$U_2Pt_{15}Si_7$: Competition between magnetism and the formation of a heavy-fermion ground state

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(Received 31 July 1995)

Via doping of $U_2Pt_{15}Si_7$ we have investigated the magnetic susceptibility and the anomaly in the low-temperature specific heat at 6.5 K. These data are compared with those for $CeAl_3$ (which also shows a low-temperature specific-heat anomaly) and UBe_{13} and $CeCu_2Si_2$ (which show a monotonic rise in C/T below 10 K). The results further the understanding on how the creation of a large electron effective mass ground state is linked to the magnetic degrees of freedom of the f electrons. The direct conversion of this entropy of disordered magnetic moments into an increase of C/T ($\propto m^*$) at low temperatures is observed in $U_2Pt_{15-x}Ni_xSi_7$ as Pt is replaced with Ni. In addition, the present results provide some insight into diverse phenomena associated with peaks in the specific heat in several heavy-fermion systems, e.g., in $CeAl_3$ where the long misunderstood peak at 0.35 K has recently been discovered to be incipient magnetism.

I. INTRODUCTION

Geibel *et al.*[1] discovered the existence of $U_2Pt_{15}Si_7$ in a face-centered-cubic structure, with an approximate U-U separation[2] of 5.9 Å. Rather than exhibiting magnetic order (a likely result due to the large $d_{\text{U-U}}$) or heavy-fermion, enhanced electron effective mass ($\propto$ large specific heat divided by temperature, C/T, as $T\rightarrow 0$) behavior, this new compound was found to exhibit a third behavior: a broad peak in the specific heat at 6.5 K coupled with a large magnetic susceptibility and ''Kondo-like'' resistivity. In the study of 5f-electron compounds with f-ion separations well above the Hill limit of 3.4 Å, the process by which the f electrons, hybridized with the neighboring ligand atom electrons, sometimes avoid a magnetically ordered ground state is not at all well understood. What is often put forward, see, e.g., Refs. 1 and 2, as a phenomenological description is that such systems are ''Kondo lattices,'' taken to mean that their properties are, in part, reminiscent of dilute Kondo alloys where theoretical description exists.[3]

The present work, using doping of $U_2Pt_{15}Si_7$, was undertaken to investigate this neither magnetic nor heavy-fermion intermediate behavior in the hopes of furthering understanding of the limiting, high effective mass case.

II. EXPERIMENT

In order to vary the U 5f-electron hybridization with its ligand atoms in $U_2Pt_{15}Si_7$ substitutions were made both for Pt and Si. In order to separate single-ion from correlation effects, certain selected samples had the U replaced by Th.

Thus, samples of $U_2Pt_{15-x}N_xSi_7$, $0\leqslant x\leqslant 4$, $U_2Pt_{13}Co_2Si_7$, $U_2Pt_{13}Ni_2Si_5Ge_2$, $U_{2-x}Th_xPt_{12}Ni_3Si_7$, $0\leqslant x\leqslant 2$, and $U_{0.4}Th_{1.6}Pt_{15}Si_7$ were prepared via melting together the appropriate amounts of pure starting material under a purified Ar atmosphere in an arc melter. Since the primary focus of the work developed to be $U_2Pt_{15}Si_7$ doped with Ni, samples of $Th_2Pt_{15}Si_7$ were also doped with Ni to serve as non-f-electron comparison compounds. All samples were annealed for 1 week at 800 °C and were, according to x-ray diffractometry, single phase. Lattice parameters are given in Table I. Ni substantially shrinks the cubic lattice (by 2% for $U_2Pt_{11}Ni_4Si_7$), while Th doping expands the lattice slightly. Attempts to prepare single-phase samples with either Cu or Au replacing Pt were unsuccessful.

