

Magnetism, spin fluctuations, and non-Fermi-liquid behavior in $(U_xLa_{1-x})_2Zn_{17}$ G.-F. von Blanckenhagen,¹ E.-W. Scheidt,¹ T. Schreiner,¹ and G. R. Stewart²¹*Institut für Physik, Universität Augsburg, Universitätsstrasse 1, 86159 Augsburg, Germany*²*University of Florida, Department Of Physics, Gainesville, Florida 32611-8440*

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We present results of the low-temperature specific heat C of samples of the series $(U_xLa_{1-x})_2Zn_{17}$, combined with measurements of the low-temperature magnetic susceptibility (χ) and resistivity (ρ). For $x > 0.8$ we find antiferromagnetic order in coexistence with heavy-fermion behavior. An extrapolation of T_N as a function of the uranium concentration implies that T_N vanishes for $x = 0.8$; at $x = 0.8$, no magnetic order is detected experimentally at temperatures above 0.06 K. The non-Fermi-liquid (NFL) behavior predicted at such a point in the magnetic phase diagram may be observed, but not as clearly as in other systems; some of the behavior is more consistent with spin fluctuations. As the uranium concentration is lowered below $x = 0.8$, C continues to rise in the low-temperature limit, while $\chi \propto \chi_0 - aT^{0.5}$, but C seems to tend towards the behavior of a Fermi liquid with spin fluctuations at the lowest temperatures ($T < 0.25$ K). First at $x = 0.3$ the temperature dependence of C/T is found to be contrary to Fermi-liquid behavior, while $\chi \propto \chi_0 - a \log T$. Thus non-Fermi-liquid behavior is not found so unambiguously at the concentration where T_N vanishes as expected by a quantum critical point theory, but rather at lower uranium concentrations. This presents the possibility that NFL behavior in $(U_xLa_{1-x})_2Zn_{17}$ is *not* due to nearness to a quantum critical point, but rather to disorder or the presence of spin fluctuations.

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INTRODUCTION

The low-temperature properties of f -electron systems have been a wide and active field of research for many years. Particularly the ground-state properties of the so-called heavy-fermion systems have been studied intensively. For these systems different ground states have been reported, including antiferromagnetic order, superconductivity, and a paramagnetic ground state (for an overview, see Refs. 1 and 2).

Recently, the observation of non-Fermi-liquid behavior in some f -electron alloys—for example, $U_{0.2}Y_{0.8}Pd_3$ (Refs. 3 and 4) and $CeCu_{5.9}Au_{0.1}$ (Ref. 5)—has raised the question of whether a non-Fermi-liquid ground state exists in these materials. What characterizes a system as a non-Fermi liquid is not unambiguous. At least a nonconstant C/T (for example, $C/T \propto \log T$) and deviations from the Fermi-liquid behavior for χ and ρ have to be seen (for an overview, see Refs. 6 and 7). To explain the non-Fermi-liquid behavior in f -electron compounds, mainly three interpretations have been invoked.^{6,7} First, there are unconventional single-ion Kondo models like the two-channel Kondo effect⁸ and the quadrupolar Kondo effect.⁹ A second model, the Kondo disorder model, assumes a distribution of Kondo temperatures in disordered metals yielding a non-Fermi-liquid behavior at low temperatures.^{10,11} Recent results of NMR studies support this as a possible interpretation for the non-Fermi-liquid behavior in $UCu_{3.5}Pd_{1.5}$.¹² The third interpretation relates the non-Fermi-liquid behavior to a $T = 0$ K phase transition.⁴ For $CeCu_{5.9}Au_{0.1}$ (Ref. 5) and $CeCu_{6-x}Ag_x$ (Ref. 13), the low-temperature properties are interpreted by the existence of a quantum phase transition due to the suppression of long-range antiferromagnetic order. The influence of the $T = 0$ K quantum phase transition on the specific heat at finite temperatures has been calculated. One finds that for an antifer-

romagnetic quantum phase transition $C/T \propto T^{0.5}$ at the lowest temperatures.^{14–16} Non-Fermi-liquid behavior has also been reported in nearness to a ferromagnetic quantum phase transition in $Th_{1-x}U_xCu_2Si_2$,¹⁷ a scenario which is well known in d metals (for example, see Ref. 18).

U_2Zn_{17} was one of the first f -electron systems that was characterized as an antiferromagnetic heavy-fermion system.¹⁹ The electronic part of the specific heat divided by temperature was high both above and below the Néel temperature of $T_N = 9.7$ K. Values of about 500 mJ/U mol K² for temperatures just above the phase transition and 200 mJ/U mol K² for $T \rightarrow 0$ K have been reported.¹⁹ Magnetization and resistivity results of alloying experiments on the uranium sites with lanthanum $(U_xLa_{1-x})_2Zn_{17}$ for $x = 0, 0.05, 0.30,$ and 0.9 have previously been published.²⁰ The magnetic order vanishes with decreasing uranium concentration, and for diluted alloys ($x < 0.1$) the data are explained²⁰ by the existence of the Kondo effect. Thus, investigating the low-temperature specific heat of samples of the series $(U_xLa_{1-x})_2Zn_{17}$ allows us to study both the interplay of magnetism and heavy-fermion behavior in magnetically ordered alloys and, for the nonmagnetically ordered samples, to investigate the observed deviations of the specific heat from Fermi-liquid behavior, which will be discussed in regard to the ongoing discussion of non-Fermi-liquid behavior.

