Critical reexamination of non-Fermi-liquid characteristics of moderately disordered UCu₄Pd

A. Otop,¹ S. Süllow,¹ E. W. Scheidt,² and J. A. Mydosh^{3,4}

¹Institut für Physik der Kondensierten Materie, TU Braunschweig, 38106 Braunschweig, Germany

²CPM, Institut für Physik, Universität Augsburg, 86159 Augsburg, Germany

³II. Physikalisches Institut, Zülpicher Strasse 77, 50937 Köln, Germany

⁴Kamerlingh Onnes Laboratory, Leiden University, 2300 RA Leiden, The Netherlands

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We present a detailed analysis of the susceptibility at temperatures 1.8-300 K of differently heat treated samples of the non-Fermi-liquid compound UCu_4Pd . We observe significant sample-to-sample dependencies, which we quantify by parametrizing the data using different model approaches previously reported in the literature. We discuss the implications of our findings and, in particular, consider the issue of evaluating the quality of data parametrizations to identify non-Fermi-liquid behavior in heavy fermion systems and related materials.

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I. INTRODUCTION

In recent years, the label non-Fermi-liquid (NFL) behavior has been coined to describe the temperature and energy dependence of certain physical properties, which deviate from the predictions of the well-established Fermi liquid theory. In particular, NFL behavior has been observed for a variety of strongly correlated and heavy fermion compounds, which are situated in the vicinity of a magnetic instability being tuned to zero temperature, i.e., quantum critical behavior.^{1–3}

As a fingerprint of NFL behavior at low *T*, the main bulk properties display unconventional *T* dependencies such as a logarithmic or power-law electronic specific heat $(C/T \sim \ln T \text{ or } \sim T^{1+\lambda})$ (Refs. 4–15) and magnetic susceptibility $[\chi \sim T^{-1+\lambda}, \sim (1-T^{1/2}) \text{ or } \sim \ln(T/T_0)]$,^{7–14,16–19} as well as a nonquadratic electrical resistivity $(\rho - \rho_0 \sim T/T_0, \sim T^{x<2})$ or $\sim T^{-1+\lambda}$).^{4–6,8,12,14,17,19} Conceptually, parametrizing these quantities with such scaling laws represents a crucial element in the analysis of NFL behavior and is used to verify the predictions of various theoretical and phenomenological models.^{9,10,20–23} Now, for many heavy fermion systems, the temperature ranges where the above NFL scaling approaches can be applied turn out to be different for C/T, χ , or ρ . Correspondingly, so far, none of the various theoretical models proposed can fully describe the *T* dependence of the physical properties of NFL materials.

Furthermore, it has been shown that the electrical resistivity of moderately disordered heavy fermion compounds reflects disorder induced localization effects.^{24,25} For NFL systems, this observation becomes relevant in so far as these are intermetallic compounds typically derived by substitution from a parent material and therefore will be crystallographically disordered because of random site occupation or metallurgical imperfections resulting from strain. In consequence, anomalous *T* dependencies of the resistivity reported for such materials do not indicate NFL behavior but instead conductivity corrections from disorder induced localization.

In view of such pronounced and ubiquitous disorder effects in the resistivity, the question arises if other properties are similarly affected by disorder. To answer this question, criteria are needed to distinguish between generic NFL behavior and disorder effects. These criteria should provide guidelines for the data analysis in the search for NFL modeling.

For instance, considering the magnetic susceptibility, as yet no such criteria exist, neither for fitted *T* ranges nor specific *T* dependencies. In experimental studies, the scaling functions and fit parameters for this property are chosen quite arbitrarily and are valid often only in very narrow temperature ranges. The question arises about the significance of a specific *T* dependence of χ for identifying the source for the NFL behavior in a given material. In order to assess this issue, we have carried out a detailed study of the magnetic properties of differently treated samples UCu₄Pd. In doing so, we introduce a procedure to assess the quality of NFL fits of the susceptibility, but which can also be used to analyze other physical properties regarding the presence and significance of NFL behavior.

