PHYSICAL REVIEW B VOLUME 54, NUMBER 2 1 JULY 1996-II

# Non-Fermi-liquid behavior in the specific heat over two decades of temperature in doped $UPt_3$

W. Trinkl, U. Weilnhammer, S. Corsépius, T. Schreiner, E.-W. Scheidt, and G. R. Stewart*
*Institut für Physik, Universität Augsburg, Memminger Strasse 6, D-86159 Augsburg, Germany*

(Received 1 February 1996; revised manuscript received 19 March 1996)

We have discovered, in contrast with most dopants $M$, that Hf (Zr) can be doped to quite high levels in $U_{1-x}M_xPt_3$, up to $x=0.15$ (0.30). Above 5% doping, the temperature dependence of the low-temperature upturn in $C/T$ (specific heat divided by temperature) changes from $T^3 \ln T$ (indicative of spin fluctuations) to $T \ln T$ [indicative of non-Fermi-liquid (NFL) behavior]. The specific heat NFL temperature dependence has been measured down to 0.1 K and is seen over more than two decades in temperature. The field dependence of the NFL specific heat is small, consistent with a quadrupolar moment model interpretation. A possible reason for the NFL behavior is discussed. [S0163-1829(96)07826-5]

## I. INTRODUCTION

Probably *the* most interesting heavy-fermion system remains today $UPt_3$, with its superconductivity coexistent[1–3] with magnetic fluctuations and magnetic order, its complicated superconducting phase diagram,[4–6] and its two superconducting transitions.[7–10] What makes $UPt_3$ so fascinating to study is superconductivity at 0.5 K in the presence of both antiferromagnetic correlations below 20 K and, separately, small moment ($0.02\mu_B$) antiferromagnetic order below $T_N=5$ K. The antiferromagnetic correlations/fluctuations below 20 K seen in neutron scattering experiments correspond to a peak in the dc magnetic susceptibility $\chi$ at 18 K; this peak in $\chi$ was in fact first thought[11] to be indicative of static, long-range antiferromagnetic order. The antiferromagnetic order in pure $UPt_3$ observed below 5 K was actually first discovered[3] after it became known that doped $UPt_3$ [$UPt_{3-x}Pd_x$ (Refs. 12,13) and $U_{1-x}Th_xPt_3$ (Refs. 13,14)] shows evidence for magnetism at $T\simeq 8$ K in the specific heat, $C$, in the magnetic susceptibility, and in the resistivity. These measurements[12–14] on doped samples led to neutron scattering experiments on $U_{0.95}Th_{0.05}Pt_3$ (Ref. 15) and $UPt_{3-x}Pd_x$.[16] Then, having found relatively strong ($\mu_{\mathrm{eff}}=0.6\mu_B$) local moment behavior in the doped samples at particular reciprocal lattice points, discovery of the small ($0.02\mu_B$) moment magnetism at the same points followed.[2]

The behavior of the doped $UPt_3$ samples had therefore direct and important bearing on understanding pure $UPt_3$. The connection between the $0.02\mu_B$ magnetic order and the superconductivity in $UPt_3$ remains a topic of continued experimental and theoretical interest, with the chances for an unconventional superconducting order parameter being a central focus.[17] The behavior of $U_{1-x}Th_xPt_3$ and $UPt_{3-x}Pd_x$ has also displayed some complexity, with the specific heat anomaly at $T_N$ disappearing[13] upon grinding as well as there being a report[18] of possible non-Fermi-liquid behavior in the low-temperature specific heat (i.e., $C$ diverges when $T\rightarrow 0$ as $T\ln T$) coexisting with magnetism in these materials. All other known systems (see, e.g., Refs. 19–25) display such a distinctive temperature dependence in $C$ *after* (but just[20,21]) magnetism has been suppressed, either via doping or pressure.[22] While trying to further understand the known magnetic behavior of doped $UPt_3$ ($U_{1-x}Th_xPt_3$ and $UPt_{3-x}M_x$, $M=$Pd, Au, are magnetic while $UPt_{3-x}Ir_x$ is not[26]) in samples of $U_{1-x}M_xPt_3$, $M=$Y, Sc,