EXPERIMENT

Polycrystalline samples of $(U_xLa_{1-x})_2Zn_{17}$ were prepared. Stoichiometric amounts of the constituting element were placed in an outgassed BeO crucible covered with a lid. This crucible was then sealed under argon in a tantalum or a quartz glass tube. The sealed tubes were then heated to 1050 °C, e.g., above the melting point of U_2Zn_{17} , then slowly (within 3–10 h) cooled down to 750 °C. After annealing

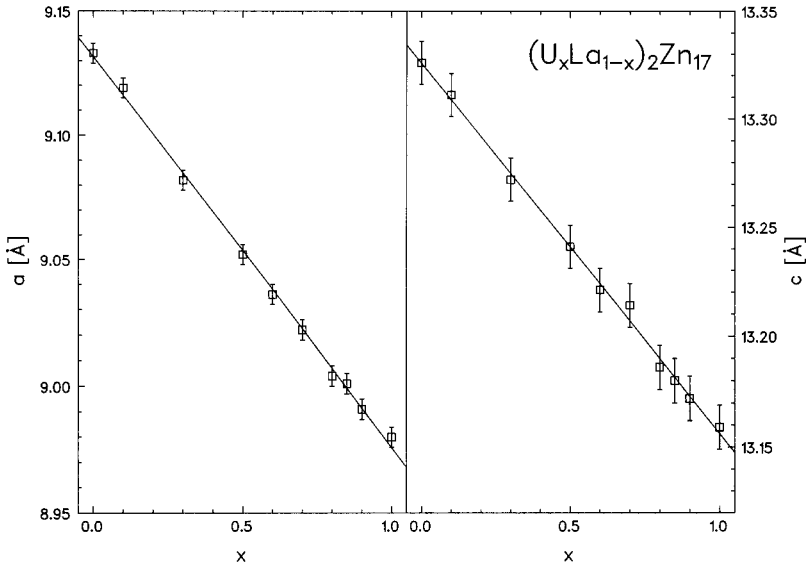


FIG. 1. Lattice parameters (given for the hexagonal setting of this rhombohedral structure) as a function of the U concentration x . Both the values of a and c vary linearly with x .

at this temperature for about 24 h, the tubes were removed from the furnace.

For x-ray diffraction we used a Siemens D5000 diffractometer. Magnetic measurements were performed with a Quantum Design superconducting quantum interference device (SQUID) magnetometer. The resistivity measurements were performed using a four-contact low-frequency ac method.

We measured the specific heat with a relaxation method^{21,22} in the temperature range 0.06–20 K. The absolute error of the specific heat is $\pm 3\%$.

RESULTS

X-ray diffraction patterns of the prepared samples show that they crystallize in the rhombohedral $\text{Th}_2\text{Zn}_{17}$ structure. Only the samples with $x=0.1$ show a weak line which could not be indexed on the basis of this structure. Our results are in agreement with the more recently published literature in that the $\text{Th}_2\text{Zn}_{17}$ structure is the one which forms out of the melt.^{23,24} Preliminary results from our group show that one obtains samples in the hexagonal $\text{Th}_2\text{Ni}_{17}$ structure via solid-state reaction of the constituent elements at about 800 °C.²⁵

The lattice parameters (given for the hexagonal setting) are plotted in Fig. 1 as a function of the uranium concentration x . Both lattice parameters vary linearly with x . This indicates the validity of Vegard's rule and supports the view that the main effect on the lattice of the alloying process is the size difference of La and U atoms. Thus, as seen in Fig. 1 (see also Table I), doping U_2Zn_{17} with La results in a quite significant (about 1.5%) lattice expansion.

In the following we first present the data of the specific-heat measurements. Of particular interest here is the electronic part of the specific heat C_{el} and C_{el}/T . To obtain C_{el}/T , the electronic part of the specific heat divided by temperature, we prepared and measured $\text{La}_2\text{Zn}_{17}$ and subtracted the lattice part of the specific heat of $\text{La}_2\text{Zn}_{17}$ and the electronic part of $\text{La}_2\text{Zn}_{17}$ $\{(1-x)\gamma(\text{La}_2\text{Zn}_{17})\}$ from the data. $\text{La}_2\text{Zn}_{17}$ has a Sommerfeld parameter of $\gamma = 12.3 \text{ mJ/mol K}^2$.

Figure 2 shows the specific heat divided by temperature of the magnetic alloys ($0.85 \leq x \leq 1$) and the first nonordered compound with $x=0.8$ of the series $(\text{U}_x\text{La}_{1-x})_2\text{Zn}_{17}$. In Fig. 2(a) we present the specific heat divided by temperature T