UCu₄Pd is a NFL system derived by Pd substitution in the Kondo lattice antiferromagnet UCu₅.¹⁴ According to Ref. 14, the electronic specific heat and magnetic susceptibility of the as-cast material are well described by a power-law function at temperatures 1–10 K (c_p) and 1.8–10 K (χ), respectively. Further, the electrical resistivity is found to be linear in *T* from 10 down to about 0.3 K. A subsequent alternative analysis of the specific heat data from Ref. 14 in the range 0.3–10 K was taken as evidence for the Kondo disorder.²⁶

These early results served as a starting point for various experimental studies on this material and the discussion of the microscopic origin for NFL behavior in UCu₄Pd and related materials. Especially, an extensive study of the crystal-lographic structure of UCu₄Pd by means of x-ray absorption fine-structure analysis²⁷ revealed that the system is disordered, with ~25% Cu/Pd random site exchange. Further, it has been demonstrated that the actual level of disorder in this compound depends on the metallurgical treatment and that the disorder level can be reduced through annealing.^{15,28} The annealing dependence of the structural properties translates into one of the magnetic ground state. Initial studies^{29,30} reported the observation of a spin glass phase for UCu₄Pd below a freezing temperature $T_f \sim 0.15$ K. Additional work



FIG. 1. (Color online) Magnetic susceptibility $\chi(T)$ and inverse susceptibility $\chi^{-1}(T)$ of as-cast and annealed samples UCu₄Pd; for details see text.

revealed that the actual nature of the ground state—spin glass and superpara- or antiferromagnet—depends on the metallurgical treatment.³¹ As well, the *T* dependence of the specific heat was demonstrated to depend on the sample quality,^{31,32} but that neither the Kondo disorder model⁹ nor the Griffiths phase scenario¹⁰ consistently explained the data below ~ 1 K.

In order to study the influence of crystallographic disorder on the magnetic properties of UCu₄Pd, we have carried out magnetic susceptibility measurements at temperatures 1.8–300 K on two different sets of samples which were prepared by the groups in Augsburg and Leiden, respectively (labeled as **A**=Augsburg and **L**=Leiden; for details concerning sample preparation, see Refs. 15 and 25). Altogether, we have analyzed the magnetic susceptibility of five samples: two as cast, **A**_{as} and **L**_{as} as well as three annealed, **A**_{1ann}, **A**_{2ann}, and **L**_{ann}. The annealing process has been performed in an evacuated quartz tube at 750 °C for 7 days (**A**_{1ann}), at 750 °C for 14 days (**A**_{2ann}), and at 900 °C for 7 days: **L**_{ann}. We compare our results to previously published data, for which different NFL scalings have been applied, and assess the applicability of these different approaches.

II. RESULTS

We have performed dc magnetic susceptibility measurement ($\chi \equiv M/H$) in a conventional superconducting quantum interference device magnetometer at a low magnetic field³³ of B=0.01 T for the **A** samples and B=0.001 T for both **L** samples at temperatures 1.8–300 K (see Fig. 1). The overall T dependence of χ for the different samples is very similar among themselves as well as to that previously published for as-cast UCu₄Pd.^{16–19} The magnetic susceptibility of the annealed samples at low temperatures is somewhat smaller than that of the as-cast samples but shows no additional features. TABLE I. Paramagnetic properties of UCu₄Pd, as obtained from Curie-Weiss fits to the high temperature susceptibility, with μ_{eff} as the effective moment and Θ_{CW} as the Curie-Weiss temperature. The temperature ranges for the fits are I=200–300 K, II=80–200 K, and III=80–300 K.