TABLE I. Parameters for $U_{1-x}M_xPt_3$.

| | | Lattice parameters | | Specific heat fit parameters[a] | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | | $a$ (Å) | $c$ (Å) | $\gamma$ (mJ/mol $K^2$) | $\beta^*$ (mJ/mol $K^4$)[b] | $\delta$ (mJ/mol $K^6$)$\times 10^{-3}$ | $\epsilon$ (mJ/mol $K^4$) | $T_{\mathrm{SF}}$ (K) | $T_K$ (K) |
| $UPt_3$ | | 5.7534 | 4.9011 | 450 | −4.23 | −1.0 | 1.99 | 26.3 | |
| $U_{1-x}M_xPt_3$ | | | | | | | | | |
| M = Zr | $x=0.05$ | 5.7465 | 4.8865 | 400 (400) | −5.98 (−6.13) | −2.0 (−1.4) | 2.39 (2.40) | 17.3 (18.2) | |
| | 0.10 | 5.7411 | 4.8667 | 380 (385) | −8.17 (−9.68) | −3.5 (−3.9) | 3.31 (3.9) | 15.2 (14.9) | |
| | 0.15 | 5.7437 | 4.8425 | | | | | | 33 (40) |
| | 0.20 | 5.7327 | 4.8216 | | | | | | 32 (34) |
| | 0.25 | 5.7187 | 4.8072 | | | | | | 34 (38) |
| | 0.30 | 5.7294 | 4.7803 | | | | | | 34 (35) |
| $M=$Hf | $x=0.05$ | 5.7452 | 4.8837 | 400 (400) | −5.79 (−5.00) | −1.6 (−3.3) | 2.28 (1.76) | 18.3 (27.8) | |
| | 0.10 | 5.7350 | 4.8698 | | | | | | 29 (43) |
| | 0.15 | 5.7301 | 4.8580 | | | | | | 31 (40) |

[a] Numbers given are for fits up to 20 K; numbers in parentheses are from fits only up to 10 K.
[b] $\beta^*$ is a sum of the coefficients from the normal phonon term $\beta_{\mathrm{ph}}T^3$ and the spin fluctuation term $-\epsilon T^3 \ln T_{\mathrm{SF}}$.

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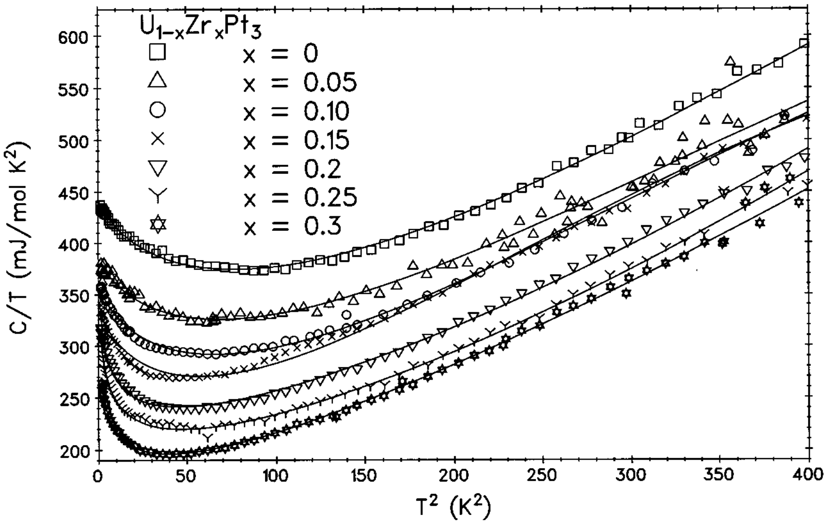