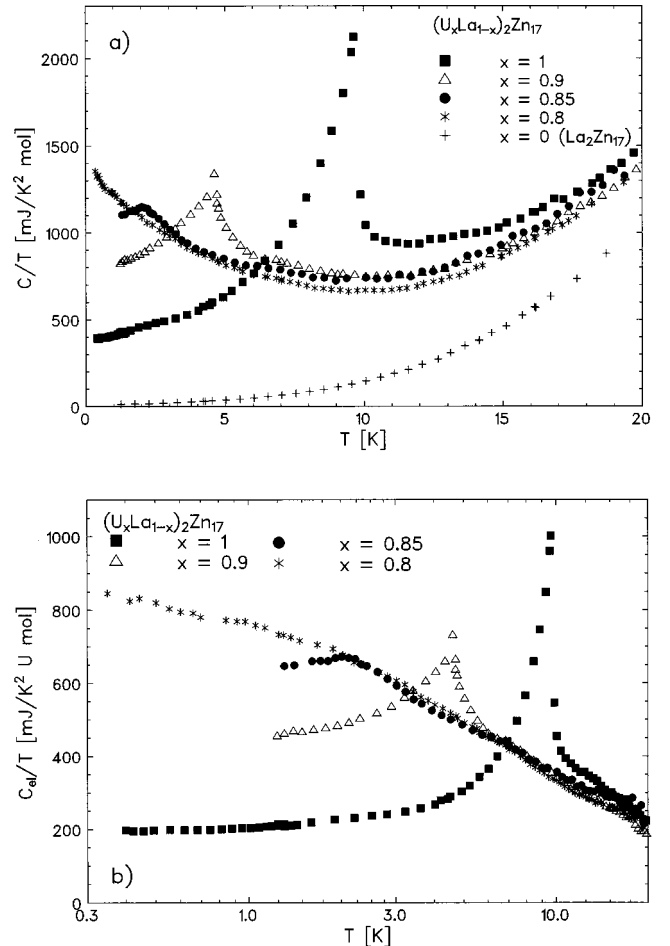


FIG. 2. (a) Specific heat and (b) electronic part of the specific heat divided by temperature, C_{el}/T , normalized to 1 mol uranium for $x \geq 0.8$. Note the logarithmic temperature axis in (b).

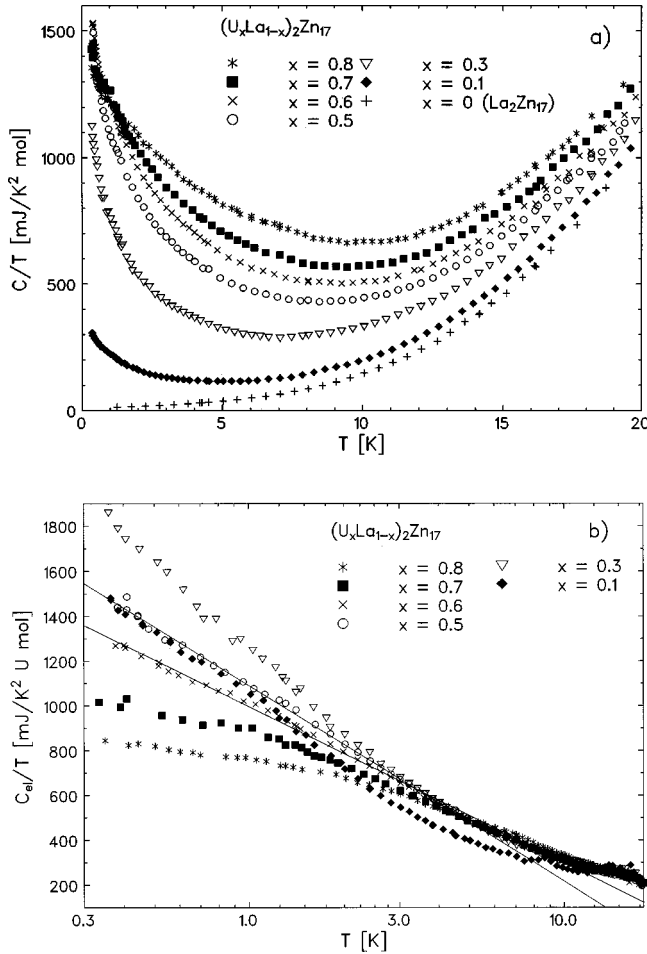


FIG. 3. (a) Specific heat and (b) electronic part of the specific heat divided by temperature, C_{el}/T , normalized to 1 mol uranium for $x \leq 0.8$. No anomaly due to a phase transition can be seen. The solid lines in (b) are fits to the data with $C/T = \gamma_0 - a \log_{10} T$. Note the logarithmic temperature axis in (b).

versus T as measured and in Fig. 2(b) the electronic part C_{el}/T normalized to 1 U mol vs $\log_{10} T$ is shown. The observed anomaly for $x > 0.8$ is due to the antiferromagnetic phase transition also present in pure U_2Zn_{17} . This is confirmed by magnetic measurements (not shown). The magnetic susceptibility as a function of temperature passes through a maximum at temperatures larger than T_N , and a maximum in $d\chi/dT$ marks the antiferromagnetic order. This behavior of the magnetic susceptibility is also well known for U_2Zn_{17} itself.¹⁹ The transition temperature T_N decreases with increasing applied field, a feature which supports the interpretation of the anomaly as an antiferromagnetic phase transition. Note that all curves in Fig. 2(b) fall together for temperatures above T_N . This indicates single-ion behavior in the paramagnetic region.

While Fig. 2 has C/T data for the magnetic alloys, Fig. 3 shows the data for the nonmagnetic samples ($x \leq 0.8$) for temperatures above 0.35 K. In Fig. 3(a) the data as measured are presented versus T and in Fig. 3(b) the electronic part C_{el}/T vs $\log_{10} T$. At low temperatures deviations from Fermi-liquid behavior are evident. For $x = 0.6$ and 0.5 , a