Sample	$rac{\mathrm{I}}{\mu_{eff} / \Theta_{CW}} \ (\mu_B / \mathrm{K})$	$\begin{matrix} \Pi \\ \mu_{eff} / \Theta_{CW} \\ (\mu_B / \mathrm{K}) \end{matrix}$	$\displaystyle egin{array}{c} \mathrm{III} \ \mu_{eff} / \Theta_{CW} \ (\mu_B / \mathrm{K}) \end{array}$
L _{as}	3.78/-213	3.46/-140	
\mathbf{L}_{ann}	3.13/-132	3.20/-140	
\mathbf{A}_{as}			3.21/-126
\mathbf{A}_{1ann}	3.12/-79	3.39/-128	
A _{2ann}			3.19/-125

In the following, we will analyze the data according to the procedures commonly used to identify NFL behavior in heavy fermion metals. In a first step, we need to verify that the samples studied here compare favorably to those studied previously. For this, we note that at high temperatures, the magnetic susceptibility appears to be Curie-Weiss-like,³⁴

$$\chi = \frac{C}{T - \Theta_{CW}},\tag{1}$$

where $C = \frac{N_A \mu_{eff}^2}{3k_B}$, Θ_{CW} =Curie-Weiss temperature, and μ_{eff} is the effective moment in units of Bohr magnetons. For \mathbf{L}_{as} , a linear fit to χ^{-1} (Fig. 1) between 200 and 300 K yields a value μ_{eff} =3.78 μ_B and a very low Θ_{CW} =-213 K. Further, in the range from 200 to 80 K χ^{-1} , is also linear in *T* but with a different slope. Here, a fit yields μ_{eff} =3.46 μ_B and Θ_{CW} =-140 K.

For L_{ann} , the same type of analysis results in μ_{eff} = 3.13 μ_B , Θ_{CW} =-132 K and μ_{eff} = 3.20 μ_B , and Θ_{CW} =-140 K for the *T* ranges 300–200 and 200–80 K, respectively. These data alone would seem to suggest that the presence of two temperature ranges in $\chi^{-1}(T)$ is related to the actual level of disorder, as for the annealed sample, the fit parameters for the two temperature regimes yield quite similar results.

However, as can be gathered from Fig. 1, this hypothesis is not confirmed by the measurements on the A samples. Here, as-cast \mathbf{A}_{as} and annealed \mathbf{A}_{2ann} samples show a very similar $\chi^{-1}(T)$ dependence, with a linear scaling over the range 300–80 K. In contrast, for the annealed sample \mathbf{A}_{1ann} , a change of slope occurs in $\chi^{-1}(T)$ around 200 K, just as for the L samples. As before, we fit the data to Eq. (1) and summarize the results in Table I.

Similar to our observations, effective moments μ_{eff} = 3.00–3.44 μ_B and Curie-Weiss temperatures Θ_{CW} ~-60 to -260 K have been observed for UCu_{5-x}Pd_x, with Pd concentrations x=0-2.2 at T>200 K (Refs. 14 and 18) and $100 \le T \le 400$ K,¹⁶ respectively. All these values for the effective moment are, with the exception of L_{as} above 200 K, slightly lower than expected for the valence states commonly assumed for free uranium ions in a metallic environment (f^2/f^3 : $\mu_{eff}=3.58/3.62\mu_B$). This probably would in-



FIG. 2. (Color online) The *T* dependence of the magnetic susceptibility of UCu₄Pd, samples L and A, plotted as $\chi - \chi_0 \sim T^{-1/3}$; for details see text.

dicate the presence of crystalline electric fields and/or the Kondo effect modifying the temperature dependence of the magnetic susceptibility.¹⁸

For temperatures $\sim 30-300$ K, Aronson *et al.*¹⁹ proposed a NFL scaling of the static magnetic susceptibility

$$\chi - \chi_0 \sim T^{-1/3}$$
. (2)

