Specific heat variation as $T^{0.5}$ in Th-doped UIr_2Zn_{20}: Consistent with weak coupled quantum critical behavior

J. S. Kim and G. R. Stewart
Department of Physics, University of Florida, Gainesville, Florida 32611-8440, USA

E. D. Bauer
Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA

(Received 18 April 2008; revised manuscript received 8 June 2008; published 18 July 2008)

By partially replacing the U in single crystals of UIr_2Zn_{20} with Th, we have suppressed the 2.1 K ferromagnetic-like transition present in the pure compound. The magnetic susceptibilities of $U_{1-x}Th_xIr_2Zn_{20}$ show enhanced low-temperature values, with χ (2 K) for $x=0.25$ around 140 memu/Umole. However, unlike the parent compound, for $U_{1-x}Th_xIr_2Zn_{20}$ ($x \geq 0.25$) the extrapolation of even the lowest temperature $1/\chi$ vs T data shows an intercept on the negative temperature axis—consistent with antiferromagnetic fluctuations. The specific heat at low temperature in $U_{0.75}Th_{0.25}Ir_2Zn_{20}$ shows no magnetic transition down to 0.4 K, but rather obeys $C/T=\gamma-aT^{0.5}$ over more than a decade of temperature, consistent with weak-coupling three-dimensional antiferromagnetic fluctuations. Low temperature resistivity data show an unusual non-Fermi-liquid temperature dependence, where $\rho=\rho_0+aT^{\alpha}$ with $\alpha<1$. The specific-heat data for $U_{0.5}Th_{0.5}Ir_2Zn_{20}$ can be fit within the normal Fermi-liquid picture by the addition of a $T^3 \log T/T_{SF}$ term, where T_{SF} is the spin-fluctuation temperature.

DOI: 10.1103/PhysRevB.78.035121   PACS number(s): 71.10.Hf, 71.27.+a, 71.20.Lp, 65.40.Ba

I. INTRODUCTION

One of the more interesting set of compounds discovered in recent years that exhibit heavy Fermion behavior is the RX_2Zn_{20} (R=lanthanide,Th,U; X=transition metal) series.[1–5] The f-ion interatomic spacings are quite large (>6 Å), and in addition to the heavy Fermion behavior observed, there is strong ferromagnetic behavior in a number of these compounds. The values of the specific-heat γ values for the samples that do not order magnetically, where γ is proportional to the large heavy Fermion effective mass m^*, are typically in the range of 500–750 mJ/mole K^2 for the Yb compounds while for the UX_2Zn_{20} (X=Fe,Ru,Co,Rh) samples γ ranges[5] between 50 and 250 mJ/mole K^2.

One of the most widely used methods for tuning magnetic samples to a possible quantum critical point is via doping.[6–8] In the case of a first-order ferromagnetic transition as seen in UIr_2Zn_{20}, in order to reach a quantum critical point the doping must change the order of the transition as it is suppressed to $T=0$ to a second-order phase transition. As well, when doping suppresses magnetic order the entropy previously associated with the magnetic order often appears as an enhanced low temperature γ. For both these reasons, the current work investigates the properties of UIr_2Zn_{20} doped with Th on the U site in order to look for a possible quantum critical point and its associated non-Fermi-liquid behavior, as well as to examine the low-temperature specific heat for enhanced heavy Fermion behavior with the ferromagnetism suppressed.

II. EXPERIMENT

Samples were prepared using a procedure similar to that in the discovery work:[1] the elements U:Th:Ir:Zn in the ratio 1-x:x:2:100, where x was either 0.25 or 0.50, were placed in an outgassed BeO crucible with lid, which was sealed in Ta under a half-atmosphere of purified Ar. This Ta container was placed in a tube furnace with high purity Ar flowing to prevent oxidation. The sample was heated to 600 °C for 12 h, then to 1050 °C at 75 °C/h for a 4 h soak, followed by a slow cool at 4 °C/h to 700 °C. The furnace was then turned off and the sample was allowed to cool. The sample was then removed from its containment and sealed under a partial atmosphere of high-purity Ar above quartz wool in a quartz tube. This tube was heated to 700 °C, and then centrifuged to remove the excess Zn. In our experience in working with these crystals grown in Zn flux over the last several years, we sometimes find less than 5% of Zn either on the surface of the crystal left from centrifuging the sample at elevated temperatures, or small inclusions within the sample that occur during the growth process which can only be detected in resistivity (i.e., not in the bulk magnetization and specific heat) measurements.

