Order and non-Fermi-liquid behavior in UCu_4Pd

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We have studied the role of disorder in the non-Fermi-liquid system UCu_4Pd using annealing as a control parameter. Measurement of the lattice parameter indicates that this procedure increases the crystallographic order by rearranging the Pd atoms from the 16*e* to the 4*c* sites. We find that the low-temperature properties depend strongly on annealing. Whereas the non-Fermi-liquid behavior in the specific heat can be observed over a larger temperature range after annealing, the clear non-Fermi-liquid behavior of the resistivity of the unannealed sample below 10 K disappears. We come to the conclusion that this argues against the Kondo disorder model as an explanation for the non-Fermi-liquid properties of both as-prepared and annealed UCu_4Pd.

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I. INTRODUCTION

Since the first observation of non-Fermi-liquid behavior in 1991,[1] many intermetallic compounds have been found with low-temperature thermodynamic properties that cannot be described within Landau's Fermi-liquid theory ($\gamma = \text{const}, \chi = \text{const}, \rho - \rho_r \propto T^2$). Typically, such alloys show a logarithmic or a weak power-law dependence in C/T and in the magnetic susceptibility χ at low temperatures, as well as a non-T^2 dependence of the electrical resistivity ρ (for example $\rho - \rho_r \propto T$). All of these compounds are close to magnetism. In searching for further examples for non-Fermi-liquid behavior, Andraka and Stewart[2] characterized UCu_4Pd as a non-Fermi-liquid. They found a power-law temperature dependence of the Sommerfeld coefficient $\gamma = C/T = \Delta T^{-\delta}$ ($\Delta = 450$ mJ/mole $K^{2-\delta}$, $\delta = 0.32$) over a decade in temperature between 1 and 10 K: the magnetic susceptibility χ also followed a power law with the same δ between 1.8 K and 10 K. Furthermore, the electrical resistivity increased linearly below 10 K with a slope six times larger than that in $U_{0.2}Y_{0.8}Pd_3$.[1] A possible explanation for this non-Fermi-liquid behavior is, according to Ref. 2, the proximity to antiferromagnetism suppressed to $T_N = 0$ in this compound. This is called the ''quantum critical point'' (QCP) scenario.

Following this seminal work, much research has been done on UCu_4Pd and its physical properties have been explained by various other theories. Castro Neto *et al.*[3] suggested that a *Griffiths phase* causes the non-Fermi-liquid behavior, whereupon de Andrade *et al.*[4] fit their UCu_4Pd data down to 0.6 K to this theory. Scheidt *et al.*,[5] however, showed that their C/T data down to 60 mK matches this model only in a small temperature range around 1 K. This was also confirmed by Vollmer *et al.*[6]

Bernal *et al.*[7] interpreted the non-Fermi-liquid behavior of UCu_4Pd within the framework of the *Kondo disorder model*. This model presupposes a distribution of microscopic exchange couplings and was argued to successfully explain the observed non-Fermi-liquid behavior in the magnetic susceptibility and the specific heat.

UCu_4Pd is particularly well suited for studying the influence of disorder because of the possibility of perfect chemical ordering in this compound, which crystallizes in the cubic $AuBe_5$ structure. In this ideal case the Pd atoms would occupy the 4*c* sites and the Cu atoms would occupy the smaller 16*e* sites. X-ray absorption fine-structure (XAFS) experiments,[8] however, indicate that in an unannealed sample of UCu_4Pd only 76% of the Pd atoms populate the 4*c* sites, and the remaining 24% are on the 16*e* sites. Another hint for disorder in this compound is, according to Ref. 9, that the change of the slope of the lattice parameter of $UCu_{5-x}Pd_x$ versus the Pd concentration x occurs at $x \approx 0.85$ and not as would be expected at $x = 1$ for an ideal, ordered compound.