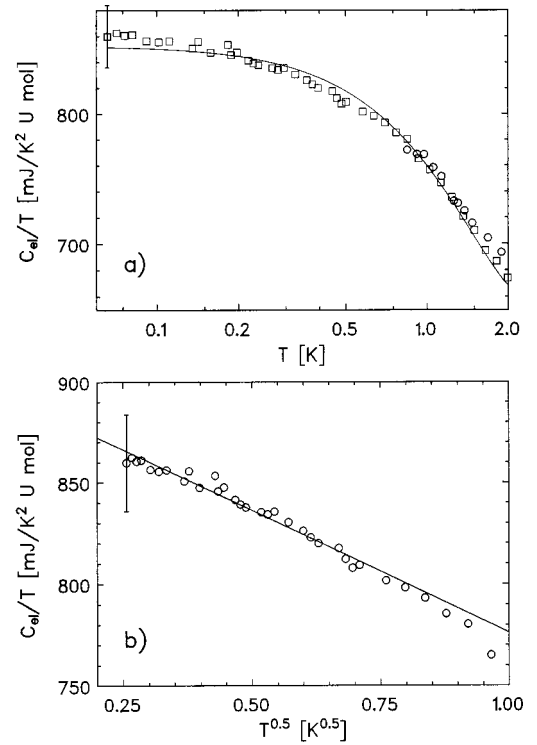


FIG. 4. Electronic part of the specific heat divided by temperature, C_{el}/T , normalized to 1 mol uranium down to the lowest temperature accessible with our experiments (0.06 K) for $(U_{0.8}La_{0.2})_2Zn_{17}$. In (a) the data are shown with a logarithmic temperature axis. The solid line represents a fit to the data with $C/T = C/T = \gamma_0 - a T^2 \ln(T/T_{SF})$ in the temperature range 0.06–2.5 K as described in the text. (b) shows the same data with an $T^{0.5}$ axis. The line in (b) is a guide to the eye (see text). The error bars sketch the error which is mainly systematic and weakly temperature dependent. Thus we think that the rise of C_{el}/T with decreasing temperature is not an artifact.

logarithmic divergence of C/T for more than one decade in temperature with decreasing temperature indicates non-Fermi-liquid behavior in these compounds, at least in the measured temperature range down to 0.35 K. The lines in Fig. 3(b) are fits with $C/T = \gamma_0 - a \log_{10} T$ to the data. As noted above for the data in Fig. 2(b), $0.8 \leq x \leq 1$, single-ion behavior is also observed [see Fig. 3(b)] at temperatures above 4 K for $x \leq 0.8$ (with some deviation for $x = 0.1$, which is, however, within the error bar).

Since the proper investigation of non-Fermi-liquid behavior requires data down to the lowest temperatures, measurements in a $^3\text{He}/^4\text{He}$ dilution refrigerator down to 0.06 K were performed on the nonmagnetically ordered ($x = 0.3, 0.5, 0.8$) samples. The results for $x = 0.8$, i.e., the composition where $T_N \rightarrow 0$, are given in Figs. 4(a) and 4(b). In Fig. 4(a) the data are presented with a logarithmic temperature axis. The solid line is a fit to the data with $C/T = \gamma_0 - a T^2 \ln(T/T_{SF})$, representing a spin fluctuation contribution to the specific heat²⁶ in the temperature range 0.06–2.5 K. The best-fit parameters are $\gamma_0 = 850 \text{ mJ/U mol K}^2$, $a = 90 \text{ mJ/K}^4 \text{ U mol}$, and $T_{SF} = 4 \text{ K}$. Figure 4(b) shows C_{el}/T vs $T^{0.5}$ at the lowest temperatures; the solid line, which is a guide to the eye, indicates that C_{el}/T can also be fitted to $T^{0.5}$ between 0.06 and 0.64 K.

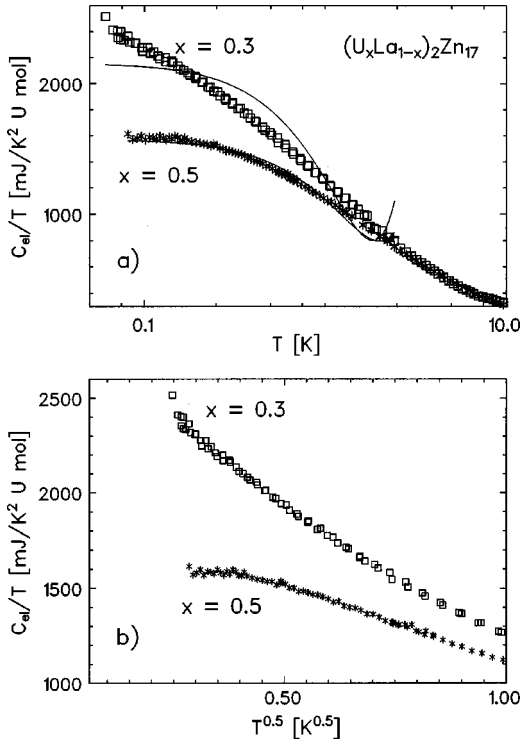


FIG. 5. Electronic part of the specific heat divided by temperature, $C_{el}T$, normalized to 1 mol uranium down to the lowest temperature accessible with our experiments for $(U_{0.5}La_{0.5})_2Zn_{17}$ (stars) and $(U_{0.3}La_{0.7})_2Zn_{17}$ (squares). In (a) the data are shown with a logarithmic temperature axis. The solid lines are fits to the data with $C/T = C/T = \gamma_0 - a T^2 \ln(T/T_{SF})$ in the temperature range 0.06–2.5 K as described in the text. (b) shows the same data with a $T^{0.5}$ axis.