X-ray diffractometer analysis indicated the correct cubic $Mg_3Cr_2Al_{18}$ structure with the lattice parameters a_0=14.2026(14.2256) Å for x=0.25(0.50). Compared to the published[1] a_0 values for pure UIr_2Zn_{20} and $ThIr_2Zn_{20}$ of 14.1783 and 14.301 Å, respectively, these values for Th-doped UIr_2Zn_{20} indicate a slightly less than linear (Vegard's law) lattice expansion with the addition of Th. Specifically, the measured lattice parameters for our doped $U_{1-x}Th_xIr_2Zn_{20}$ indicate that the actual Th composition of our samples is approximately 20% less than the nominal composition, i.e., the as-grown samples have the respective compositions $U_{0.80}Th_{0.20}Ir_2Zn_{20}$ and $U_{0.61}Th_{0.39}Ir_2Zn_{20}$. Thus, we achieved the desired approximate doubling of the Th composition in the second sample, however, apparently some of the Th in the reaction crucible was not taken up in the crystals. Hereafter the nominal compositions will be used in labeling the samples.

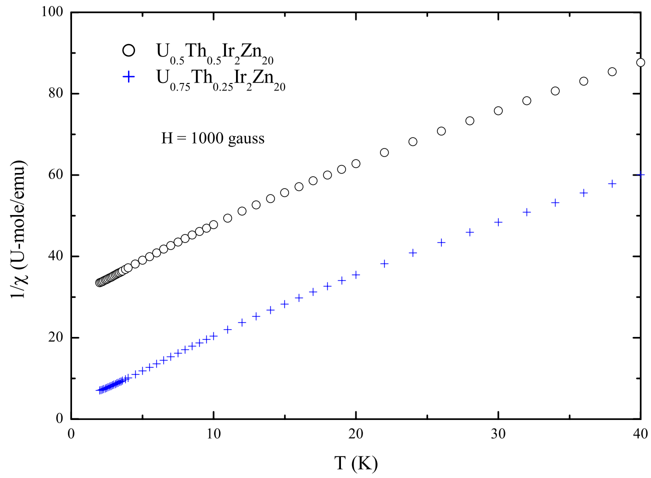

FIG. 1. (Color online) Inverse magnetic susceptibility of $U_{1-x}Th_xIr_2Zn_{20}$ (x=0.25,0.50) vs temperature.

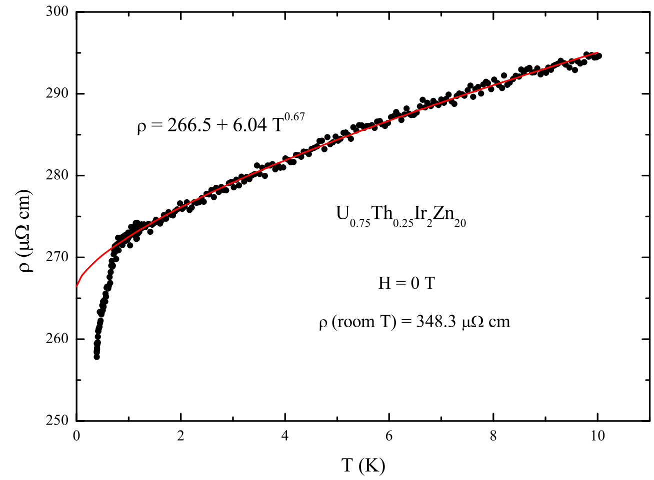

FIG. 2. (Color online) Low temperature resistivity of single crystal $U_{1-x}Th_xIr_2Zn_{20}$ (x=0.25). The dip in the resistivity below 0.8 K is likely due to Zn inclusions from the flux growth process. A small field above the critical field of elemental Zn of 0.1 T is sufficient to suppress this (data not shown). Note that a field of 0.1 T, rather than just above the 0.005 T critical field of pure Zn, was used to avoid any problems setting the field in the magnet due to remanent field in the 20 T magnet in the SCM-1 facility at NHMFL, Tallahassee, which was approximately 0.03 T on the day these measurements were made.

