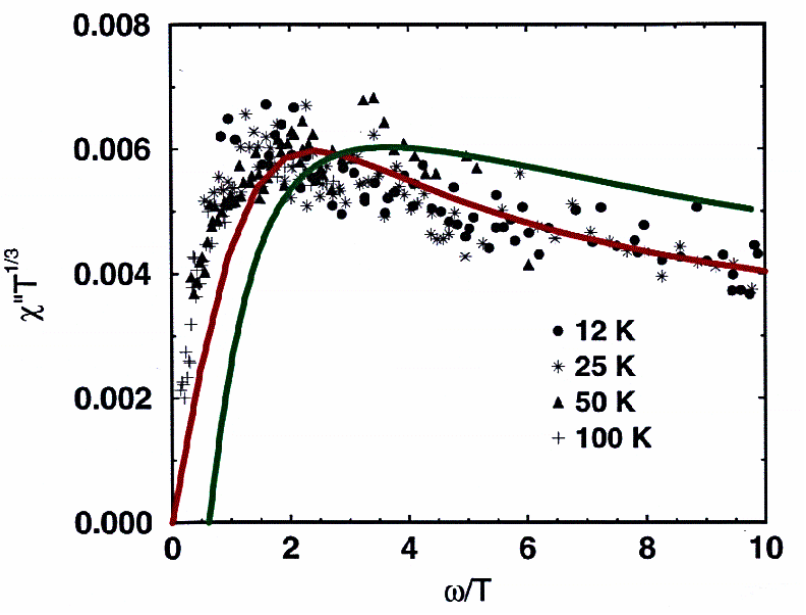

Figure 5. Scaling behaviour of $\chi''(\omega)$ in UCu_4Pd for temperatures in the range 12–100 K. The full red line is equation (5) with $\Delta = \frac{1}{3}$. The full green line is equation (8) with $T_0 = 150$ K.

The conformal field theory further predicts that the electrical resistivity has a temperature-dependence [10]

$$\rho(T) = \rho_0 \left[1 - \left(\frac{T}{T_0} \right)^{\Delta} \right]. \tag{6}$$

We have plotted the measured $\rho(T)$ of UCu_4Pd and $UCu_{3.5}Pd_{1.5}$ as functions of $T^{1/3}$ in figure 6. For both compounds, at temperatures between room temperature and about 40 K for UCu_4Pd and about 10 K for $UCu_{3.5}Pd_{1.5}$, $\rho(T)$ agrees moderately well with this expression.

The analysis presented so far suggests that, for temperatures and energies between about 1–2 meV and about 25 meV, the temperature-dependences of the resistivity and static susceptibility mirror the dynamics of individual uranium ions. Magnetic excitations on this energy range are scaled by temperature, which is a generic signature of quantum impurity models such as the multi-channel Kondo model. Further information about the effective moment degeneracy and the nature of the coupling to the conduction electrons is required to identify the nature of the impurity state itself.

There is increasing experimental evidence that the scaling behaviour associated with this impurity-like fixed point does not extend to the lowest temperatures. To demonstrate this, we have summarized the temperature-dependences of the electrical resistivity and

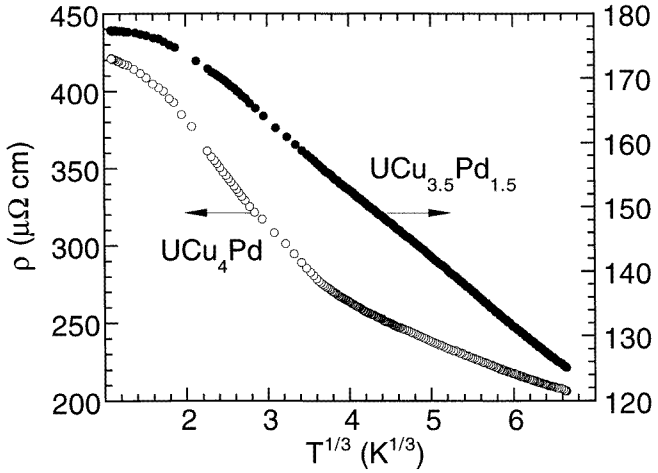

Figure 6. The temperature-dependences of the electrical resistivity $\rho(T)$ for UCu_4Pd and $UCu_{3.5}Pd_{1.5}$. For both compounds, $\rho(T) \sim T^{1/3}$ for $T^* \leqslant T \leqslant 300$ K. $T^* \simeq 50$ K for UCu_4Pd and $T^* \simeq 10$ K for $UCu_{3.5}Pd_{1.5}$.

