

# Tuning through the quantum critical point in $\text{UCu}_{5-x}\text{Ni}_x$ : Rapid variations in the specific heat

D. J. Mixson, J. S. Kim, M. Swick, T. Jones, and G. R. Stewart  
*Department of Physics, University of Florida, Gainesville, Florida 32611-8440, USA*

E.-W. Scheidt and W. Scherer  
*Fachbereich Physik, Universitaet Augsburg, 86159 Augsburg, Germany*

T. Murphy and E. C. Palm  
*National High Magnetic Field Laboratory, 1800 E. Paul Dirac Drive, Tallahassee, Florida 32310, USA*

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Using specific heat down to 0.05 K, we determine the variation of the non-Fermi liquid behavior in strongly disordered  $\text{UCu}_{5-x}\text{Ni}_x$ ,  $0.5 \leq x \leq 1.2$ , as the Ni doping is varied through the quantum critical concentration,  $x_{\text{QC}}$ , that suppresses the 16.5 K antiferromagnetism in pure  $\text{UCu}_5$ . Contrary to the case of Pd-doped  $\text{UCu}_5$ , where the Pd experiences partial sublattice ordering, Ni—which unlike Pd is *smaller* than Cu—goes equally on the four equivalent smaller Cu sites in  $\text{UCu}_5$  at a concentration of about 25% at  $x_{\text{QC}}=1$ . This ensures strong lattice disorder. This disorder is confirmed by large high angle x-ray linewidths and large measured residual resistivity values of  $\geq 700 \mu\Omega \text{ cm}$ .  $C/T \approx \log T$  over the whole temperature range of measurement in the well-disordered  $\text{UCu}_{5-x}\text{Ni}_x$  only within 5% of  $x_{\text{QC}}$ . By  $x=1.1$ , a temperature dependence for  $C/T$  sets in below 0.3 K that is more divergent than *either* the  $\log T$  or  $T^{-1+\lambda}$  fits to the higher temperature data, similar to behavior recently observed and analyzed in  $\text{YbRh}_2\text{Si}_2$ . This progression of the temperature dependence of the specific heat in  $\text{UCu}_{5-x}\text{Ni}_x$  around  $x_{\text{QC}} \approx 1$  as a function of  $x$  is rapid compared to other known doped non-Fermi liquid systems.

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## I. INTRODUCTION

Soon after the discovery<sup>1</sup> of unusual non-Fermi liquid (nFl) behavior in  $\text{U}_{0.2}\text{Y}_{0.8}\text{Pd}_3$  in 1991, it was recognized<sup>2</sup> that nFl behavior often occurs where a magnetic transition has been suppressed in a phase diagram. Typical nFl behavior is  $\rho \approx T$  or  $T^\alpha$ ,  $\alpha < 2$ , and  $\chi$  as well as  $C/T \approx \log T$  or  $T^{-1+\lambda}$ . The usual explanation<sup>3</sup> for such nFl behavior is that there is a second-order phase transition, e.g., antiferromagnetism, whose ordering temperature has been suppressed using doping, pressure, or magnetic field. Such a second-order phase transition at  $T=0$  creates a quantum critical point (QCP) that has a large influence on the measured properties at *finite* temperatures (see Ref. 3 for a review).

Most experimental studies of nFl behavior have used<sup>3</sup> doping, with its accompanying disorder, to reach the QCP. Due to the inherent complexity of disorder in a doped strongly correlated electron system, and the widely differing amounts of disorder introduced by the various dopings, the existing experimental status of, and the theoretical models for understanding, nFl behavior at a QCP reached by doping are still in the developmental stage. While pure, presumably well-ordered nFl systems often can have<sup>3</sup> residual resistivity ( $\rho_0$ ) values, which are a measure of order, lower than  $1 \mu\Omega \text{ cm}$ , doped nFl systems reported to date have<sup>3</sup>  $\rho_0$  values that vary between  $1.5 \mu\Omega \text{ cm}$  to over  $500 \mu\Omega \text{ cm}$ .

The present work presents the characterization of the bulk specific heat of a very disordered system ( $\rho_0 \geq 700 \mu\Omega \text{ cm}$ ) in the vicinity of its QCP, to investigate disorder and its effect on nFl behavior near a QCP and provide input about a well-disordered system to further theoretical work.

## A. Existing theories

There are numerous theories, still in flux,<sup>4,5</sup> that make at least qualitative predictions for a three-dimensional system near in phase space to where  $T_N \rightarrow 0$ .