Comparing the C/T data for $x=0.8$ in Fig. 2 with Fig. 4(b), one can ask if these data are consistent with the weak interaction theory¹⁵ of Moriya and Takimoto, where at the lowest temperatures $C/T \propto 1 - \sqrt{T}$, while $C/T \propto -\log T$ for a limited ($\sim 60\%$ of a decade) range of temperature at (Fig. 2) higher temperature. Unfortunately, such a simple solution to the difficulty demonstrated in Figs. 4(a) and 4(b) in distinguishing the temperature dependence of the lowest temperature C/T data does not work. First, as the data in Fig. 2 show, C/T for $x=0.8$ obeys $(-\log T)$ over a much broader temperature range than predicted by Moriya and Takimoto. Second, as will be seen below in the discussion of ρ , the Moriya-Takimoto prediction of $\rho = \rho_0 + AT^{1.5}$ at low temperatures is not followed at all. Thus, as will be discussed for ρ and also χ data further below, this ambiguity in what the low-temperature C/T data imply for $x=0.8$ is not resolvable using the data reported on herein with current theoretical understanding. [The possibility of fitting C/T for $(U_xLa_{1-x})_2Zn_{17}$ using the disorder model—as done, e.g., in Ref. 12 for $UCu_{5-x}Pd_x$ —will be discussed below for the more disordered $x=0.3$.]

The low-temperature C data for $x=0.5$ and 0.3 are shown in Fig. 5. Again, in Fig. 5(a) the data are plotted versus a logarithmic temperature scale. The solid lines are spin fluctuation fits, like in Fig. 4(a), in the temperature range 0.06–2.5 K. Whether or not the spin fluctuation fit to the $x=0.5$

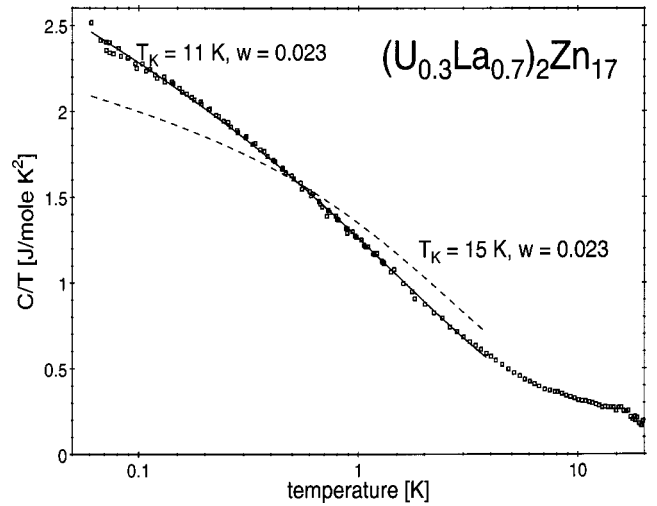


FIG. 6. Specific-heat data for $(U_{0.3}La_{0.7})_2Zn_{17}$ plotted vs $\log T$ and fit (shown by the solid line through the data) to the disorder model of Bernal *et al.* (Ref. 12). The average Kondo temperature $\langle T_K \rangle$ and the width of the distribution of Kondo temperatures, w , which best fit the specific-heat data are 11 K and 0.023, respectively; the disorder model fit is a good approximation of the data between 0.06 and ~ 4 K. [The other two parameters in the disorder model of Bernal *et al.* (Ref. 12), the effective moment and the Fermi energy, are taken from the literature (Ref. 19).] A disorder model fit to the magnetization data (not shown) as a function of temperature and field yields the same distribution width of Kondo temperatures and a slightly different $\langle T_K \rangle$ of 15 K. Using the parameters from the fit to the magnetization data would result—as shown by the dashed line—in a qualitatively much worse fit to the specific heat data. In the theory of Bernal *et al.* (Ref. 12), the parameters for fitting both the specific heat and magnetization should be the same.

C/T data, with its small systematic deviations visible in Fig. 5(a), is a good representation of the data is an open question. In any case, for $x=0.5$ a constant value of C_{el}/T , i.e., Fermi-liquid behavior, is reached at temperatures below 0.15 K.

The low-temperature data for $x=0.3$ show, in contrast, non-Fermi-liquid (NFL) behavior down to the lowest temperature: i.e., C/T does not tend to a constant value. An attempt to fit the data with an additional spin fluctuation contribution to the specific heat failed for this concentration, as can be seen by the large deviation of the fit (solid line) from the data in Fig. 5(a). When plotted against a $T^{0.5}$ axis [Fig. 5(b)], no linear region is found for $x=0.3$ or 0.5 . Since there is inherent disorder in the $(U_{0.3}La_{0.7})_2Zn_{17}$ sample, can the NFL disorder phenomenological model of Bernal *et al.*¹² give a convincing fit to the C/T data? The answer, as shown in Fig. 6, is a qualified yes. Assuming a distribution of Kondo temperatures caused by disorder changing the local Kondo compensation of the U $5f$ -electron moments, the disorder model of Bernal *et al.* (using four fit parameters) in fact does succeed in fitting the data between 0.07 and ~ 4 K. However, it should be pointed out that the fit parameters derived from fitting the magnetization data (not shown) of $(U_{0.3}La_{0.7})_2Zn_{17}$ as a function of temperature and field—which parameters according to the model¹² should also fit the

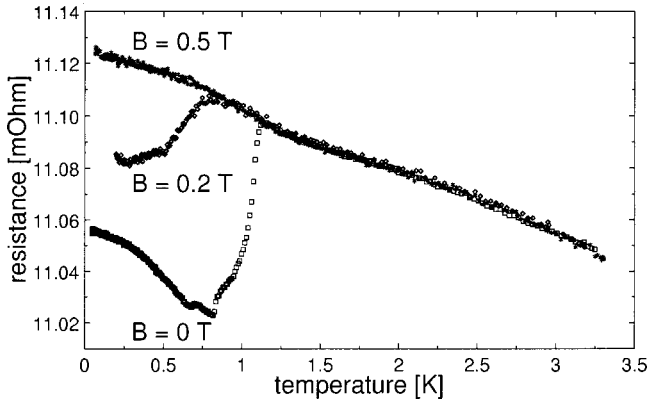


FIG. 7. Low-temperature resistance of $(U_{0.8}La_{0.2})_2Zn_{17}$ in 0, 0.2, and 0.5 T where the applied field serves to suppress the superconducting transition (believed due to a slight second phase of Zn, $T_c=0.85$ K).

specific-heat data—give a different result for C/T than that measured, see Fig. 6.