FIG. 1. Low-temperature specific heat divided by temperature vs temperature squared for $U_{1-x}Zr_xPt_3$, $0 \leq x \leq 0.30$; 1.4 K$\leq T \leq$20 K. The solid lines are fits to the data as discussed in the text using either an additional spin fluctuation term ($x \leq 0.15$) or a non-Fermi-liquid term ($x \geq 0.20$). Note the change from a gradual, spin-fluctuation-caused upturn in $C/T$ below 10 K ($C \propto T^3 \ln T$ term) for pure $UPt_3$ to a much steeper, NFL upturn ($C \propto T \ln T$ term) with increasing Zr doping.

Lu, Hf, and Zr, we discovered[27] that Hf and Zr were unusual. While only up to 7% of Y, Sc, Lu could be doped into the $UPt_3$ DO19 structure, 15% (30%) of Hf (Zr) doping remained single phase. We report here on the unusual behavior of these Hf and Zr samples; the results help to explain the unique coexistence[18] in doped $UPt_3$ of bulk magnetism and what is otherwise uniquely a ''nearly magnetic''-only phenomenon, i.e., $C \propto T \ln T$. The present work has also implications for understanding pure $UPt_3$ and the diverse magnetic character (antiferromagnetic fluctuations in one part of reciprocal space below 20 K and local small moment magnetism below 5 K in another part of reciprocal space) therein.

## II. RESULTS AND DISCUSSION

Samples of $U_{1-x}M_xPt_3$, $M$=Hf and Zr, were prepared by arc melting together stoichiometric quantities of the pure elements (see, e.g., Ref. 27). Samples were characterized by x-ray diffraction to check for second phases (typically visible if $\geq$ 5% of the total sample) and to track the decrease in the hexagonal $a$ and $c$ lattice parameters caused by the smaller size of Hf and Zr compared to U. As seen in Table I these lattice parameters behave approximately linearly (Vegard's law) with doping, which is a further useful method for determining the limit of solubility of dopants in the host lattice. Samples of 40% Zr and 20% Hf show $a=5.719$ Å, $c=4.777$ Å and $a=5.728$ Å, $c=4.855$ Å, respectively, indicating—based on the data in Table 1—a maximum solubility of 32% (15%) for Zr(Hf) in the $UPt_3$ lattice.

The specific heat divided by temperature, $C/T$, for $U_{1-x}Zr_xPt_3$, $0 \leq x \leq 0.3$, is shown in Fig. 1. One sees that the gradual upturn in $C/T$ below 10 K characteristic of the $T^3 \ln T$ spin fluctuation term in the specific heat of $UPt_3$ (Refs. 1,28) is, with increasing Zr, gradually replaced with a steeper increase in $C/T$ at low temperatures. (We find similar results for $U_{1-x}Hf_xPt_3$, $0 \leq x \leq 0.15$, discussed below.) In order to better follow the progression of this increase in $C/T$, Fig. 2 shows these data down to 0.4 K expanded at low temperatures, together with two fits to each set of data where $C = \gamma T + \beta T^3 + \delta T^5 + \epsilon f(T)$, with $f(T)$ either $T^3 \ln T$ (spin fluctuations[28]) or $T \ln T$ (non-Fermi-liquid behavior[19–25]). As clearly visible in Figs. 1 and 2, the $T^3 \ln T$ fit is clearly superior for $x=0$ and 0.05, and the $T \ln T$ fit is clearly superior for $x \geq 0.15$ for Zr. One might ask the following question: If the temperature region over which the data are fit is restricted to some $T(< 20$ K), over how narrow a temperature range must