III. RESULTS AND DISCUSSIONS

The low-temperature susceptibility data for $U_2Pt_{15-x}Ni_xSi_7$, $0\leqslant x\leqslant 4$, are shown in Fig. 1, with χ(1.8 K), which ranges between 90 memu/U mol ($x=0$) and 55 memu/U mol ($x=4$), tabulated in Table I. [Typical values for χ(1.8

TABLE I. Lattice parameter a_0, magnetic susceptibility χ(1.8 K) and amount of saturation in magnetization vs field (M vs H sat). The saturation is here defined as the difference of the linear magnetization at low fields extrapolated to 7 T and the measured M(7 T) divided by the extrapolated value, i.e., ($[M(7\ \text{T})_{\text{extra}}-M(7\ \text{T})_{\text{meas}}]/M(7\ \text{T})_{\text{extra}}$).

	a_0(Å)	χ(1.8 K) (memu/U mol)	M vs H sat. (%)
$U_2Pt_{15}Si_7$	16.810	90	40
$U_2Pt_{14}NiSi_7$	16.735	67	23
$U_2Pt_{13}Ni_2Si_7$	16.666	65	22
$U_2Pt_{12}Ni_3Si_7$	16.615	60	21
$U_2Pt_{11}Ni_4Si_7$	16.576	55	18
$U_{1.6}Th_{0.4}Pt_{12}Ni_3Si_7$	16.622	56	20
$U_{0.4}Th_{1.6}Pt_{12}Ni_3Si_7$	16.651	44	16
$U_{0.2}Th_{1.8}Pt_{12}Ni_3Si_7$	16.670	42	19
$U_{0.1}Th_{1.9}Pt_{12}Ni_3Si_7$	16.671	48	27
$U_{0.05}Th_{1.95}Pt_{12}Ni_3Si_7$	16.671	48	25
$U_2Pt_{13}Ni_2Si_5Ge_2$	16.746	55	18

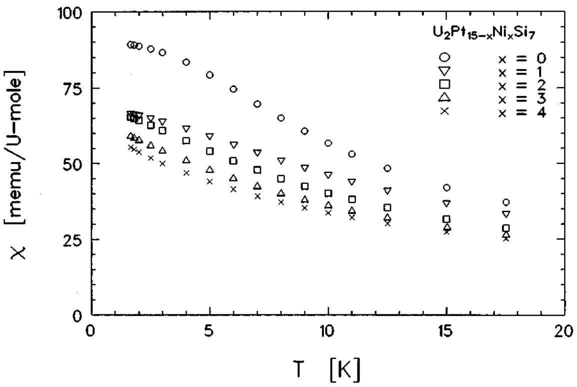

FIG. 1. Low-temperature magnetic susceptibility as a function of temperature for $U_2Pt_{15-x}Ni_xSi_7$, $0\leqslant x\leqslant 4$. Note the rapid decrease in $\chi(T\rightarrow 0)$ upon first doping with Ni, followed by a more gradual decrease with increasing Ni doping.

K) for heavy-fermion systems range from 6.9 memu for UPt_3 to 36 memu for $CeAl_3$.) Clearly, Ni doping suppresses the low-temperature χ monotonically. A further measurement useful for qualitatively determining the amount of magnetic character of a sample is that of the magnetization as a function of field at low (here 2 K) temperature. At low fields M vs H is linear and then as the field is increased, a point is reached where the M vs H curve bends over, i.e., rises less steeply. Heavy-fermion systems in general have linear M vs H behaviors to quite high fields [e.g., 24 T for UBe_{13} (Ref. 4)], while compounds with more magnetic character display a tendency towards saturation at lower fields. The $U_2Pt_{15-x}Ni_xSi_7$ samples all display a saturation, which, however, decreases with increasing x (see Table I) consistent with the observed decrease in χ(1.8 K) also observed with increasing x.