The resistivity data for $x=0.8$ are shown in Fig. 7. Due to a trace of superconducting Zn, $T_c \sim 0.85$ K, the data were measured in an applied field of up to 5000 G. As shown in Fig. 7, this field does not change the resistivity above 1 K and it is reasonable to consider these (relatively low) field data as characteristic of $(U_{0.8}La_{0.2})_2Zn_{17}$. As may be seen from the data, $\rho - \rho_0 - AT^1$, albeit with some waviness to the linear behavior. The ρ data certainly fit neither Fermi-liquid behavior ($\rho = \rho_0 + AT^2$) nor the $\rho = \rho_0 + AT^{1.5}$ prediction¹⁵ of Moriya and Takimoto. In the disorder model of Rosch for the resistivity,²⁷ such an exponent α (≈ 1) in $\rho = \rho_0 - AT^\alpha$ is possible over such an extended temperature range only for rather well-ordered samples, which is not consistent with the disorder inherent in the $x=0.8$ sample where 20% of the U has been substituted by La.

Resistivity data for $(U_xLa_{1-x})_2Zn_{17}$, $x=0.05, 0.3, 0.9$, and 1.0, have been previously reported.²⁰ Of interest for the discussion below are the data for $x=0.3$, which show little measurable temperature dependence below 1 K down to their lowest temperature of measurement (0.3 K). However, what little temperature dependence there is is consistent with $\rho \propto \rho_0 - AT^\alpha$, where $\alpha < 1$ —again inconsistent with either Fermi-liquid, Moriya-Takimoto, or Rosch²⁷ predicted behavior.

The low-temperature magnetic susceptibility data are shown in Fig. 8. The χ data for $x=0.8$ show no pure temperature dependence, instead showing an inflection point around 4 K. Remembering the dichotomy displayed in Fig. 4 for the temperature dependence of C/T at low temperature, where one of the fits [Fig. 4(a)] was due to the temperature dependence of a spin fluctuation system, it is interesting to note that the χ data for $x=0.8$ shown in Fig. 8(a) are very reminiscent²⁸ of χ data for the canonical spin fluctuation compound UA_2 . In contrast, the χ data in Fig. 8 for $x=0.7, 0.6$, and 0.5 obey a canonical NFL dependence, $\chi = \chi_0 - a\sqrt{T}$, and the χ data for $x=0.3$ and 0.1 both obey $\chi \propto -\log T$, again a standard⁶ NFL temperature dependence. When considering the Griffiths phase disorder model of Cas-

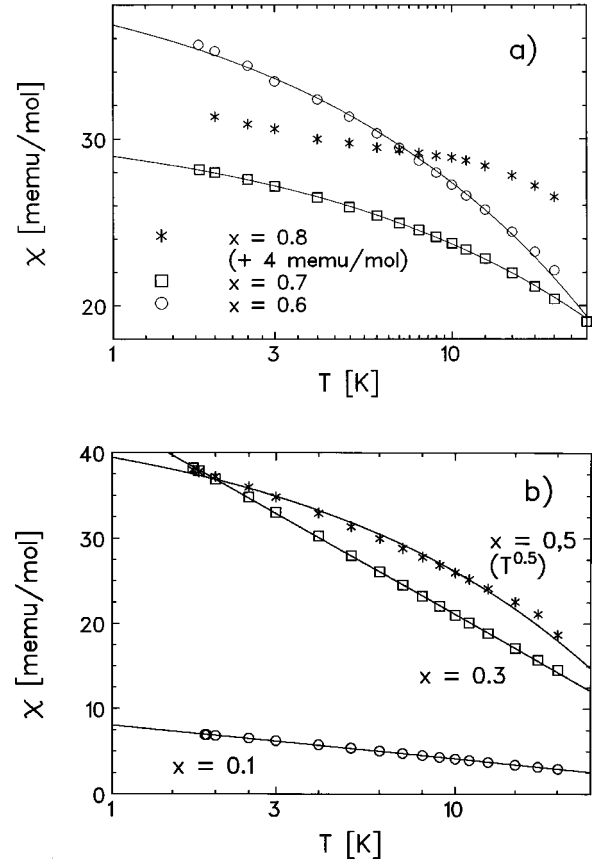


FIG. 8. Low-temperature magnetic susceptibility for (a) $x=0.6-0.8$ and for (b) $x=0.1$ (circles), $x=0.3$ (squares), and $x=0.5$ (stars) in $(U_xLa_{1-x})_2Zn_{17}$ vs $\log T$. The curved solid lines through the data for $x=0.5-0.7$ are fits to $\chi = \chi_0 - a T^{0.5}$, while the straight lines through the $x=0.1$ and 0.3 data show the $\chi = \chi_0 - a \log T$ behavior.

tro Neto *et al.*,²⁹ a possible sign of the Griffiths phase rare magnetic clusters is spin-glass-like behavior, including disagreement between $\chi_{\text{field cooled}}$ and $\chi_{\text{zero field cooled}}$. Both the $x=0.3$ and $x=0.1$ samples show such a divergence at around 25–30 K, which is consistent with the disorder model of Castro Neto *et al.* However, the Griffiths phase model of Castro Neto *et al.* would predict that $C/T \sim T^{-1+\lambda}$, which is not observed (see Fig. 9) for either sample below about 1 K.