Qualitatively (see Ref. 3 for a more complete discussion), a saturation of  $C/T$  as  $T \rightarrow 0$  would be better described by the Kondo disorder model<sup>6-8</sup> without a large number of spins remaining uncompensated at low temperatures.  $C/T \approx a - b\sqrt{T}$  as  $T \rightarrow 0$  would imply weakly coupled spin fluctuations in the Moriya theory.<sup>9</sup>  $C/T \approx \log T$  as  $T \rightarrow 0$  over more than a decade in temperature would be consistent with the Kondo disorder model in the “marginal case”<sup>8</sup> with remaining uncompensated spins at even lower temperature.  $C/T \approx T^{-1+\lambda}$  as  $T \rightarrow 0$  over more than a decade in temperature would be consistent with the Griffiths phase disorder model,<sup>4,10,11</sup> with the same power law temperature dependence included<sup>8</sup> in the Kondo disorder model as one limiting case. Measurements of  $C/T$  to typically less than 0.1 K are required<sup>3,12</sup> to distinguish between power law and logarithmic temperature dependence when the power law exponent is small, i.e., when  $\lambda$  is close to 1.

Going beyond these three models, there are systems<sup>3</sup> with apparently no disorder, or where disorder is found<sup>13</sup> by other measurement techniques not to dominate the measured behavior, where the Kondo disorder model is not applicable and still  $C/T \approx \log T$  as  $T \rightarrow 0$ . This  $\log T$  temperature dependence is, phenomenologically, found<sup>3</sup> in the majority of systems at their QCP. Thus, such a  $\log T$  dependence for  $C/T$  may be indicative of a universality class at a QCP that transcends the details of how (via doping, pressure, or magnetic field) the critical point was reached, or whether disorder is

present.<sup>14</sup> One of the questions of the present work is whether this  $\log T$  behavior in  $C/T$  is also present at the QCP of a strongly disordered system.

In addition, saturation in  $C/T$  at low temperatures need not have any complicated theoretical reason, but can just be due to the system's displaying Fermi liquid behavior because it has not been tuned precisely to a QCP.

Finally, there exist experimental results in, e.g.,  $\text{YbRh}_2\text{Si}_2$  (see Ref. 15) and  $\text{Y}_{1-x}\text{U}_x\text{Pd}_3$ ,<sup>3</sup> where  $C/T$  diverges as  $\log T$  down to  $\approx 0.3$  K and then, at still lower temperatures, diverges *more* rapidly than  $\log T$ . Thus, in contrast to the systems<sup>3</sup> that follow the Moriya model of  $C/T \approx \log T$  at higher temperatures followed by a crossover to a limiting, *less* divergent behavior as  $T \rightarrow 0$ , some systems follow  $C/T \approx \log T$  behavior succeeded by, at lower temperatures, a more divergent behavior. The results in  $\text{YbRh}_2\text{Si}_2$  have been recently theoretically addressed<sup>16</sup> as being possibly due to an excitation belonging to a new universality class. This new kind of QCP physics, where  $C/T$  diverges at low temperatures faster than either  $\log T$  or a comparable<sup>12</sup> weak ( $\lambda$  close to 1) power law,  $T^{-1+\lambda}$ , is predicted<sup>16</sup> to occur "close" to a QCP and is dependent on the relative strengths of the antiferromagnetic and Kondo interactions.

### B. Current experimental status of the effect of disorder at a QCP

$\text{UCu}_{5-x}\text{Pd}_x$  is a very well studied system in which nFl properties have been achieved by doping to suppress  $T_N \rightarrow 0$  and where the influence of disorder has been studied. A range of different measured properties has been studied in this system as a function of small variations in doping around the quantum critical concentration  $x_{\text{QC}}$ .  $\text{UCu}_4\text{Pd}$  is putatively at (or at least very near) a quantum critical point, since  $T_N$  is suppressed to  $T=0$  in  $\text{UCu}_{5-x}\text{Pd}_x$  at  $x \approx 1$ . Even in this well studied system, however, the relative roles of the quantum criticality versus that of disorder in determining the nFl behavior have not been clearly resolved. The reason for this uncertainty hinges on the question of preferential sublattice ordering of the larger Pd atoms on the larger Be 1 site in the cubic  $\text{AuBe}_5$  structure. Work by Booth *et al.*,<sup>17</sup> using x-ray absorption fine structure (XAFS) studies of unannealed and 14 day at 750 °C annealed  $\text{UCu}_4\text{Pd}$  samples shows an increase of the Pd occupancy of the larger Be 1 site from  $\approx 73\%$  in the unannealed sample to  $\approx 81\%$  after annealing. This result implied improved preferential sublattice ordering of the Pd atom but still with disorder present. Weber *et al.*<sup>18</sup> showed that the effect of improved ordering on the nFl behavior in  $C/T$  and  $\rho$  in the annealed  $\text{UCu}_4\text{Pd}$  is substantial. Specifically, Weber *et al.*<sup>18</sup> found a change of 20% in the magnitude of  $C/T$  at 0.1 K, a factor of 7 decrease (from<sup>3</sup> 375 to 50  $\mu\Omega$  cm) in  $\rho_0$ , and changed temperature dependences at low temperature for both  $C/T$  and  $\rho$ . In fact, annealing even affects where  $T_N \rightarrow 0$  as a function of  $x$  in  $\text{UCu}_{5-x}\text{Pd}_x$ ;  $x$  for  $T_N=0$  changes<sup>19</sup> from  $x_{\text{QC}}=1.1$  to  $x_{\text{QC}}=1.0$  upon annealing. Despite these clear signs of the effect of improved order on  $C/T$  and  $\rho$  in the data<sup>18</sup> of Weber *et al.* on annealed  $\text{UCu}_4\text{Pd}$ , MacLaughlin *et al.*<sup>13</sup> still find—consistent with the XAFS results—clear signs of disorder in