In this work we study annealed UCu_4Pd samples, where the Pd atoms are rearranged from the 16*e* sites to the 4*c* sites. This allows us to study UCu_4Pd with less (but not zero) intrinsic disorder in order to analyze the importance of disorder for its non-Fermi-liquid behavior. The original discovery work[2] reported the effect of annealing on $UCu_{3.5}Pd_{1.5}$, but not on UCu_4Pd. The only other annealing study of which we are aware reported that,[10] within their error bar, no difference in disorder in ball milled (which introduces strain that broadens diffraction peak linewidths) unannealed and annealed (at 925 °C for one week) samples of UCu_4Pd could be found in elastic neutron diffraction measurements.

We show by x-ray, ac-susceptibility, resistivity, and specific heat data that the physical properties of UCu_4Pd are extremely sensitive to annealing. We conclude that disorder may not be the underlying mechanism for the non-Fermi-liquid behavior in the specific heat and resistivity.

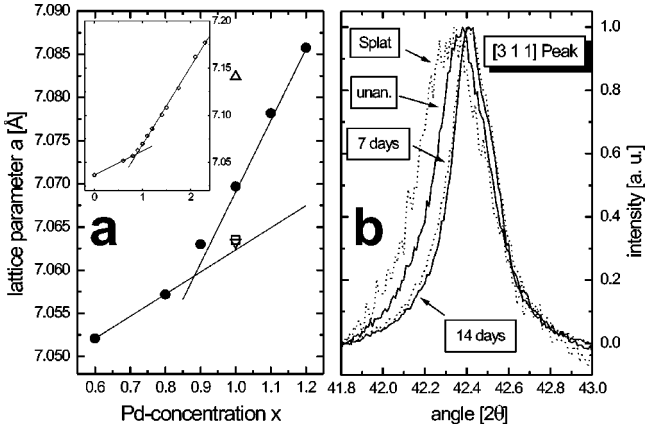

FIG. 1. (a) The lattice parameter a as a function of the Pd concentration x of unannealed $UCu_{5-x}Pd_x$ (solid circles). The open square shows the lattice parameter of our 7-day annealed sample, the open down triangle the 14-day annealed sample. Both lie nearly on the straight line (see inset) that fits the data lower than $x=0.8$. The splat-cooled sample (open up triangle) has a lattice parameter similar to that of $UCu_{3.8}Pd_{1.2}$. The inset shows the lattice parameter of unannealed $UCu_{5-x}Pd_x$ over a large range of concentration x (Ref. 9). Notice that the lattice expansion changes at $x\approx 0.85$. The straight lines are linear fits to the data for $x\leqslant 0.8$ and $x\geqslant 1.1$. (b) The [3 1 1] peak of all samples of UCu_4Pd. Upon annealing the peak width becomes smaller, which indicates increasing order: a quantitative analysis gives for the full width at half maximum 0.39°, 0.31°, 0.27°, 0.25° with $\pm 0.01°$ accuracy in this sequence.

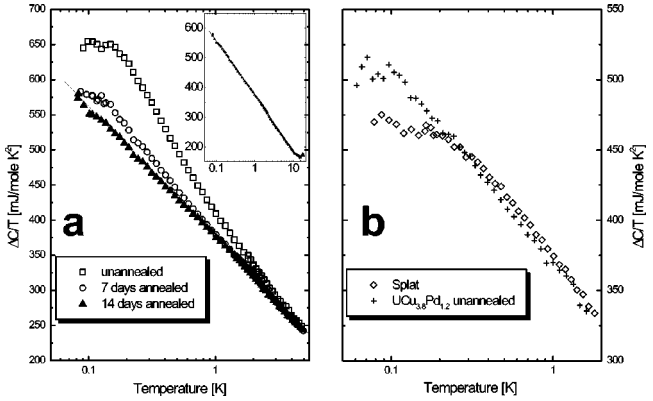

FIG. 2. The electronic part of the specific heat $\Delta C/T$ over the temperature as a function of temperature for (a) UCu_4Pd unannealed and annealed and (b) UCu_4Pd splat-cooled and $UCu_{3.8}Pd_{1.2}$. With increasing annealing time the non-Fermi-liquid regime expands up to more than two decades (inset). The data of splat-cooled UCu_4Pd matches $UCu_{3.8}Pd_{1.2}$ above 200 mK.