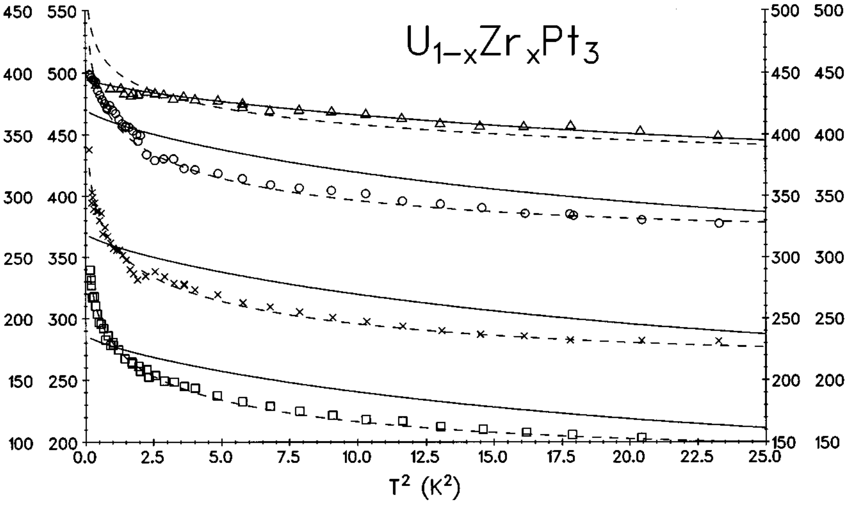

FIG. 2. Low-temperature specific heat divided by temperature vs temperature squared, $T \geq 0.4$ K on expanded scales [rightmost scale for $x=0$ (triangles), leftmost scale for $x=0.30$ (squares), middle left scale for $x=0.15$ (circles), and middle right scale for $0=0.25$ ($x$'s)]. The solid lines are fits to $C/T = \gamma + \beta T^2 + \delta T^4 + \epsilon T^2 \ln T$ over the entire temperature range (i.e., up to $T$=20 K), while the dashed lines are similar fits with the non-Fermi-liquid term $\ln T$ replacing the spin fluctuation $T^2 \ln T$ term. The change from the one behavior to the other is clear.

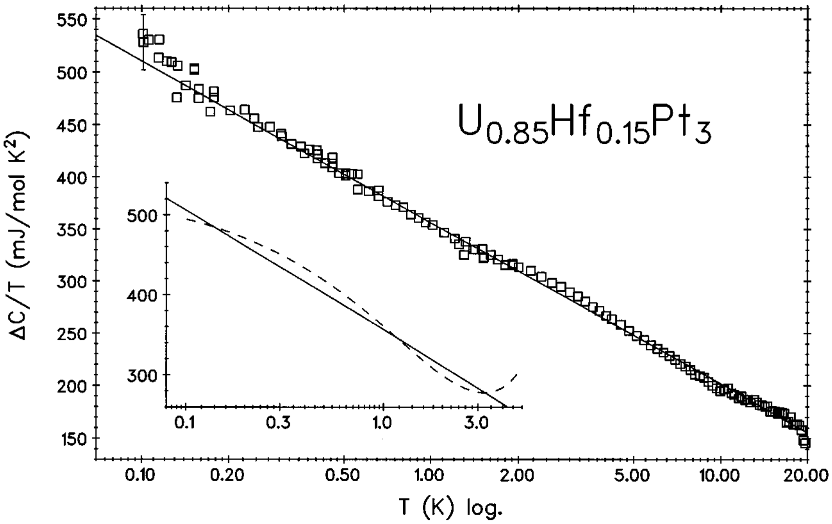

FIG. 3. The electronic contribution to the specific heat, $\Delta C$ (see text), divided by temperature vs $\ln T$ for $U_{0.85}Hf_{0.15}Pt_3$. (The $\Delta C/T$ data taken in the dilution refrigerator, 0.1 K$\leqslant T \leqslant$0.7 K, are linear vs $\ln T$ with the same slope as the data taken in the $^3$He apparatus for $T>0.4$ K, but—due to a calibration error still under investigation—are shifted slightly ($-4\%$) with respect to the higher-temperature data. The lowest-temperature data have been multiplied by a constant factor (1.04) to coincide with the higher-temperature data.) The error bar shown corresponds to $\pm 5\%$. Shown in the inset are the data—represented by the straight line—fitted to a $T^3\ln T$ fit (dashed line) restricted to $T\leqslant 5$ K. It is clear that such a fit is not a good representation of the data.