their muon relaxation experiments on annealed  $\text{UCu}_4\text{Pd}$ . MacLaughlin *et al.*<sup>13</sup> conclude that the nFl behavior in the muon relaxation response in this material is due to disorder.

In order to avoid the partial sublattice order in  $\text{UCu}_4\text{Pd}$  and to study nFl behavior at  $x_{\text{QC}}$  in a thoroughly disordered system where doping suppresses  $T_N$ , we have carried out the present work. The system chosen is, in the sense of the question of order versus disorder, rather at the opposite extreme from the  $\text{UCu}_4\text{Pd}$  case; there is guaranteed strong lattice site disorder where  $T_N \rightarrow 0$ . In order to build on the thorough studies in  $\text{UCu}_{5-x}\text{Pd}_x$ , the system chosen is  $\text{UCu}_{5-x}\text{Ni}_x$ , where Ni is smaller than Cu. Since Ni is isoelectronic to Pd, and since the suppression of the antiferromagnetism in  $\text{UCu}_5$  is a hybridization effect between the  $Uf$  electrons and the ligand  $d$  electrons, the expectation (born out by the results reported below) is that  $T_N$  is depressed to  $T=0$  (creating a quantum critical point) at around the same  $x \approx 1$  concentration for Ni as reported for the  $\text{UCu}_{5-x}\text{Pd}_x$  system. In contrast to the better than 80% occupation of the larger Be 1 sublattice site by the larger Pd atom reported<sup>17</sup> by Booth *et al.* in annealed  $\text{UCu}_4\text{Pd}$  (with statistically less than 5% Pd occupation of each of the four smaller Be 2 sites), the smaller Ni in  $\text{UCu}_4\text{Ni}$  will be distributed statistically at slightly less than the 25% level on the four smaller Be 2 sublattice sites with some occupation of the one larger Be 1 site. This will guarantee significant disorder (as will be clear from the x-ray and residual resistivity results discussed below) at the 4:1 Cu:Ni concentration, as well as at neighboring concentrations. As  $x$  is varied through  $x_{\text{QC}} \approx 1$ , the disorder will change continuously in  $\text{UCu}_{5-x}\text{Ni}_x$ . Any nonmonotonic behavior in the nFl behavior will not be due to nonmonotonic changes in the disorder. A very fine variation in dopant concentration is used for this work (for previous work on  $\text{UCu}_{5-x}\text{Ni}_x$ ,  $x=0.5, 0.8, 1.0, \text{ and } 1.5$ , see Refs. 20–23), so that results essentially at the critical concentration  $x_{\text{QC}}$  where  $T_N \rightarrow 0$  can be compared to samples away from the critical concentration. This care is necessary because of the possibility of sensitivity of the properties at the quantum critical point to small variations away from the critical concentration.

As a companion effort to the present work, we have measured and reported elsewhere<sup>24</sup> the resistivity and magnetic susceptibility as a function of temperature as well as the magnetization as a function of field for the  $\text{UCu}_{5-x}\text{Ni}_x$  system. The question of the relative influence of disorder near  $x_{\text{QC}}$  on the non-Fermi liquid behavior could not be clearly determined in this companion study<sup>24</sup> of  $\rho$  and  $\chi$ .

This is because, *firstly*, the resistivity of the disordered  $\text{UCu}_{5-x}\text{Ni}_x$  samples (just as recently concluded<sup>25</sup> for unannealed, partially disordered  $\text{UCu}_4\text{Pd}$ ) was found to be mostly unconnected with the intrinsic, highly correlated electron behavior. Instead, the resistivity was highly dependent on disorder effects in the lattice and at the grain boundaries. Thus, the  $\rho = \rho_0 - AT$  nFl behavior measured<sup>24</sup> in  $\text{UCu}_{5-x}\text{Ni}_x$  between 2 and 15 K comprises only about 10% of the magnitude of  $\rho$  at low temperatures, which includes a 30% temperature-independent term (i.e., of order 200  $\mu\Omega$  cm) possibly due<sup>24</sup> to grain boundaries and/or microcracks. In addition, the  $\rho \sim \log T$  behavior (possibly due<sup>24</sup> to the Kondo effect or localization effects) below 2 K down to where  $\rho$  saturates at  $\sim 0.1$  K comprises only about 3% of the magni-