Magnetic susceptibility and magnetization measurements were carried out in a Quantum Design MPMS. The resistivity was measured using a four-point-contact method and dc current, with the current reversed and the resultant voltage drop averaged for each point. Multiple measurements were taken at each temperature and averaged in order to reduce scatter due to electrical noise, particularly at low temperatures (down to 0.1 K) in the SCM-1 dilution refrigerator facility at NHMFL Tallahassee. Specific heat measurements in zero and applied magnetic fields were performed on the small single-crystal samples using established techniques.[9]

III. RESULTS AND DISCUSSION

The inverse magnetic susceptibility, $1/\chi$, plotted vs temperature is shown in Fig. 1 for $U_{1-x}Th_xIr_2Zn_{20}$ (x =0.25,0.50). In the parent compound, UIr_2Zn_{20}, a rapid, ferromagneticlike increase in χ (to ~1.4 emu/mole) occurs[1] below 3 K with the accompanying intercept of the $1/\chi$ vs T plot occurring on the positive T axis. As seen in Fig. 1, the effect of the Th doping has been to suppress this ferromagneticlike transition, with χ (x=0.25) ~140 memu/Umole at 2 K. The intercept of the extrapolations of the low-temperature $1/\chi$ data in Fig. 1 are −2.5 and −16 K, respectively, for x=0.25 and 0.50, indicating antiferromagnetic fluctuations.

The low-temperature electrical resistivity for $U_{1-x}Th_xIr_2Zn_{20}$ (x=0.25) was measured and is shown in Fig. 2. There is a slight transition below about 0.8 K which is most likely due to residual filamentary Zn inclusions in the crystal measured. In order to check this supposition, a 0.1 T (above the critical field for pure Zn) field was applied and the resistivity data (not shown) at low temperatures were remeasured. ρ data down to 0.1 K in an applied field of 0.1 T showed no signs of a transition; ρ (0.1 K, 0.1 T) was found to be 270.4 $\mu\Omega$ cm. As is clear from Fig. 2, the resistivity increases less than linearly in temperature with increasing temperature. This was also the case[1] above T_{Curie} in the parent compound. Thus, there may be some remanent behavior from the parent compound that is dominating the resistivity (but not the $1/\chi$) in the Th-doped sample. There are several non-Fermi-liquid compounds where[6,7] a sublinear resistivity temperature exponent of $\alpha \sim 2/3$ is observed, for example[10] $UCu_{3.5}Al_{1.5}$. Whether some particular significance attaches to the value of α in $U_{1-x}Th_xIr_2Zn_{20}$ (x=0.25) in $\rho=\rho_0+aT^{\alpha}$ for understanding the non-Fermi-liquid behavior is problematic, since we cannot exclude the possibility that the presence of filaments of Zn could affect the temperature dependence. It should be noted that such low values of the temperature exponent α are not consistent with current theories of weak-coupling quantum critical behavior, which instead predict[6] α=1.5. It is interesting to note, however, that low-field magnetoresistance data at 0.3 K (not shown) on the $U_{1-x}Th_xIr_2Zn_{20}$ (x=0.25) sample show (after the suppression of the transition) a linear in field suppression of the resistivity up to 2 T, which might[11] be consistent with suppression of spin-fluctuation scattering in a dirty system near a weak coupled antiferromagnetic quantum critical point.

The specific heat divided by temperature vs temperature is shown in Fig. 3 for both $U_{1-x}Th_xIr_2Zn_{20}$ (x=0.25 and 0.50) samples, as well as for the parent compound UIr_2Zn_{20}. Considering first the magnitude of the specific heat, the specific-heat coefficient γ (=C/T as $T\to 0$), proportional to the effective mass m^*, is 415(1035) mJ/U mole K^2 for $U_{1-x}Th_xIr_2Zn_{20}$ [x=0.50(0.25)]. Thus, as discussed above the suppression of the magnetic order in UIr_2Zn_{20} has indeed resulted in a significant enhancement over the γ values observed in the other UX_2Zn_{20} compounds as the entropy formerly involved in the transition has been shifted into a rise in C/T at lower temperatures, at least for x=0.25. Another way of stating this is that the conservation of entropy, S, (=$\int(C/T)dT$) in the system implies that the magnetic entropy is found in the magnitude of C/T at low temperature in the