Correspondingly, we analyze the magnetic susceptibility of \mathbf{L}_{as} , \mathbf{A}_{as} , and \mathbf{A}_{2ann} by plotting $\chi - \chi_0$ as a function of $T^{-1/3}$ (Fig. 2). For A_{as} and A_{2ann} , and to a lesser degree for L_{as} as well, Eq. (2) does reasonably well reproduce the experimental data down to ~100 K. However, the values χ_0 obtained from fits using this equation yield quite unreasonable diamagnetic values $(\chi_0 = -2.3/-2.8/-3 \text{ memu/mole})$ for $\mathbf{L}_{as}/\mathbf{A}_{as}/\mathbf{A}_{2ann}$, respectively), questioning this approach. Further, for L_{ann} and A_{1ann} , a dependence as in Eq. (2) is not observed. In Fig. 2, we include the data for L_{ann} and A_{1ann} after forcing fits to Eq. (2) to yield the best matching $(\chi_0 = -2.5/-4 \text{ memu/mole for } \mathbf{L}_{ann}/\mathbf{A}_{1ann})$. Fixing χ_0 at zero obviously does not improve the situation, and it does nothing to remove the qualitatively different behavior of the various samples.

In previous studies^{17,18} on UCu₄Pd, the susceptibility of as-cast samples at low temperatures ≤ 12 K has been analyzed using a NFL-like logarithmic temperature dependence proposed to indicate Kondo disorder,⁹

$$\chi = \chi_0 \ln\left(\frac{T}{T_0}\right). \tag{3}$$

Correspondingly, we parametrize our data between 1.8 and 12 K in the same way. The result is depicted in Fig. 3(a), with the parameters obtained from the fits summarized in Table II. Here, we include two literature values. Altogether, the matching between fit and data is quite reasonable. Only for A_{as} , there appears to be a systematic deviation visible in the normalized difference between data and fit, $(\chi - \chi_{fit})/\chi$ [Fig. 3(b)], and which would imply that here a ln(*T*) parametrization fails. Aside from the values reported in Ref. 17 the fit parameters in Table II are of similar order of magnitude. The extensive deviation of the Ref. 17 values seems to indicate an incorrect fit formula.

A priori, there is no reason to restrict fits using Eq. (3) to



FIG. 3. (Color online) (a) Low temperature susceptibility of UCu₄Pd, samples L and A. Data are as in Fig. 1, but shifted for clarity. Solid lines represent fits to Eq. (3). (b) Normalized difference between measured susceptibility and fits of UCu₄Pd; for details see text.

a certain temperature range. Within the "NFL spirit," the common assumption is that NFL behavior occurs at low T and that subsequently the T range of NFL behavior can be determined by extending the fitted T range. Here, a criterion is missing to measure the adequacy of this approach, i.e., how the quality of the fit evolves as the fitted temperature range changes. One such measure would be a relative deviation between fit and data per number of data points,

$$\Gamma[T(N)] = \frac{\sum_{i=1}^{N} \frac{|\chi_i(T) - \chi_{fit,i}(T)|}{\chi_i(T)}}{N},$$
(4)

where N is the number of data points in a particular fit, with

TABLE II. Fit parameters of the magnetic susceptibility, using as fit function $\chi = \chi_0 \ln \frac{T}{T_0}$; for details see text.

Sample	Scaling range	χ ₀ (memu/mole)	<i>Т</i> ₀ (К)
$\overline{\mathbf{L}_{as}}$	3–12	-3.9	299
Lann	2-12	-2.7	616
\mathbf{A}_{as}	1.8-12	-4.1	227
\mathbf{A}_{1ann}	1.8-12	-3.3	539
A _{2ann}	1.8-12	-3.0	518
Ref. 17	2-10	8.7	11
Ref. 18	2–12	-3.3	428



FIG. 4. (Color online) Temperature dependence of the fit-quality function $\Gamma(T_{max})$, as defined in Eq. (4), for L samples UCu₄Pd; lines are guides to the eyes; for details see text.

the count beginning at the lowest temperature and increasing with *T*. Thus, the number of data points *N* specifies a range $\Delta T(N) = [T(1), ..., T(N)]$, starting at the lowest measured temperature $T(1) = T_{low}$ up to the highest fitted one T(N) $= T_{max}$. $\chi_i(T)$ denotes the *i*th data point in $\Delta T(N)$ and $\chi_{fit,i}(T)$ represents the corresponding fit point. For each range $\Delta T(N)$, a set of fit parameters is determined and χ_{fit} calculated. The argument of the sum represents the relative deviation between fit and data at temperature T_i [Fig. 3(b)]. The sum provides the measure for the overall mismatch between fit and data in the range $\Delta T(N)$, viz., the fit quality. Division by *N* yields the average mismatch per point.