TABLE I. Parameters for  $\text{UCu}_{5-x}\text{Ni}_x$ <sup>a</sup>

$x$	$a_0$ (Å)		$T_N$ (K) <sup>b</sup>	$\lambda$ in $C/T \approx T^{-1+\lambda}$
	unann.	ann. two weeks		
0.5	7.0089	7.0101	6.1	
0.6	7.0176	7.0022	3.8	
0.75	6.9982	6.9905	1.9	
0.8	6.9935	6.9927	1.1	
0.9	6.9836	6.9871	0.43	0.63
1.0	6.9828	6.9825	<0.05	
1.05	6.9797	6.9788		0.74
1.1	6.9791	6.9761		
1.2	6.9711	6.9728		

<sup>a</sup>All sample properties presented in this work were measured on material annealed two weeks at 750 °C, except for the unannealed lattice parameter data.

<sup>b</sup> $T_N$  was primarily determined by the plateau in  $C/T$ , as discussed in the text. However, for  $x=0.5$  and  $0.6$  (i.e., where  $T_N > 2$  K, the lowest temperature of measurement of the Quantum Design MPMS system) this “plateau method” determination of the magnetic ordering temperature was confirmed both by dc and ac magnetic susceptibility data (see Ref. 24).

tude of the low- temperature  $\rho$  value. Clearly, extrinsic effects may dominate<sup>24</sup> the low- temperature resistivity. Indeed, pieces of the *same* sample of  $\text{UCu}_{5-x}\text{Ni}_x$  showed<sup>24</sup> 20% variation in the value of  $\rho_0$ , with values for  $x \geq 0.8$  in the range of 700–1100  $\mu\Omega$  cm.

*Secondly*, the temperature dependence of the magnetic susceptibility in  $\text{UCu}_{5-x}\text{Ni}_x$  below 10 K was difficult to determine due to the influence of a broad, field-dependent maximum in  $\chi$  centered at around 25 K.

## II. EXPERIMENTAL DETAILS

Samples of  $\text{UCu}_{5-x}\text{Ni}_x$ ,  $x=0.5, 0.6, 0.75, 0.8, 0.9, 1.0, 1.05, 1.1,$  and  $1.2$  were prepared by arc-melting the constituent elements (depleted U from Los Alamos National Laboratory, 99.9+ % purity, Ni and Cu from Johnson Matthey Aesar, 99.999% purity) under a purified Ar atmosphere. To check if there was any unexpected dependence of properties on annealing, pieces of each sample were wrapped in Ta foil and sealed in evacuated quartz tubes for annealing for two weeks at 750 °C (just as done<sup>18</sup> in the annealing studies of  $\text{UCu}_{5-x}\text{Pd}_x$ ). Samples were x-rayed using powder diffractometry in the University of Florida Major Analysis Instrumentation Center. Specific heat was measured down to 0.3 K in a <sup>3</sup>He cryostat, and separately in a dilution refrigerator from Oxford Instruments down to 0.050 K at NHMFL, Tallahassee or at the University of Augsburg, Germany using established<sup>26</sup> time-constant measurement techniques.

## III. RESULTS

Upon adding Ni to  $\text{UCu}_5$ , the cubic lattice parameter of the annealed samples decreases linearly (Vegard’s law) with increasing Ni concentration (see Table I). No consistent

change between  $x=0.8$  and  $1.2$  in  $a_0$  or in high angle line-width with annealing for two weeks at 750 °C is evident; i.e., in contrast to the behavior observed<sup>18</sup> in  $\text{UCu}_{5-x}\text{Pd}_x$ . Although, as will be seen in the discussion below, there are slight ( $\sim 5\%$ ) changes in the specific heat with annealing, there is no evidence that annealing at 750 °C for two weeks affects the order of the sample. This is evidenced either through changes in the lattice parameter (see Table I) or through measurement of the width of the high angle x-ray diffraction lines. For example, the [731] line centered at approximately  $115.9^\circ 2\Theta$  for the  $\text{UCu}_4\text{Ni}$  sample shows the same line width ( $\sim 0.72^\circ 2\Theta$  full width at half-maximum, versus an instrumental broadening of less than  $0.01^\circ 2\Theta$ ) for both unannealed and annealed samples. This is in contrast to the observed<sup>18</sup> sharpening of even a relatively low angle diffraction line (at  $42.4^\circ 2\Theta$ ) in  $\text{UCu}_4\text{Pd}$  under the same annealing treatment by 20%.