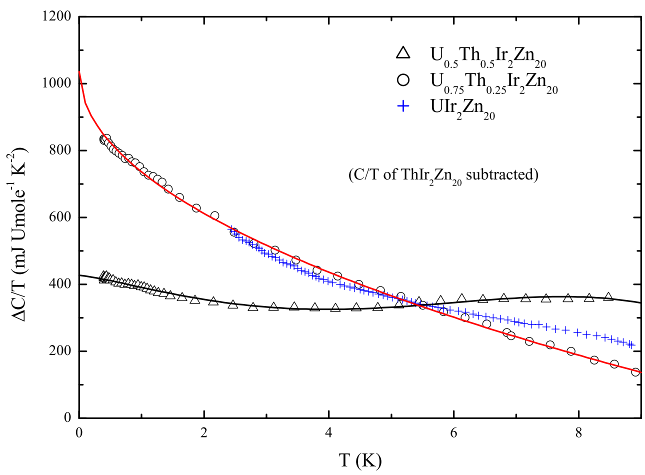

FIG. 3. (Color online) Low temperature specific heat, C, normalized per U mole divided by temperature, T, vs T for $U_{1-x}Th_xIr_2Zn_{20}$ ($x=0,0.25,0.50$), with the data for $x=0$ only shown down in temperature to just above the magnetic transition. A fit (C/T=22.4+0.0725 T^2+0.02324 T^4 in units of mJ/mole-K^2 for C/T) to the specific heat of $ThIr_2Zn_{20}$ from Ref. 1 has been subtracted from the measured data. The $x=0.25$ data can be fit to the weak coupled Millis-Hertz form $C/T=\gamma-a\sqrt{T}$ ($\gamma=1035$ mJ/Umole K^2 and $a=300$ mJ/Umole K$^{5/2}$) over the whole temperature range of measurement, consistent with antiferromagnetic fluctuations near a quantum critical point. Even though there are only two adjustable parameters, note the rather good (less than $\pm 2\%$ deviation) fit [as good as that (Ref. 12) for, e.g., U_2Co_2Sn—another example of $C/T=\gamma-a\sqrt{T}$]. As Th doping is increased to $x=0.5$, the strong upturn in C/T at low temperatures observed for $x=0.25$ is decreased to a broad minimum in C/T at 3.5 K with only a 30% increase down to 0.3 K. Interestingly, this temperature dependence in C/T for $x=0.5$ can be fit ($C/T=415-22.74\ T^2-0.0969\ T^4+31.2\ T^2 \log T$ in units of mJ/Umole K^2 for C/T)—albeit with more adjustable parameters than in the Millis-Hertz fit to the $x=0.25$ data—to a $\delta T^2 \log T/T_{SF}$ behavior, indicative of ferromagnetic fluctuations (Ref. 13) ("paramagnons") and consistent with Fermi liquid behavior. (Note that the large negative T^2 coefficient is primarily from $-\delta \log T_{SF}$.)

doped system. For $x=0.5$ the entropy per U mole is still present, just not so concentrated at the lowest temperature (i.e., not found so much in γ) but rather spread over a broader temperature range, see Fig. 3. In this connection, note that the specific heat, normalized per U mole, for the $x=0.25$ sample agrees in magnitude (see Fig. 3) with that of pure UIr_2Zn_{20} just above the parent compound's T_{Curie}, although the temperature dependence is somewhat altered.

Considering now the temperature dependence of the specific-heat results shown in Fig. 3, the data for $x=0.25$ over the whole temperature range of measurement, 0.4–9 K, fit a simple two parameter fit of C/T to $\gamma-aT^{1/2}$, which is[6] the predicted Millis-Hertz weak-coupling fluctuation quantum critical non-Fermi-liquid behavior for a three-dimensional sample with antiferromagnetic fluctuations. This is in agreement with the temperature axis intercept of the $1/\chi$ data from Fig. 1.

As the Th content is increased to $x=0.5$, the sample properties such as χ (2 K) (Fig. 1) and C/T ($T\to 0$) are expected to be less enhanced (see, e.g., Ref. 6) since it is further from the critical concentration (inferred to be $0<x_c<0.25$) where the magnetism is suppressed in the phase diagram. In addition, as composition is varied away from the critical-point concentration x_c where T_C of pure UIr_2Zn_{20} is suppressed to $T=0$, the temperature dependence predicted[6] by Millis-Hertz near x_c should become less critical, or more Fermi-liquid-like. This is exactly what is seen in Fig. 3: the specific-heat data can be fit to an additional $\delta T^3 \log T/T_{SF}$ term which is a sign of paramagnons[13] and is consistent with Fermi-liquid behavior. (T_{SF} is the characteristic energy of the paramagnon fluctuation spectrum.)