the data be limited so that the $T^3\ln T$ fits for $x \geqslant 0.15$ appear equally as good as the $T\ln T$ fits up to 20 K? The answer is that the higher concentration data do not really fit the $T^3\ln T$ fit (as is visually clear already in Figs. 1 and 2) unless the fit range is restricted to below 5 or 6 K. (Even then, using a fit with four parameters, there is an unphysical downward curvature in the fit at around 4 K.)

In order to verify the non-Fermi-liquid $T\ln T$ behavior in $C$ over as broad a temperature range as possible (which has also been requested by theorists[29]), we have extended our specific heat data down to 100 mK for $U_{0.75}Zr_{0.25}Pt_3$ and $U_{0.85}Hf_{0.15}Pt_3$. These data, plotted as $\Delta C/T$ vs $\ln T$ (where $\Delta C/T = C/T - \beta T^2 - \delta T^4$ i.e., $\Delta C$ is the electronic contribution) are shown in Figs. 3 and 4. Considering first the $U_{0.85}Hf_{0.15}Pt_3$ data of Fig. 3, we see that the $C/T$ data obey a $T\ln T$ behavior over more than two decades. (The inset to Fig. 3 shows clearly that a $T^3\ln T$ fit, even when restricted to $T\leqslant 5$ K, does not fit the data.) In order to show visually the progression from $T^3\ln T$ to $T\ln T$ behavior for the Hf data, just as shown for $U_{1-x}Zr_xPt_3$, $x\geqslant 0.15$, in Fig. 2, Fig. 5 is a plot of the $C/T$ data for $U_{1-x}Hf_xPt_3$, $0\leqslant x\leqslant 0.15$ with both the $T\ln T$ and $T^3\ln T$ fits. As for Zr doping, $x=0.10$ for Hf is a transition region, while for $x=0.15$ the fit to the Hf data down to 0.1 K *must* follow the $T\ln T$ dependence: Even if the fit range is limited to 0.1–5 K, the property of the spin fluctuation fit that it levels off at low temperature gives an unphysical waviness (see Fig. 3). Considering now the $T^3\ln T$ fit parameters for Zr, $x\leqslant 0.10$, and Hf, $x\leqslant 0.05$ (see Table I), one sees that these parameters behave systematically (independent of fit range) and that the spin fluctuation temperature $T_{SF}(=e^{(\beta_{ph}-\beta^*)/\epsilon})$ decreases monotonically upon doping.

Thus, we have discovered here two new materials where non-Fermi-liquid behavior is observed over more than two decades in temperature. (Previously the largest region of $T\ln T$ behavior in the specific heat was observed in $U_{0.2}Y_{0.8}Pd_3$, where $\Delta C \propto T\ln T$ between[19,30] 0.2 and 15 K, with[30] a distinct deviation at lower temperatures 0.05 $K\leqslant T\leqslant 0.2$ K.) From these $C/T$ vs $\ln T$ curves, one may calculate[31] a characteristic Kondo temperature $T_K$, via