The suppression of  $T_N$  in  $\text{UCu}_{5-x}\text{Ni}_x$  from 16.5 K at  $x=0$  with increasing  $x$ , determined by specific heat data (and by<sup>24</sup> ac and dc magnetic susceptibility for  $T_N > 2$  K, i.e., up to  $x=0.6$ ), is shown in Table I. The antiferromagnetic ordering temperature as determined by peaks in  $\chi_{ac}$  (whole temperature range) and  $\chi_{dc}$  (for  $T > 2$  K) corresponds to plateaus in  $C/T$  in numerous systems when  $T_N$  has been suppressed below  $\sim 1/2$  of the undoped  $T_N$ , not the normal peak in  $C/T$  observed in pure systems. Systems in which this plateau behavior of  $C/T$  at the (depressed) ordering temperature has been observed include  $\text{CeCu}_{6-x}\text{Ag}_x$ ,<sup>27</sup>  $\text{CeCu}_{6-x}\text{Au}_x$ ,<sup>28</sup> and—most importantly for the present work—the analog system  $\text{UCu}_{5-x}\text{Pd}_x$ ,<sup>19</sup> down to 0.2 K.<sup>19</sup> In each case,  $T_N$  determined by the peak in the magnetic susceptibility corresponds within a few percent to the midpoint of the beginning of the plateau in  $C/T$ ; see, e.g., the arrows in the plot (discussed below) of  $C/T$  versus  $\log T$  for  $\text{UCu}_{5-x}\text{Ni}_x$  shown in Fig. 1. The ordering temperature for  $\text{UCu}_{5-x}\text{Ni}_x$  falls somewhat more rapidly with initial doping than in<sup>19</sup>  $\text{UCu}_{5-x}\text{Pd}_x$  but, as  $T_N \rightarrow 0$ , the two sets of data become quite similar. For example, at  $x=0.9$ ,  $T_N=0.53$  K for<sup>19</sup> Pd doping, and 0.43 K in the present work for Ni doping, both determined from specific heat measurements.

### A. Specific heat

The specific heat divided by temperature ( $C/T$ ) versus  $\log T$  is shown in Fig. 1 for  $\text{UCu}_{5-x}\text{Ni}_x$ ,  $0.6 \leq x \leq 1.2$ , from 0.3 to 10 K. As the antiferromagnetic transition temperature is suppressed with increasing Ni doping, the upturn in  $C/T$  above  $T_N$  is extended to lower and lower temperatures, giving a large magnitude to the value of  $C/T$  as  $T \rightarrow 0$ . After  $T_N$  is suppressed for  $x > 0.9$  (see Table I), Fig. 1 shows that this rather large value of  $C/T$  at low temperatures then falls monotonically with increasing  $x$ , as does<sup>24</sup> the value of the magnetic susceptibility,  $\chi$ , at low temperature. This is because the magnetic fluctuations and associated degrees of freedom related to the antiferromagnetic ordering—which contribute to the  $f$ -electron part of the entropy,  $S = \int (C/T) dT$ , at low temperatures—are depressed as  $T_N \rightarrow 0$ .

In this plot of the specific heat divided by temperature ( $C/T$ ) versus  $\log T$ , although it is tempting to fit the  $C/T$  data