With this notation, $\Gamma(T_{max})$ represents a measure for the quality of a fit in the temperature range $\Delta T(N)$ =[T_{low}, \ldots, T_{max}], provided that N is not too small.³⁵ If a fit formula does adequately describe the data, then $\Gamma(T_{max})$ is constant, its absolute value being controlled by the scatter of the data. Conversely, an inappropriate fit formula leads to a continuously increasing $\Gamma(T_{max})$ with T_{max} since with a larger range $\Delta T(N)$, the mismatch will become worse.

To analyze if a function such as $\Gamma(T_{max})$ provides a tool to derive the proper temperature range for fits of the susceptibility of UCu₄Pd using Eq. (3), we have calculated its value for the L samples (Fig. 4). Initially, for both samples, as temperature increases from T_{min} , $\Gamma(T_{max})$ rises continuously. Subsequently, for L_{ann} , there is a plateau in $\Gamma(T_{max})$ up to about 12 K, consistent with the range $\Delta T(N)=1.8-12$ K being adequately described by Eq. (3). In contrast, for L_{as} , there is no plateau, implying that a T dependence $\propto \ln(T)$ does not in full detail account for the data (the same behavior is observed for the A samples). In essence, it implies that the slight structure seen in $(\chi - \chi_{fit})/\chi$ for L_{as} [see Fig. 3(b)], with a maximum mismatch between fit and data <1%, already implies that Eq. (3) does not fully reproduce the experimental data in the temperature range $T \le 12$ K.

A central prediction of a disorder-based model, the Griffiths phase scenario,^{10,36} is that a power-law behavior should be observed in quantities such as magnetic susceptibility or specific heat above a crossover temperature T^* . Below T^* , magnetic susceptibility and specific heat should diverge even stronger than a power law.^{10,36} This model is of particular interest for UCu₄Pd because in this material, disorder clearly is an issue.^{27,29–32} Hence, in Fig. 5, we analyze the magnetic



FIG. 5. (Color online) (a) Low temperature susceptibility of UCu_4Pd , samples L and A. Data are shifted for clarity. Solid lines represent fits to Eq. (5). (b) Normalized difference between measured susceptibility and fits of UCu_4Pd ; for details see text.

susceptibility for temperatures 1.8-12 K using as fit function

$$\chi = AT^{-\eta}.$$
 (5)

Again, the resulting fit parameters are listed in Table III. Overall, compared to the approaches used above, the matching between data and fit is the best obtained, with less than 1% normalized difference [see Fig. 5(b)]. Further, the parametrization yields quite consistently a value $A \sim 0.020$ K⁷ emu/mole and $\eta \sim 0.25$ for all the samples studied here as well as for samples investigated by other groups.^{16,18}

As before, we need to verify the stability of our fit and assess the validity of the choice for the fitted temperature range. For this, we have determined the values A and η at temperature $T=T_{max}$ obtained by fitting our

TABLE III. Parameters from fits to the magnetic susceptibility below 12 K with $\chi = AT^{-\eta}$ (with the exception of Ref. 16, where the scaling range is 2–20 K). In Refs. 16 and 18, as-cast samples have been measured and A values have not been reported; for details see text.

Sample	η	$A \\ (K^{\eta} emu/mole)$
$\overline{\mathbf{L}_{as}}$	0.26	0.024
Lann	0.21	0.018
\mathbf{A}_{as}	0.27	0.024
A _{1ann}	0.18	0.025
A _{2ann}	0.22	0.020
Ref. 16	0.27	
Ref. 18	0.26	



FIG. 6. (Color online) The values A and η obtained by fitting the susceptibility of UCu₄Pd, samples L and A, to Eq. (5) in the temperature range $\Delta T(N) = [T_{low}, \ldots, T_{max}]$ and the corresponding value of the fit-quality function $\Gamma(T_{max})$. Error bar in the $\Gamma(T_{max})$ plot indicates typical uncertainty for this quantity; for details see text.

susceptibility data to Eq. (5) in the temperature range $\Delta T(N) = [T_{low}, \dots, T_{max}]$. In addition, we have calculated the corresponding value of the fit-quality function $\Gamma(T_{max})$. In Fig. 6, we plot the result of our analysis.