static susceptibility, together with the scaling properties of χ'' in table 1. We stress that, although some of these results have already appeared in the literature [1–3], the temperature-dependences of all the quantities reported in table 1, with the exception of the specific heat, were obtained on the same samples of UCu_4Pd and $UCu_{3.5}Pd_{1.5}$. Tsvelik and Reizer have proposed [11] that these temperature- and field-dependences result from proximity to a three-dimensional, zero-temperature phase transition of unspecified origin, possibly related to the finite-temperature antiferromagnetic or spin glass order found at different compositions in the phase diagram.

Table 1. Temperature-dependences of various quantities of UCu_4Pd and $UCu_{3.5}Pd_{1.5}$.

UCu_4Pd	2 K $\leqslant T \leqslant$ 12 K	85 K $\leqslant T \leqslant$ 300 K
$\chi(T)$	$\log T$, $T^{-0.24}$	$T^{-1/3}$
$\rho(T)$	T	$T^{1/3}$
C/T	$T^{-1/3}$ *	?
$\chi''(\omega, T)$	?	$\mathcal{F}(\omega/T)^{\dagger}$
$UCu_{3.5}Pd_{1.5}$	2 K $\leqslant T \leqslant$ 25 K	20 K $\leqslant T \leqslant$ 300 K
$\chi(T)$	$\log T$, $T^{-0.21}$	$T^{-1/3}$
$\rho(T)$	$T^{\ddagger}$	$T^{1/3}$
C/T	$\log T$**	?
$\chi''(\omega, T)$	?	$\mathcal{F}(\omega/T)^{\dagger}$

* From [1] C/T saturates at lower temperatures.
** From [1], 0.32 K $\leqslant T \leqslant$ 10 K.
† From [4] 10 K $\leqslant T \leqslant$ 300 K, 2 meV $\leqslant \omega \leqslant$ 25 meV.
‡ $\rho(T)$ saturates at lower temperatures.

Although no direct study of the scaling properties of the magnetic excitations at temperatures and energies below 1–2 meV by means of neutron scattering has yet been published, the existing specific heat, magnetization and electrical resistivity are consistent

with an impurity-lattice cross over, analogous to those observed in many heavy-fermion systems [12]. That is, at high temperatures, the moment dynamics in UCu_4Pd and $UCu_{3.5}Pd_{1.5}$ are increasingly constrained and longer lived as the temperature is lowered due to their coupling to the conduction electrons. The spatial range of the temporal correlations presumably grows, inducing interactions among the moments. Ultimately, it is these long-range correlations which herald the approach of a three-dimensional magnetic phase transition, at or near zero temperature, whose critical fluctuations control the properties of the low-temperature, non-ordered metal.