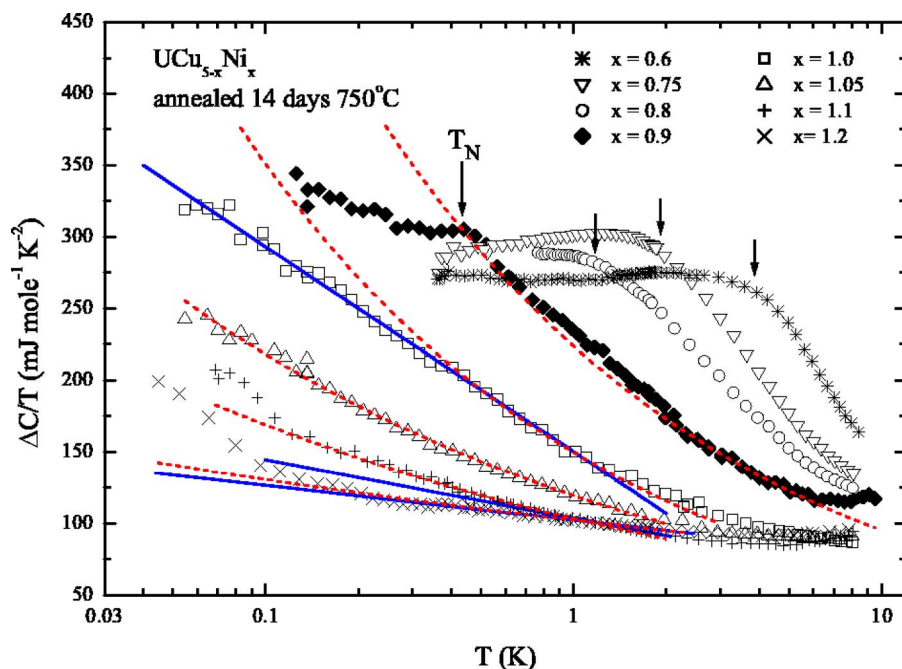


FIG. 1. (Color online)  $\Delta C/T$  (where  $C/T$  is corrected for the lattice contribution for  $\text{UNi}_5$  from Ref. 20), for  $\text{UCu}_{5-x}\text{Ni}_x$ ,  $x=0.6-1.2$ , annealed for two weeks at  $750^\circ\text{C}$  vs  $\log T$ .  $C/T$  for unannealed,  $x=1.0$ , is about 5% less than for the annealed data shown here at 0.4 K, while for  $x=1.1$  the unannealed data are about 5% larger than for the annealed sample. The straight solid line (—) fit to the  $x=1.0$  data over more than a decade of temperature between 0.05 and 1 K shows that  $C/T \approx \log T$  only at this one composition. The  $x=1.05$  data fit neither the Moriya theory nor  $\log T$  behavior, but, as shown by the dashed line (---) fit power law behavior over more than a decade in temperature. Note the rapid divergence above both the  $\log T$  and the  $T^{-1+\lambda}$  extrapolations, shown as a solid line and as a dashed line, respectively, of the data above 0.3 K at low temperature of the data for  $x=1.1$  and 1.2.

for  $x=0.8$  and 0.9 (which tend to saturate at low temperatures) to the Moriya theory, these fits are necessarily limited in temperature range to above  $T_N$ . Indeed, Moriya fits (not shown) to  $x=0.8$  and 0.9 yield a parameter  $y_0$  that decreases towards 0 (0.065 and 0.011, respectively); i.e., consistent with a quantum critical point, as  $x \rightarrow 1.0$ . However, even for  $x=0.9$  the fit is only between 0.6 and 3 K; as will be discussed below, the data for  $x=0.9$  can be fit over a wider temperature range to a power law temperature dependence. The data shown in Fig. 1 for  $x=1.0, 1.05, 1.1$ , and 1.2 do not fit the Moriya theory since below 1 K the data for  $x=1.0$  continue to rise as  $\log T$  and the data for  $x>1$  rise more quickly than a  $\log T$  rise.

Considering further the temperature dependence of the specific heat data, the  $C/T$  versus  $\log T$  data for  $x=0.6-1.2$  shown in Fig. 1 show that *only* the data for  $x=1.0$  obey  $C/T \approx \log T$  over at least a decade in temperature.<sup>29</sup> This  $\log T$  behavior observed down to 0.05 K is inconsistent with the Griffiths phase model but consistent (at least *a priori*) with a number of models, including the Kondo disorder model,<sup>6-8</sup> predictions<sup>3</sup> for a two-dimensional quantum critical point, or a system with nested Fermi surfaces.<sup>14</sup> There are, of course, problems, as discussed generally above in Sec. I, with any of these models being applied to the present  $\text{UCu}_{5-x}\text{Ni}_x$  system under discussion. For the Kondo effect, there is the question<sup>24</sup> as to whether such a large resistivity allows even the discussion of the Kondo effect which, however, has been used<sup>6</sup> in the literature to describe the specific heat behavior in the  $\sim 400 \mu\Omega \text{ cm}$   $\text{UCu}_4\text{Pd}$  system. Further,

it is unclear that  $\text{UCu}_{5-x}\text{Ni}_x$  can be considered either as having nested Fermi surfaces or being two-dimensional. As stated in the introduction, experimentally  $C/T \approx \log T$  has been found<sup>3</sup> in numerous three-dimensional systems (ordered and disordered) at their quantum critical point, a possible universality class that is outside current theoretical understanding. Work is under way<sup>30</sup> to measure other properties of  $\text{UCu}_4\text{Ni}$ , including NMR, in order to check the applicability of the Kondo disorder model.

Considering now fits of the  $C/T$  data in Fig. 1 to a power law behavior,  $T^{-1+\lambda}$ , the dashed line fits shown for this temperature dependence show that the data for  $x=1.05$  obey  $C/T \approx T^{-1+\lambda}$  from 2 K down to 0.05 K (the lowest temperature of measurement); i.e., well over a decade in temperature. In addition, the data for  $x=0.9$  can be fit to a power law temperature dependence between 6 and 0.5 K, below which the remanent antiferromagnetic transition at this composition occurs. As seen visually in Fig. 1,  $C/T$  for  $x=1.0$  clearly fits  $\log T$  (the straight line) better than a power law (dashed line), while for  $x=1.05$   $C/T$  is better fit by a power law behavior. These visual impressions of the applicability of the fits are confirmed numerically by considering the chi-square deviations for the fits. For  $x=1.0$ , the  $\log T$  fit for the  $C/T$  data gives a factor of 3.7 smaller chi-square deviation than a power law fit to the same points, while for  $x=1.05$  the power law fit to the  $C/T$  data has a chi-square deviation a factor of 2.3 smaller than a fit of the same data to  $\log T$ . Note again that the degree of disorder for  $x=0.9$  or 1.05 in  $\text{UCu}_{5-x}\text{Ni}_x$  is very similar to that for  $x=1.0$ .