$$T_K = \frac{-0.25R}{\delta},$$

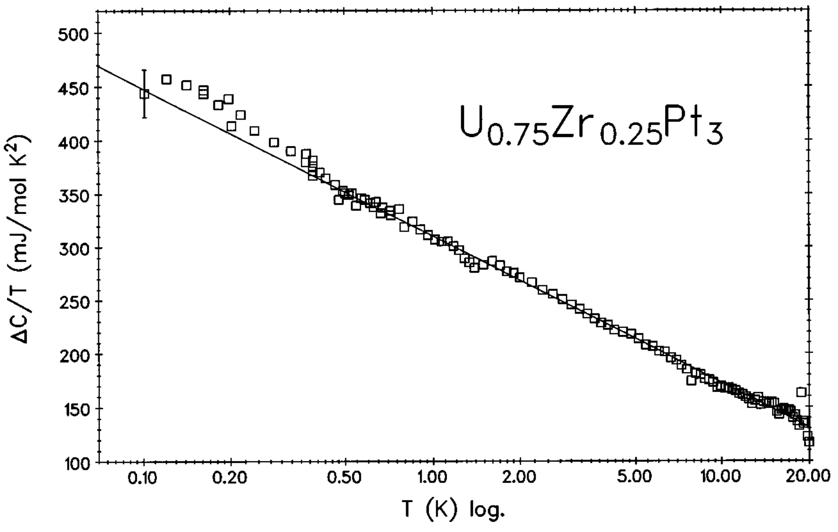

FIG. 4. Similar to Fig. 3, plotted here is the electronic term in the specific heat, $\Delta C$, divided by temperature for $U_{0.75}Zr_{0.25}Pt_3$ vs $\ln T$. (The dilution refrigerator data are scaled by 1.04 as discussed in Fig. 4.) The error bar shown corresponds to $\pm 5\%$.

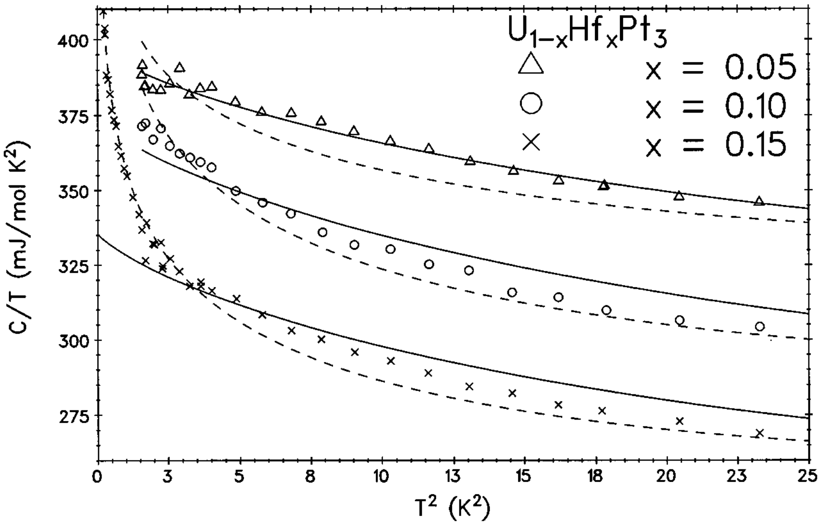

FIG. 5. Low-temperature specific heat divided by temperature vs temperature squared for $U_{1-x}Hf_xPt_3$, $0 \leqslant x \leqslant 0.15$. The solid lines are $T^3 \ln T$ fits and the dashed lines are $T \ln T$ as discussed in the text. Although the $T \ln T$ fit for $x=0.15$ does not pass perfectly through the data near $T=3.5$ K, below 2 K (see also Fig. 3) the $T \ln T$ fit is the only fit appropriate to the data.

where $R$ is the gas constant (8.3 J/mol) and $\delta$ is the slope of the $C/T$ vs $\ln T$ curve. As seen in Table I, $T_K$ for all five samples ($U_{1-x}M_xPt_3$, $x = 0.15$, 0.20, 0.25, 0.30 for Zr and $x=0.15$ for Hf) is, within a small deviation, the same. Thus, the already observed fact[32,33] that single-ion effects play an important role in the specific heat of $UPt_3$ seems—even in the rather concentrated doping regime considered—also to be the case here, since the characteristic temperature $T_K$ for the non-Fermi-liquid (NFL) behavior is concentration independent.