II. SAMPLE PREPARATION

The samples of UCu_4Pd presented in this work were all obtained from the same batch. This batch was arc-melted under a highly purified argon atmosphere. To obtain the highest possible homogeneity, the sample was flipped over four times and remelted. The loss in weight was 0.2%. The sample was then cut in several pieces. These pieces were treated in different ways: one was measured as cast, one was annealed in an evacuated quartz glass tube for seven days at 750 °C, one for 14 days at 750 °C and another one was splat-cooled (cooling rate more than 10^4 K/s) as a way to introduce more disorder than in the as-cast sample. All these samples were identified as single phase ($AuBe_5$ structure) in extra long counting time powder diffraction measurements. Also in specific heat and ac-susceptibility measurements no impurity phases such as UCu_5 or UPd_3 were detected within 1% accuracy. The annealed samples showed no weight loss within 0.1% accuracy.

III. RESULTS AND DISCUSSION

In the inset of Fig. 1 the lattice parameter a of unannealed $UCu_{5-x}Pd_x$ is plotted versus the Pd concentration x. The concentration dependence of the lattice parameter a can be described by two straight lines with an intersection at $x\approx 0.85$. This points to the beginning of augmented (i.e., above random disorder present for $x<0.85$) occupation of the smaller 16*e* sites by the Pd atoms at this $x=0.85$ concentration.[9] The expanded plot around $x=1$ [Fig. 1(a)] shows in addition the lattice parameters of the two annealed and the one splat-cooled samples. One observes that the lattice parameters of the annealed samples nearly lie on the straight line that fits the data points from unannealed samples with Pd concentrations lower than $x=0.8$. The decrease of the lattice parameter with annealing can be attributed to a thermally activated movement of the Pd atoms from the smaller 16*e* sites to the bigger 4*c* sites. The lattice parameter of the splat-cooled sample, however, is clearly much larger than the unannealed one, which is indicative of increased disorder. The lattice parameter of the splat-cooled UCu_4Pd sample is almost the same as that of unannealed $UCu_{3.8}Pd_{1.2}$.

A further test for increasing order upon annealing is the full width at half maximum of an x-ray peak. As an example, Fig. 1(b) shows the [3 1 1] peak from x-ray powder diffraction measurements of the splat-cooled, the unannealed, and the 7- and the 14-day annealed UCu_4Pd samples. The full width at half maximum clearly gets smaller in this sequence, which is a strong indication of increasing order with annealing the samples. The resistivity ratio of the 7-day and 14-day annealed samples between $T=1.8$ K and $T=400$ K decreases by 11% (see Fig. 3), which also supports increasing order for extended annealing.

With these samples with different degrees of disorder, we are now able to make a systematic study of the role that disorder plays for the physical behavior of UCu_4Pd. In Fig. 2(a) $\Delta C/T$ is displayed versus $\ln_{10} T$ for all these samples except the splat-cooled one. It can be seen that the antiferromagnetic transition[9] of the unannealed UCu_4Pd sample ($T_N\approx 170$ mK) is suppressed by annealing. The 7-day annealed sample of UCu_4Pd still shows an inkling of such a transition (≈ 140 mK), whereas UCu_4Pd 14-day annealed follows a logarithmic dependence down to the lowest measured temperature of 0.08 K. This logarithmic temperature dependence expands upon annealing to cover more than two decades as can be seen in the inset of Fig. 2(a).