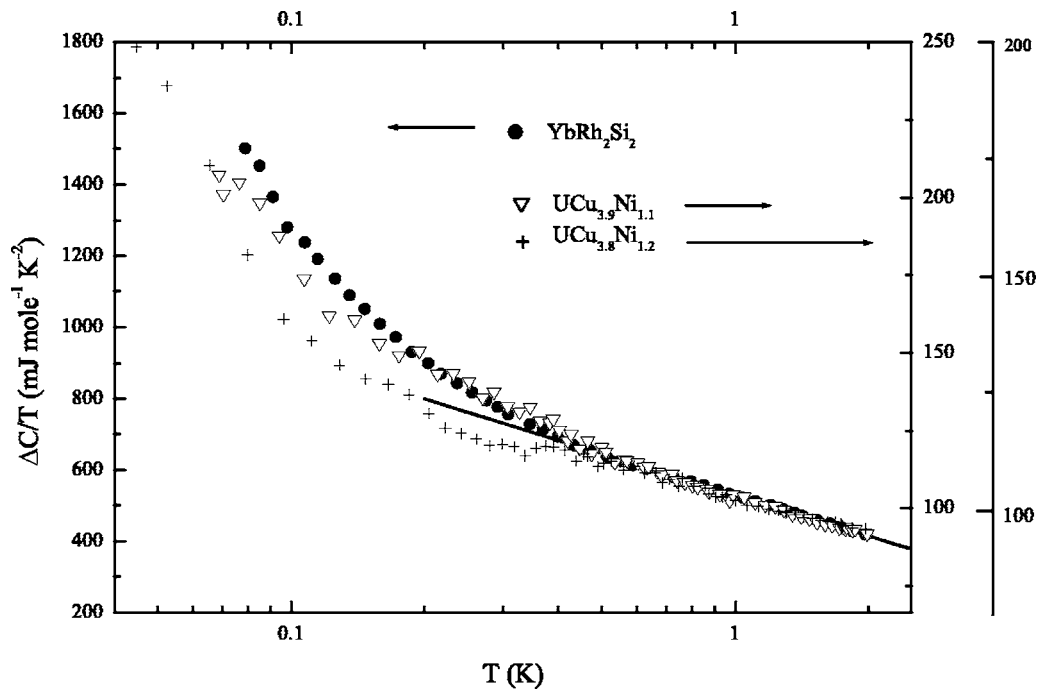


FIG. 2.  $\Delta C/T$  for  $\text{UCu}_{5-x}\text{Ni}_x$ ,  $x=1.1$  and  $1.2$ , plotted using the right hand axes and for (see Ref. 15)  $\text{YbRh}_2\text{Si}_2$  plotted using the left hand axis versus  $\log T$ , with the straight line serving as a guide to the eye to the  $\log T$  temperature dependence common to all three sets of data above about  $0.3$  K. The  $\text{UCu}_{5-x}\text{Ni}_x$   $C/T$  data have had their vertical zeros shifted and the  $y$  axes scaled so that the regions of  $C/T \approx \log T$  for all three materials overlap in the figure shown here. The upturns in  $\Delta C/T$  below  $0.3$  K for all three samples have similar temperature dependences.

Thus, both  $x=0.9$  and  $1.05$  samples have  $C/T \approx T^{-1+\lambda}$  over more than a decade in temperature, consistent with either the Griffiths phase disorder model or with the Kondo disorder model. Measurements of the specific heat as a function of field<sup>24</sup> (not shown) up to  $9$  T were made to check for the particular field and temperature dependence of the field-induced peak in  $C/T$  predicted<sup>11</sup> by the Griffiths phase rare spin cluster model, and seen in  $C/T$  field data for  $\text{Ce}_{1-x}\text{La}_x\text{RhIn}_5$ <sup>31</sup> and  $\text{Ce}_{1-x}\text{Th}_x\text{RhSb}$ .<sup>32</sup> The predicted Griffiths phase form<sup>11</sup> was *not* observed<sup>24</sup> either for the  $x=0.9$  or the  $x=1.05$  sample of  $\text{UCu}_{5-x}\text{Ni}_x$ .