DISCUSSION

The chief results of this work are (a) the interplay between magnetism and heavy-fermion behavior for the magnetically ordered alloys of the series $(U_xLa_{1-x})_2Zn_{17}$ and (b) the appearance of non-Fermi-liquid behavior in the nonordered alloys.

As shown in Fig. 2 and Table I, we find for the magnetic samples ($0.85 \leq x \leq 1$) coexistence of heavy-fermion behavior and antiferromagnetic order. The values of C/T for $T \rightarrow 0$ K increase from 195 mJ/mol K² for $x=1$ to 640 mJ/mol K² for $x=0.85$, while T_N decreases from 9.6 to 2 K. The high values of C_{el}/T , which have been found at least for U_2Zn_{17} down to 0.35 K, are almost temperature independent.

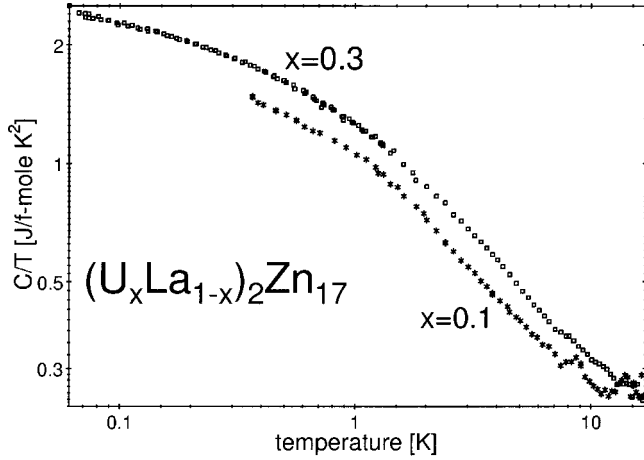


FIG. 9. Specific heat divided by temperature vs temperature of $(U_xLa_{1-x})_2Zn_{17}$ for $x=0.1$ and 0.3 shown on a log-log plot. If the Griffiths phase model of Castro Neto *et al.* (Ref. 29) were to describe the data, then C/T would behave as a power law, $T^{1+\lambda}$, and $\log C/T$ vs $\log T$ would be a straight line. This is the case between about 1 and 8 K for both sets of data, but below 1 K the power law dependence predicted by the Griffiths phase model is not obeyed by the data.

This indicates a true heavy-fermion ground state since fluctuations of the magnetic order parameter (which can cause an increase in C_{el}/T and thus approximately mimic an increase in γ) should give a temperature-dependent contribution to

TABLE I. Lattice parameters a and c , the Néel temperature T_N , and the electronic part of the specific heat divided by temperature for samples of the series $(U_xLa_{1-x})_2Zn_{17}$.

$(U_xLa_{1-x})_2Zn_{17}$	a [Å] ± 0.004	c [Å] ± 0.01	T_N [K]	C_{el}/T [mJ/U mol K ²] ^a
$x=1$	8.980	13.159	9.6 ^b 9.6 ^c	400 (10 K) 195 (0.4 K)
$x=0.9$	8.991	13.172	4.6 ^b 4.7 ^c	450 (6.3 K) 450 (1.3 K)
$x=0.85$	9.001	13.180	2.0 ^b 2.2 ^c	630 (3 K) 640 (1.3 K)
$x=0.8$	9.004	13.186		850 (0.35 K)
$x=0.7$	9.022	13.214		1000 (0.33 K)
$x=0.6$	9.036	13.221		1250 (0.38 K)
$x=0.5$	9.052	13.241		1450 (0.38 K)
$x=0.3$	9.081	13.288		1850 (0.36 K)
$x=0.1$ ^d	9.119	13.311		1500 (0.35 K)
$x=0$ (La_2Zn_{17})	9.133	13.326		12.3 ^e

^aThe lattice contribution to the specific heat and the electronic part of $La_2Zn_{17}(1-x)\gamma(La_2Zn_{17})$ have been subtracted from the data.

^bMaximum of the specific heat.

^cMaximum in $d\chi/dT$. Here (T) is measured in a field of $B_0 = 5$ kG.

^dThe x-ray pattern shows a small contribution of an unidentified second phase.

^e $C/T = \gamma + \beta T^2$ ($T < 6$ K). Here $\gamma = 12.3$ mJ/mol K² and $\theta_D = 333$ K.

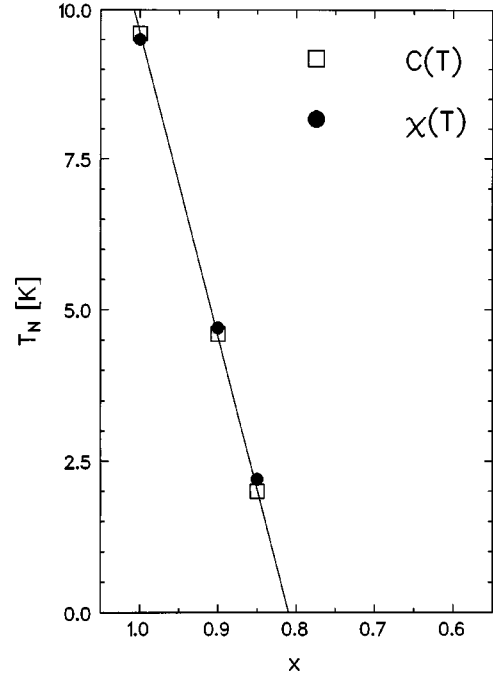


FIG. 10. Néel temperature as a function of uranium concentration x .