To compare our specific heat data to the Griffiths phase model[3] that predicts a power-law behavior $C/T\propto T^{-1+\lambda}$ with $\lambda<1$, we have performed a careful χ^2 analysis of our data:

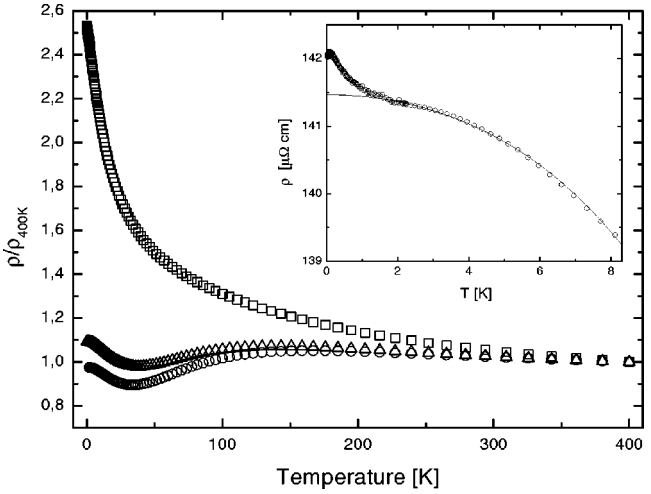

FIG. 3. The electrical resistivity $\rho/\rho_{400\ \mathrm{K}}$ as a function of temperature for UCu_4Pd unannealed (open circle), UCu_4Pd 7-day annealed (open up triangle), and UCu_4Pd 14-day annealed (open square). The qualitative run of the curve changes dramatically: Above 2 K the non-Fermi-liquid behavior $\rho-\rho_r \propto T$ disappears upon annealing. The inset shows an expanded plot of the resistivity of the 14-day annealed sample down to 30 mK. The solid line is a fit to our data in the region between 2 K and 8 K with a Fermi-liquid expansion $\rho-\rho_r=A\,T^2+B\,T^4$ ($\rho_r=141.5\ \mu\Omega$ cm; $A=-0.024\ \mu\Omega\,\mathrm{cm\,K^{-2}}$; $B=-0.000\,13\ \mu\Omega\,\mathrm{cm\,K^{-4}}$). The small upturn of the resistivity by about 0.5% below 1 K seen in the inset is so far not understood.

(i) A two-parameter logarithmic fit $a_1+a_2 \ln T$ leads to $\chi^2=9.0$ and $\chi^2=6.1$ for fits in the temperature regions $T<10$ K and $T<1$ K, respectively. (ii) A three-parameter power-law fit $a_1'+a_2' T^{-1+\lambda}$ converges to the *same* χ^2 values with the fit parameter $\lambda \to 1.0$ and a_1', a_2' large and with opposite signs.[11] One easily sees that this is effectively equivalent to our two-parameter logarithmic fit (i). From this statistical analysis we conclude that our specific heat data below 10 K has a clear logarithmic temperature behavior.

It is remarkable that increasing order in UCu_4Pd does not destroy its non-Fermi-liquid behavior in the specific heat, but rather expands the temperature range of its logarithmic behavior. Thus this work reports an alternative tuning parameter for non-Fermi-liquid behavior besides pressure,[12] doping,[2] or magnetic field:[13] crystallographic order.

The splat-cooled UCu_4Pd, as expected for the most disordered sample, shows, however, a completely different behavior. The temperature dependence is more like that of $UCu_{3.8}Pd_{1.2}$ (which has almost the same lattice parameter as our ''splat-cooled'' sample, see Fig. 1) as can be seen in Fig. 2(b).

The electrical resistivity shows a particularly strong sensitivity to annealing. Fig. 3 shows the electrical resistivity of the unannealed and the annealed samples. The residual resistivity drops strongly by a factor of about 2.5. Also an enormous change in the qualitative behavior of the curve can be observed. While the resistivity of the unannealed sample increases continuously below 400 K, the resistivity of the annealed samples has a Kondo-like minimum at about 35 K. In the temperature region between 2 K and 8 K our resistivity data for the 14-day annealed sample can be well described by a Fermi-liquid expansion $\rho-\rho_r=A\,T^2+B\,T^4$ (see the inset of Fig. 3). The small upturn of the resistivity by about 0.5% below 1 K with an inkling of a maximum at 70 mK seen also in the inset of Fig. 3 is so far not understood. It is also observed in the unannealed sample (not shown here). Except for this small upturn below 1 K we conclude that the behavior of the resistivity of the annealed sample is clearly no longer non-Fermi-liquid-like with $\rho-\rho_r \propto T$ below 10 K as first reported by Andraka and Stewart[2] for the unannealed case. In fact, the observed behavior of the resistivity below 10 K is neither consistent with Fermi-liquid nor with non-Fermi-liquid behavior with a unique power-law dependence. Also notice that the sign of A in our fit is unusual for f moments forming a Kondo lattice.