As has been done[19,21,23] in several NFL systems in order to try to distinguish between the various theoretical models, the specific heat of $U_{0.7}Zr_{0.3}Pt_3$ was measured in 0 and 13 T and is shown in Fig. 6. Within our measurement accuracy ($\pm 1\%$), there is no change in the specific heat with field. This result is in stark contrast with large ($\sim 30\%$ in 13 T) changes observed[19,21,23] in other systems showing $T \ln T$ temperature in the specific heat, and is consistent with the quadrupolar model of Cox.[34]

That this $C \propto T \ln T$ behavior occurs in, at least at first glance (Figs. 1–3), nonmagnetic $U_{1-x}M_xPt_3$, $M=$Hf, Zr, as was previously seen[13] in magnetically ordered $U_{1-x}Th_xPt_3$ and $UPt_{3-x}Pd_x$, presents us with obvious questions. Where do the interactions come from which are still temperature dependent as $T \to 0$ and therefore prevent the achieving of the Fermi-liquid ($C \propto \gamma T$) ground state in these systems?

Such interactions, based on the correlation[19–22] observed between NFL behavior and nearness to magnetism, might well be of magnetic character. From neutron scattering experiments[3] it is known, as mentioned above, that there are two kinds of magnetic behavior in $UPt_3$: the antiferromagnetic correlations (responsible for the peak in $\chi$ at 18 K) and the weak antiferromagnetic order below $T_N=5$ K (which is strengthened by Pd and Th doping into rather strong magnetic order beginning below $T_N \sim 8$ K). Thus, we need to consider the progression with Hf and Zr doping of both these behaviors in $UPt_3$.

First, let us consider the weak order at $\sim 5$ K. In the case of Pd and Th doping, this order is strengthened vis-á-vis pure $UPt_3$, but remains in the same temperature range, i.e., does not coincide with the NFL model[20–22] of temperature-dependent interactions suppressed to $T=0$. Since, as will be shown in a forthcoming work,[35] Hf- and Zr-doped $UPt_3$

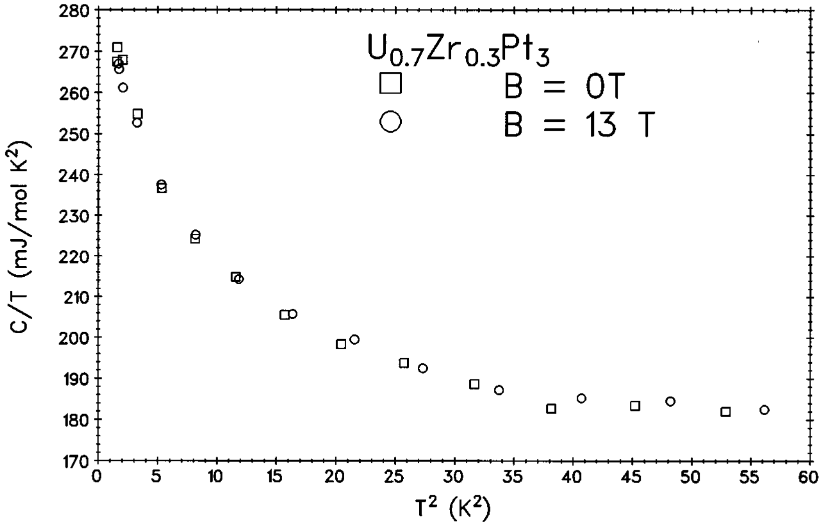

FIG. 6. Low-temperature specific heat divided by temperature vs temperature squared for $U_{0.7}Zr_{0.3}Pt_3$ in 0 (squares) and 13 T (circles) magnetic field.