IV. CONCLUSIONS

Doping of UIr_2Zn_{20} with nominal concentrations 25% and 50% Th on the U site results in a suppression of the ferromagnetism at 2.1 K observed in the pure compound. The magnetic susceptibility shows an intercept of $1/\chi$ with the negative temperature axis, consistent with antiferromagnetic fluctuations. This is in agreement with the $\sqrt{T}$ temperature dependence in the low-temperature C/T data for $U_{1-x}Th_xIr_2Zn_{20}$ ($x=0.25$) over more than a decade of temperature, which is predicted for systems with weakly coupled antiferromagnetic fluctuations. The sublinear temperature dependence of the low-temperature resistivity of $U_{1-x}Th_xIr_2Zn_{20}$ ($x=0.25$), although clearly not Fermi liquid in nature ($\rho \sim T^2$ for a Fermi liquid), is not consistent with current non-Fermi-liquid theories.

ACKNOWLEDGMENTS

Work at the University of Florida performed under the auspices of the U.S. Department of Energy, Contract No. DE-FG02–86ER45268. Work at Los Alamos performed under the auspices of the U.S. Department of Energy. Work at the National High Magnetic Field Laboratory in Tallahassee supported by the U.S. National Science Foundation.

[1] E. D. Bauer, A. D. Christianson, J. S. Gardner, V. A. Sidorov, J. D. Thompson, J. L. Sarrao, and M. F. Hundley, Phys. Rev. B **74**, 155118 (2006).

[2] S. Jia, S. L. Bud'ko, G. D. Samolyuk, and P. C. Canfield, Nat. Phys. **3**, 334 (2007).

[3] M. S. Torikachvili, S. Jia, E. D. Mun, S. T. Hannahs, R. C. Black, W. K. Neils, D. Martien, S. L. Bud'ko, and P. C. Canfield, Proc. Natl. Acad. Sci. U.S.A. **104**, 9960 (2007).

[4] S. Jia, N. Ni, S. L. Bud'ko, and P. C. Canfield, Phys. Rev. B **76**, 184410 (2007).

[5] E. D. Bauer, C. Wang, V. R. Fanelli, J. M. Lawrence, E. A. Goremychkin, N. R. de Souza, F. Ronning, J. D. Thompson, A.

V. Silhanek, V. Vildosola, A. M. Lobos, A. A. Aligia, S. Bobev, and J. L. Sarrao (unpublished).
[6] G. R. Stewart, Rev. Mod. Phys. **73**, 797 (2001).
[7] G. R. Stewart, Rev. Mod. Phys. **78**, 743 (2006).
[8] H. von Löhneysen, A. Rosch, M. Vojta, and P. Wölfle, Rev. Mod. Phys. **79**, 1015 (2007).
[9] G. R. Stewart, Rev. Sci. Instrum. **54**, 1 (1983); B. Andraka, G. Fraunberger, J. S. Kim, C. Quitmann, and G. R. Stewart, Phys. Rev. B **39**, 6420 (1989).
[10] H. Nakotte, K. Prokes, E. Brück, K. H. J. Buschow, F. R. de Boer, A. V. Andreev, M. C. Aronson, A. Lacerda, M. S. Torikachvili, R. A. Robinson, M. A. M. Bourke, and A. J. Schultz, Phys. Rev. B **54**, 12176 (1996).
[11] A. Rosch (private communication); see, for contrastingly relatively clean systems, A. Rosch, Phys. Rev. B **62**, 4945 (2000).
[12] J. S. Kim, J. Alwood, S. A. Getty, F. Sharifi, and G. R. Stewart, Phys. Rev. B **62**, 6986 (2000).
[13] Although paramagnons are ferromagnetic spin fluctuations, they are observed via $C \sim \delta T^3 \log T/T_{SF}$ behavior in compounds such as UPt_3 where [see G. R. Stewart, Rev. Mod. Phys. **56**, 755 (1984)] a plot of $1/\chi$ extrapolates to zero with a negative intercept on the temperature axis just as seen in Fig. 1 for $x=0.50$.