Furthermore, a strong indication for a change of the ground state in this system is the change from a negative magnetoresistance at 1.8 K of the unannealed sample to a positive magnetoresistance of the annealed samples (not shown). This indicates a transition from a spin-disordered compound to a ''normal'' metal.

Finally, we observe that also the spin glass behavior found in unannealed UCu_4Pd (Refs. 5 and 14) is sensitive upon annealing. The spin glass temperature T_{SG} determined from ac-susceptibility measurements shifts to lower temperature with decreasing frequency (95 Hz, 995 Hz) in the unannealed sample.[5] In the annealed samples we could still observe a maximum in the ac-susceptibility at 65 mK (7-day annealed) and at 55 mK (14-day annealed), but no temperature shift with frequency (not shown). This temperature shift of the maximum might be due to a suppression of the antiferromagnetic transition of the unannealed sample as recently discussed by Körner *et al.*,[9] supporting increasing order between the 7-day and 14-day annealed samples. Further work on the effects of annealing upon spin glass behavior is in progress.

IV. CONCLUSIONS

Summarizing, we have shown that in UCu_4Pd intrinsic order yields a new tuning parameter for non-Fermi-liquid behavior in the specific heat in addition to the usual parameters of doping, pressure, and magnetic field. Therefore samples must also be characterized by their intrinsic order: X-ray linewidth and lattice parameter measurements indicate a rearrangement of the Pd atoms from the 16*e* sites to the 4*c* sites in UCu_4Pd leading to increased sublattice order upon annealing.

Increasing order has a markedly different effect on the behavior of the specific heat and the resistivity: (i) A higher degree of crystallographic order suppresses the antiferromagnetic transition temperature T_N to <0.08 K. The best ordered sample then shows a logarithmic behavior in C/T over more than two decades in temperature from 0.08 K to 10 K. Therefore annealing expands the range of non-Fermi-liquid behavior in the specific heat.

(ii) On the other hand, the behavior of the resistivity in the annealed sample shows a T^2 dependence with higher-order corrections between 2 K and 8 K. In addition, the magne-

toresistance changes sign: The non-Fermi-liquid behavior ($\rho-\rho_r \propto T$) of the unannealed samples observed by Andraka and Stewart[2] in the same temperature range thus vanishes for increased crystallographic order.

Since the Kondo disorder model would predict non-Fermi-liquid behavior for the specific heat (logarithmic dependence of the Sommerfeld coefficient on temperature[7]) *and* for the resistivity[15] ($\rho-\rho_r \propto T$) in about the same temperature range, this model is therefore clearly not applicable in annealed UCu_4Pd. With respect to unannealed UCu_4Pd, it seems furthermore plausible from Fig. 2(a) to expect that the low-temperature behavior of its Sommerfeld coefficient is due to the same physical mechanism as in annealed UCu_4Pd. Thus, since disorder (at least as understood in the Kondo disorder model) is not the underlying mechanism in C/T for non-Fermi-liquid behavior in annealed UCu_4Pd as shown above, it seems possible that it is not the origin for non-Fermi-liquid behavior in unannealed UCu_4Pd either. With respect to the Griffiths phase model[3] we have found that our specific heat data are not consistent with the theoretically predicted power-law behavior $C/T \propto T^{-1+\lambda}$ with $\lambda<1$. Therefore these two theoretical models, which presuppose disorder for explaining the non-Fermi-liquid behavior in UCu_4Pd are not applicable. Our observations lead support to a quantum critical point scenario that might reconcile our specific heat, susceptibility and resistivity data. Further theoretical and experimental work, in particular muon spin resonance and NMR studies on the annealed samples, is in progress in order to establish such a scenario.

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