The figure reveals two salient points. First, as long as the fitted temperature range is small (in our case up to about 5 K), the resulting fit is comparatively unstable, with A and η varying by up to 10%. Since in this T range the fit-quality function $\Gamma(T_{max})$ for all samples rises from its N=1 fixed value of 0 to a data scattering controlled plateau at ~5 K, it suggests that the variations of A and η are equally scattering controlled.

Second, for *T* exceeding ~5 K, the temperature dependence of *A* and η is weak and without structure. For three of the samples, a broad plateau in the *T* dependence of the fit-quality function Γ occurs, with the plateau ranging up to ~18/17/10 K for $\mathbf{L}_{ann}/\mathbf{A}_{as}/\mathbf{A}_{1ann}$, respectively. For the remaining two samples \mathbf{L}_{as} and \mathbf{A}_{2ann} , plateaus appear to exist, but only for very narrow temperature ranges up to 7 and 6 K. This observation implies that at least for limited temperature ranges, a fit using Eq. (5) appropriately describes the experimental data. Further, the absolute value of the fit-quality function in Fig. 6 is 1 order of magni-

tude smaller than fitting the data by Eq. (3). Thus, the Griffiths phase scenario with Eq. (5) yields a much better matching between fit and data than the Kondo disorder model via Eq. (3). Taking all these findings together, we conclude that the best data parametrization of the susceptibility in the temperature range $1.8 - \sim 10$ K is provided by the Griffiths phase scenario with an expression $\chi = AT^{-\eta}$, with $A \sim 0.021 \pm 0.004$ K^{η} emu/mole and $\eta \sim 0.23 \pm 0.05$. The large error bars for A and η reflect sample-to-sample variations, as seen in Fig. 6.

At face value, of all the NFL models proposed to account for the behavior of UCu₄Pd so far, the Griffiths phase appears to be the most successful one. Yet, we stress that the above analysis represents only a data parametrization. From an experimental study such as ours, no physical justification for the use of a particular fit function can be obtained. One point of criticism previously raised against the Griffiths phase scenario, i.e., that the model does not provide criteria to assess if a certain value for a fitted parameter η or A is meaningful, is still not resolved.

III. CONCLUSION

Summarizing, we have performed a detailed analysis of the susceptibility of differently heat treated samples of the non-Fermi-liquid compound UCu₄Pd. We observe significant sample-to-sample dependencies, which we quantify by parametrizing the data using different approaches previously reported in the literature. Of all the approaches used, only a parametrization suggested within the Griffiths phase scenario yielded a consistent set of fitting parameters and good agreement between experiment and fit for all samples in the temperature range studied here.

There are still various issues which need to be clarified in the future. First of all, there are various objections raised against the Griffiths phase scenario as such, which have been discussed in literature previously. Further, there is the even more fundamental question of why a thermodynamic quantity such as the susceptibility for samples which are clearly in the thermodynamic limit should exhibit such a pronounced sample dependence as observed in our study. More cleaner samples-preferably studies on single crystalline-or more comparative studies on different samples, and all this down to much lower temperatures, will be required to deduce the intrinsic physical properties of such materials. Additional investigations of the sample dependencies via nonlinear susceptibility and specific heat are warranted.

Regarding the analysis of such data, the ambiguity of NFL fits and the carelessness of using them is highlighted by our detailed data analysis. In this context, the parametrization approach tested here lends itself to be applied instead. In particular, it can be used similarly to analyze other quantities such as the specific heat or the resistivity, which we believe would help to gain a better understanding of NFL behavior in correlated electron systems.

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