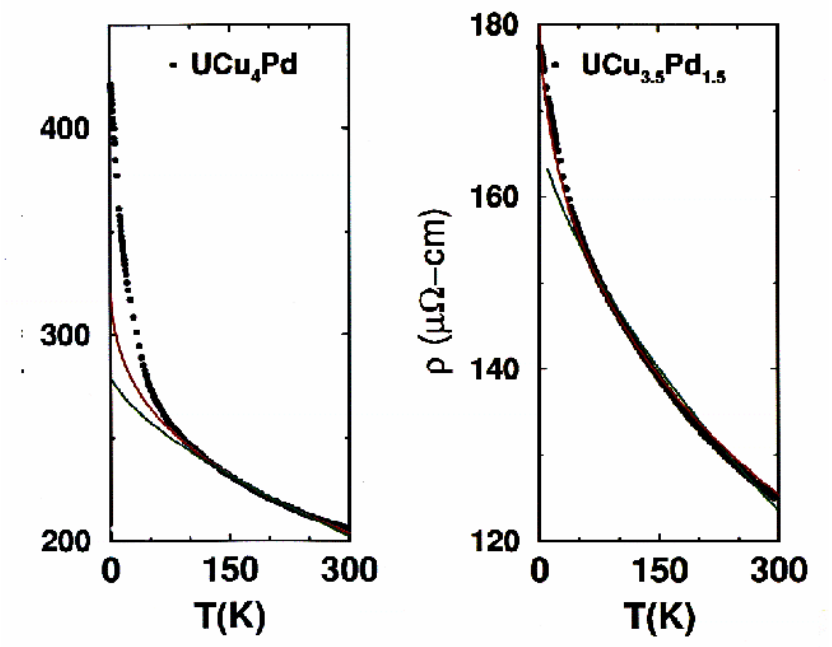

Figure 7. The electrical resistivities of UCu_4Pd (left) and $UCu_{3.5}Pd_{1.5}$ (right) with best fits $T^{1/3}$ (red) and $T^{2/3}$ (green) temperature-dependences. The former describes the resistivity in both compounds over a more extensive range of intermediate temperatures, although both forms fail below about 10 K in $UCu_{3.5}Pd_{1.5}$ and about 40 K in UCu_4Pd.

So far, our description of the non-Fermi liquid scaling properties of UCu_4Pd and $UCu_{3.5}Pd_{1.5}$ has been purely phenomenological and has not appealed to a specific microscopic mechanism. In a different view, it has recently been proposed [5, 13] that the non-Fermi liquid behaviour in UCu_4Pd and $UCu_{3.5}Pd_{1.5}$, and possibly in other materials, results from disorder. Specifically, these models assume that a wide distribution of local Kondo temperatures $P(T_K)$ exists in the system, with the most direct evidence for such a distribution being a strong inhomogeneous broadening of the copper NMR line [5]. Hence, the local moments can be divided, at a given temperature T, into two populations. *Quenched moments* have $T_K > T$, whereas *unquenched moments* have $T_K < T$. As the temperature goes to zero, the fraction of unquenched moments $f_u(T) = f_0^T P(T_K)\,\mathrm{d}T_K$ vanishes, in a manner which reflects the behaviour of $P(T_K)$ near $T_K = 0$.

The contributions of these two populations to physical quantities can be roughly estimated by saying that each quenched moment contributes to the resistivity a constant value close to the unitarity limit, $\rho \simeq \rho_0$, and to the specific heat and susceptibility

a constant value proportional to the moment's Kondo temperature, $C/T \sim \chi \propto 1/T_K$. Unquenched moments, to a first approximation, do not act as scattering centres ($\rho \simeq 0$), while contributing a Curie law to $\chi \propto 1/T$ and a negligible amount to C/T.

The logarithmically divergent specific heat ratio observed at low temperature both in UCu_4Pd and in $UCu_{3.5}Pd_{1.5}$ could thus be compatible with a distribution $P(0)$ of Kondo temperatures which is constant down to $T_K = 0$, so that $C/T \sim P(0)\ln(1/T)$. Miranda *et al* [13] also observed that, since the resistivity is entirely dominated by quenched moments ($\rho \simeq \rho_0[1 - f_u(T)]$), the linear drop in resistivity observed at low temperatures both in UCu_4Pd and in $UCu_{3.5}Pd_{1.5}$ is also consistent with this distribution, since $f_u(T) \sim P(0)T$ at low temperature. Bernal *et al* [5] used the distribution of Kondo temperatures deduced from their NMR measurements to obtain a temperature-dependent susceptibility which compares very favourably with measured values between 300 K and about 10 K in UCu_4Pd, although some deviations between the data and the predicted $\chi(T)$ were observed at lower temperatures. However, the simplest version of the Kondo disorder model with $P(T_K = 0) = P(0)$, which correctly reproduces the temperature-dependences of the specific heat and resistivity, predicts a logarithmically divergent susceptibility. As a result, the low-temperature Wilson ratio would be expected to be constant, $R_W = \chi T/C$, which is inconsistent with the experimental findings summarized in table 1. Of course, RKKY exchange interactions between local moments modify this simple estimate but can only lead to a less divergent Wilson ratio. In the context of the Kondo disorder model, these discrepancies suggest that $\chi(T)$ is much more sensitive to the detailed form of $P(T_K)$ than are the specific heat and resistivity, and that $\chi(T)$ might not have a universal temperature-dependence.