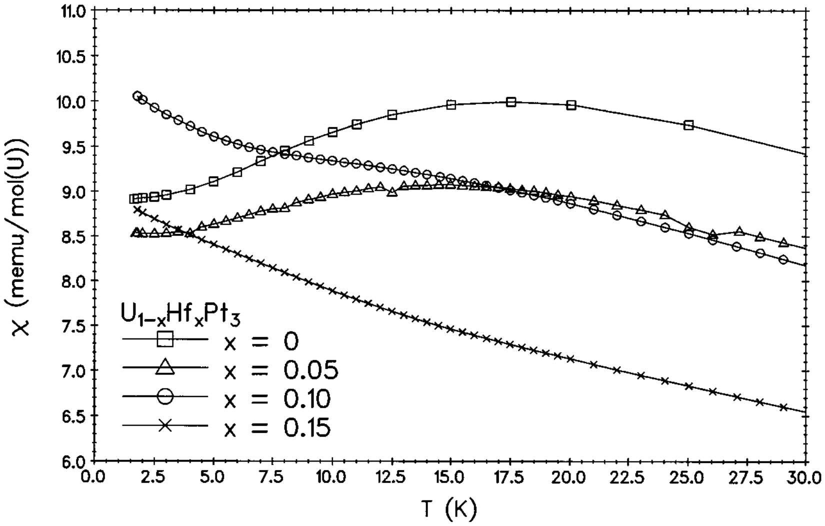

FIG. 7. Low-temperature magnetic susceptibility of $U_{1-x}Hf_xPt_x$, $0 \leqslant x \leqslant 0.15$. Note the suppression of the peak temperature with increasing $x$, as well as a broadening of the peak itself. By taking the first derivative of these $\chi(T)$ data, one can see the remanent peak at 8.5 K for $x = 0.10$. The temperature dependence of the $x = 0.15$ data shown here is neither a power law nor a ln$T$ behavior—the two usual $T$-dependences for a non-Fermi-liquid system—but rather can be fitted as a sum of *both* dependences.

show also evidence that the ~5 K magnetic interactions remained fixed in temperature, let us turn to a consideration of the peak in the susceptibility; see Fig. 7 for Hf-doped $UPt_3$. This peak is broadened and suppressed to lower temperatures (15 K for 5% Zr doping, 8.5 K for 10% Zr doping) for small doping levels, but is no longer visible, even as a slight change in slope, in $\chi$ for $x \geqslant 0.25$ (0.15) Zr (Hf) doping. Instead, the $\chi$ rises monotonically with decreasing temperature; see Fig. 7. Also the magnitude of the susceptibility for higher doping concentrations is suppressed, arguing that the correlations responsible for the 18 K peak in $\chi$ in pure $UPt_3$ have at least been shifted to lower temperatures, if not entirely suppressed. This suppression of the $\chi$ peak corresponds roughly with the suppression of the spin-fluctuation-derived $T^3 \ln T$ term in the specific heat[36] (see Fig. 5).

Thus, it is at least plausible that these antiferromagnetic correlations in $UPt_3$, upon suppression to lower temperatures, supply the mechanism for the non-Fermi-liquid $T \ln T$ dependence of $T$ for Hf and Zr doped $UPt_3$ shown in Figs. 3–4. Whether these correlations are in fact suppressed to $T = 0$ is, from our $\chi$ data, difficult to determine. Neutron scattering experiments, if possible to perform due to the unavoidable smearing out (visible also in Fig. 7) of the correlations, could resolve this question.

Thus, in conclusion, we have discovered non-Fermi-liquid behavior in Hf and Zr-doped $UPt_3$ via a $T$lnT term in C that extends over two decades in temperature. The mechanism that produces the $T \rightarrow 0$ temperature-dependent interactions necessary to prevent the formation of the Fermi-liquid ground state is possibly the suppressed antiferromagnetic correlations present in $UPt_3$ below 18 K.

## ACKNOWLEDGMENTS

Work at the University of Florida was supported by the U.S. Department of Energy, Contract No. DE-FG05-86ER45268.

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[*] Also at Dept. of Physics, University of Florida, Gainesville, Fl 32611.

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