This sequence of rapidly changing nFI temperature dependences for  $C/T$  for fine gradations of  $x$  near  $x_{\text{QC}}$  in the present work where strong disorder is present is an interesting result of the current work. Previous work<sup>3</sup> on doped nFI systems has not observed the same rapid evolution. For example, in the case of the antiferromagnet  $\text{UPd}_2\text{Al}_3$ ,  $T_N = 14.6$  K, Maple *et al.*<sup>33</sup> report that  $x_{\text{QC}} \approx 0.3$ , and that  $C/T \approx \log T$  for  $x=0.4, 0.6$ , and  $0.8$ . For unannealed  $\text{UCu}_{5-x}\text{Pd}_x$ , where  $x_{\text{QC}} \approx 1.1$ ,  $C/T$  follows  $\log T$  down to the lowest temperature of measurement only for  $x_{\text{QC}}$ , but follows a  $\log T$  dependence between  $10$  and  $0.5$  K up to  $x=1.5$ , with saturation behavior at lower temperature.<sup>34</sup> It is, of course, the case that not all doped systems necessarily have their properties dominated by disorder. For example,  $\mu\text{SR}$  and NMR measurements have been used by MacLaughlin *et al.*<sup>35</sup> to determine that systems such as  $\text{CeCu}_{5.9}\text{Au}_{0.1}$  and  $\text{Ce}(\text{Ru}_{0.5}\text{Rh}_{0.5})_2\text{Si}_2$  do not have disorder playing an essential role in their nFI behavior.

In the case of  $\text{UCu}_{5-x}\text{Ni}_x$ , at present we can only speculate as to the cause for the rapid variation of the temperature

dependence of  $C/T$  with composition. Experiments on other strongly disordered nFI systems near their QCP are planned to investigate whether the primary cause is due to the strong disorder that is present, or due to another effect, such as perhaps some unusual physics generated by the specifics of the changing ligand/ $f$ -ion hybridization as  $x$  is varied through  $x_{\text{QC}}$  in  $\text{UCu}_{5-x}\text{Ni}_x$ . In any case, this rapid changing of the nFI temperature dependence of  $C/T$  cannot be due to any non-monotonicity of the disorder as  $x$  is varied through  $x_{\text{QC}}=1$ .

As a last interesting feature, note how the  $C/T$  data shown in Fig. 1 for  $x=1.1$ , which fit a power law between about  $0.15$  and  $1$  K and a  $\log T$  dependence not at all, show at lowest temperature already upwards curvature. This upwards curvature means that  $C/T$  rises even faster than either the  $\log T$  or the power law dependence that fits the data at higher temperature. By  $x=1.2$ , the data shown in Fig. 1, which are well fit neither by a power law nor a  $\log T$  dependence, show that this extraordinary upturn in  $C/T$  continues to grow. When the temperature dependence of these two upturns plotted versus  $\log T$  in Fig. 1 is compared in Fig. 2 to the upturn in  $C/T$  above  $\log T$  behavior below  $\approx 0.3$  K in  $\text{YbRh}_2\text{Si}_2$ , the exponents derived in the present work are both within 10% of that reported,<sup>15</sup>  $C/T \approx T^{-0.3}$ , for  $\text{YbRh}_2\text{Si}_2$ . Thus, the temperature dependence of  $C/T$  for  $\text{UCu}_{5-x}\text{Ni}_x$ ,  $x=1.1$  and  $1.2$ , below  $0.3$  K is at least consistent with the new kind<sup>36</sup> of physics recently discussed<sup>16</sup> for  $\text{YbRh}_2\text{Si}_2$  and also qualitatively observed<sup>3</sup> to be similar to results in  $\text{Y}_{0.8}\text{U}_{0.2}\text{Pd}_3$  and  $\text{Y}_{0.9}\text{U}_{0.1}\text{Pd}_3$ .  $\text{YbRh}_2\text{Si}_2$  is well ordered, with  $\rho_0 \approx 2 \mu\Omega$  cm, while  $\text{Y}_{1-x}\text{U}_x\text{Pd}_3$ ,  $x=0.1$  and  $0.2$ , appears<sup>37</sup> to be a typical disordered system, with<sup>3</sup>  $\rho_0 \approx 60-300 \mu\Omega$  cm. Work is under way to search for this more divergent behavior in  $C/T$

below 0.3 K in other systems to try to find a common link between these apparently quite different systems.

#### IV. CONCLUSIONS

In summary, as the Ni concentration is tuned through  $x_{QC}$  in  $UCu_{5-x}Ni_x$ , the temperature dependence of the specific heat is dominated at  $x=x_{QC}$  by the quantum critical fluctuations just as experimentally often seen in both well-ordered and disordered systems where<sup>3</sup>  $C/T \approx \log T$  is found at  $x_{QC}$ . As  $x$  is increased above  $x_{QC}$ , the temperature dependence passes through a narrow compositional range where  $C/T$  be-

comes as a power law over more than a decade in temperature. At slightly higher Ni concentrations,  $C/T$  rather quickly attains an even faster divergence with decreasing temperature previously seen only in a limited number of systems, e.g.,  $YbRh_2Si_2$  and  $Y_{1-x}U_xPd_3$ .

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