The Kondo disorder model also has a definite prediction for the scaling of the dynamical susceptibility $\chi''(\omega, T)$ at low temperature and low frequency ($T \leqslant 12$ K, $\omega \leqslant 2$ meV), which will have to be compared to experiments when data become available in this regime. Indeed, taking the approximate form for the dynamical susceptibility of a single impurity in the quenched regime: $\chi''(\omega, T) \simeq \omega/(\omega^2 + T_K^2)$ (whereas $\chi'' \simeq 1/\omega$ is the unquenched regime) and averaging over the distribution $P(T_K)$, one obtains the following scaling form:

$$\chi''(\omega, T) = P(0)\left[\frac{\pi}{2} = \tan^{-1}\left(\frac{T}{\omega}\right)\right]. \tag{7}$$

Focusing next on the intermediate frequency and temperature regime (30 K $< T <$ 300 K for UCu_4Pd, 10 K $< T <$ 300 K for $UCu_{3.5}Pd_{1.5}$), we discuss the relevance of the Kondo disorder model to the experimental results described above. In this regime, the temperature-dependence of the susceptibility is very well fitted by a power law $\chi \simeq T^{-1/3}$ [9]. As discussed above, this temperature-dependence is not consistent with constant $P(T_K)$, as proposed by Miranda, but would be consistent with a leading behaviour for $P(T_K) \simeq T_0^{-2/3}/T_K^{1/3}$ for the lowest values of the Kondo temperature in this intermediate range (T_0 is a characteristic scale). Averaging over $P(T_K)$ as above leads to the following scaling form for the dynamical susceptibility:

$$\chi''(\omega, T) = \frac{1}{T_0}\left(\frac{T_0}{T}\right)^{1/3} f\left(\frac{\omega}{T}\right). \tag{8}$$

Note that the universal scaling function depends only on the specific power law assumed for $P(T_K)$:

$$f(x) = x\int_1^{+\infty} \frac{\mathrm{d}y}{y^{1/3}} \frac{1}{x^2 + y^2}. \tag{9}$$

This function is compared in figure 5 to equation (5), which represents the conformal field theory result for single-impurity scaling. Although both the single-impurity and the Kondo-disorder models display ω/T scaling in this range of ω and T, the single-impurity scaling appears to be a somewhat better fit to the data. We note that including the full $P(T_K)$ deduced from the NMR measurements does not appreciably affect the ω/T scaling described here.

A more compelling conclusion is drawn when considering the resistivity in the intermediate-temperature range. Indeed, since the fraction of unquenched moments varies as $f_u(T) \propto (T/T_0)^{2/3}$ in the model above, the resistivity should behave as $\rho = \rho_0 - C(T/T_0)^{2/3}$. This is to be contrasted with the conformal field theory prediction, assuming that a single power of an unknown leading irrelevant operator contributes, as in the two-channel Kondo effect: $\rho = \rho_0 - AT^{1/3}$. As shown in figure 7, the $T^{1/3}$ law does appear to provide a somewhat better fit to the data in the intermediate temperature range, thus favouring single-impurity scaling. We cannot exclude the possibility that a more accurate treatment of the cross over regime in the Kondo disorder model could improve the agreement with resistivity data in this regime, and more experimental and theoretical work is certainly needed to settle the issue.

Acknowledgments

The authors acknowledge A J Millis, P Coleman, E Miranda and D E MacLaughlin for valuable discussions, and D Gajewski and C L Seaman for performing the electrical resistivity and magnetic susceptibility measurements. MCA is grateful to the Phoenix Memorial Project, University of Michigan and to the NATO International Scientific Exchange Programmes for travel support. Research at the University of California, San Diego was supported by the National Science Foundation under grant DMR-94-08835. Work at Argonne was performed under the auspices of the USA Department of Energy, Office of Basic Energy Sciences, under contract W-31-1090-ENG-38.

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