C_{el}/T which would vanish at the lowest temperatures. The entropy of the f -electron system at 20 K, calculated by integrating C_{el}/T after having extrapolated the data to obtain values for $T=0$ K, increases with increasing La doping (rising from $1.2R \ln 2$ for $x=1$ to $1.35R \ln 2$ for $x=0.85$). The data for U_2Zn_{17} are in agreement with Ref. 30, where the entropy in excess of the value for a possible ground-state doublet has been attributed to either crystal field effects or the possibility of hopping yielding a band structure. The observed transfer of entropy from higher to lower temperatures with decreasing x cannot be explained without a model for the high-temperature entropy for pure U_2Zn_{17} itself.

The decrease of T_N with decreasing U concentration is shown in Fig. 10; the data are describable by a linear behavior of T_N as a function of temperature. An extrapolation to $T_N=0$ K yields a U concentration of $x \approx 0.8$ for the critical concentration where T_N vanishes. Thus, for the sample with $x=0.8$, non-Fermi-liquid behavior at the lowest temperatures might be expected due to the nearness of a quantum critical point ($T_N=0$ K). In the literature the occurrence of non-Fermi-liquid behavior at such a so-called magnetic instability is well established on both theoretical and experimental grounds. Several calculations^{14–16} of the specific heat give non-Fermi-liquid behavior at low temperatures at the quantum critical point ($T_N=0$ K) with $C/T = \gamma_0 - aT^{0.5}$. Experimental evidence for a non-Fermi-liquid ground state ($C/T = \gamma_0 - aT^{0.5}$ or $C/T = \gamma_0 - a \log T$) at this critical concentration was found in systems like $CeCu_{6-x}Au_x$ (Ref. 5) and $CeCu_{6-x}Ag_x$ (Ref. 13). As shown in Fig. 4(b), the data for $x=0.8$ can be described by $C/T = \gamma_0 - aT^{0.5}$, a behavior expected for an antiferromagnetic quantum critical point (three-dimensional-antiferromagnetism), up to 0.64 K followed by $C/T \propto -\log T$ above 2 K. However, although no saturation in

the values of C/T can be seen in the inset of Fig. 4(a), these data can also be described with $C/T = \gamma_0 - aT^2 \ln(T/T_{SF})$, the temperature dependence predicted for a Fermi-liquid with spin fluctuations,²⁶ as shown in Fig. 4(a). Considering now the (higher-temperature) χ data for $x=0.8$ (Fig. 8) and the low-temperature ρ data (Fig. 7), we see that these measurements also show *both* spin-fluctuation-consistent behavior (χ is like that of UAl_2 in its temperature dependence and is certainly *not* characteristic of NFL systems) *and* behavior characteristic of NFL systems ($\rho \propto \rho_0 - aT$, whereas $\rho_{UAl_2} \sim \rho_0 + aT^2$). Obviously, lower-temperature susceptibility data might help resolve this conflict.

For samples with $x < 0.8$, non-Fermi-liquid behavior, $C/T = C/T = \gamma_0 - a \log T$ and $\chi \propto \chi_0 - aT^{-0.5}$ for $x = 0.5-0.7$ and $\chi \propto \chi_0 - a \log T$ for $x = 0.1-0.3$, is observed in large temperature regions [see Figs. 8 and 3(b)]. However, when regarding the C data obtained at the lowest temperatures (Fig. 5), the situation is less clear. Plotted against $\log T$, all curves show signs of saturation at the lowest temperatures. For $x=0.5$ a constant value of C/T is found below 0.15 K, indicating rather conclusively a Fermi-liquid ground state. This view is further supported by the fact that below 2.5 K the data are well represented by a constant γ_0 and a spin fluctuation contribution [see Fig. 5(a)]. The C data for the sample with $x=0.3$ continue, on the other hand, to increase with decreasing temperature down to the lowest temperatures. Fitting these data with a spin fluctuation contribution is not possible [see Fig. 5(a)]. Although there is a disagreement in the parameters derived from the C/T and magnetization data, the $x=0.3$ C/T data (see Fig. 6) can, however, be phenomenologically fit to the disorder model of

Bernal *et al.* Thus, as Bernal *et al.*¹² showed, disorder resulting in a distribution of Kondo temperatures can cause an increasing C/T down to the lowest temperatures.^{12,31} A thorough investigation of this topic demands microscopic probes like NMR or muon spin resonance (μ SR) to measure a possible spatial inhomogeneity of the physical properties because of a local distribution of the Kondo temperatures. (The quadrupolar Kondo model, while in principle a possible mechanism in uranium compounds, is in the present case not a suitable model because it should not appear in rhombohedral systems.³²)

Recently, non-Fermi-liquid behavior has been reported in alloys of UPt_3 [$U_{1-x}(Hf, Zr)_xPt_3$ in Ref. 33] and UAl_2 ($U_{1-x}Y_xAl_2$ in Ref. 34), also far away from any detectable magnetic instability. Both these systems, as well as $(U_xLa_{1-x})_2Zn_{17}$, show evidence of spin fluctuations in the specific heat. Thus the occurrence of NFL behavior in $(U_xLa_{1-x})_2Zn_{17}$ could possibly be, depending on the results of investigation of the role of disorder, a member of a new class of NFL systems, one where the long-range magnetic excitations necessary to prevent achieving Fermi-liquid behavior are not derived from the nearness to a critical point in the phase diagram where $T_{mag} \rightarrow 0$.

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