The low-temperature specific heat divided by temperature vs temperature for $U_2Pt_{15-x}Ni_xSi_7$, $0\leqslant x\leqslant 4$, is shown in Fig. 2. The broad, rather ill-defined anomaly in C/T around 6.5 K, which was also observed in Refs. 1 and 2 becomes sharper and is shifted to lower temperatures with increasing Ni doping. In order to focus on purely the $5f$-electron specific heat, we have measured also $Th_2Pt_{15-x}Ni_xSi_7$, $0\leqslant x\leqslant 4$, and subtracted these results from the data in Fig. 2. The resulting curves for C^{5f}/T vs T then all lie atop one another for $T>8$ K up to our upper temperature of measurement (20 K), and decrease with increasing temperature only gradually. This nearly constant behavior of C^{5f}/T, $8<T<20$ K, is due to the presence at higher temperatures of a Schottky anomaly, due to crystal-field split levels, as determined in Ref. 2. The independence of Ni concentration implies that the Schottky anomaly is unaffected by the Ni doping. Thus, in order to concentrate on the low-temperature electronic behavior associated with the peak in C/T, we plot $(C^{5f}/T-C^{\mathrm{Schottky}}/T)$ vs T in Fig. 3.

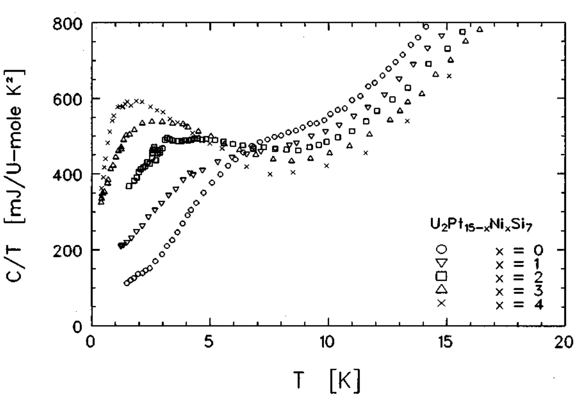

FIG. 2. Low-temperature specific heat, C, divided by temperature, T, vs T for $U_2Pt_{15-x}Ni_xSi_7$, normalized per U mol. Note that any peak in a plot of C/T is also a peak in a plot of C; in order to be able to focus both on the anomaly in C and on the possible behavior of m^* ($\propto C/T$ as $T\rightarrow 0$), the data are plotted here as C/T.

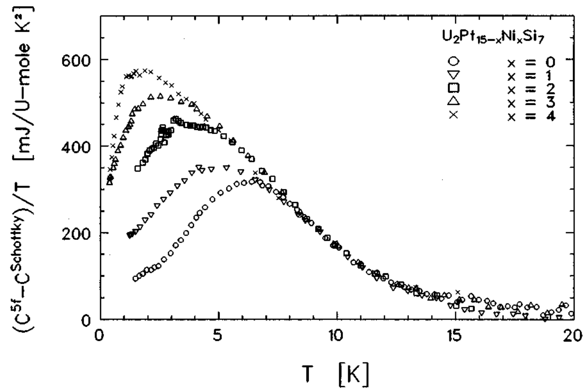

FIG. 3. Here are plotted the data from Fig. 2, $C/T(U_2Pt_{15-x}Ni_xSi_7)$, with the respective Th homologies, $C/T(Th_2Pt_{15-x}Ni_xSi_7)$, and the common crystal-field anomaly (Ref. 2) level splitting δ=55 K, subtracted off. What is surprising in this figure is that increasing Ni content has the effect of continuing to even lower temperature the heavy-fermion-like rise in C/T observed above 8 K for undoped $U_2Pt_{15}Si_7$, i.e., the anomaly *per se* is not just simply shifted to lower T.

Figure 3 shows that the anomaly in the specific heat of $U_2Pt_{15}Si_7$ is of an unusual character. The causes that one usually considers when discussing broad specific-heat anomalies (e.g., Schottky, Kondo, spin glass) all have a characteristic energy (δ=level splitting, T_K=Kondo temperature, T_F=spin-freezing temperature). This energy, when changed by some influence or other, determines a new position for the maximum in the anomaly, i.e., changes in these characteristic energies lead to *shifts* in the anomalies, not extension of the anomaly in the lower temperature direction with no change at all in the high-temperature half of the anomaly. Thus, from Fig. 3 what appears to be the case for $U_2P_{15-x}Ni_xSi_7$ is that a process is at work below 20 K which increases C/T with decreasing temperature. At some temperature, dependent within certain limits on Ni concentration, this process fails, or goes over into another process which leads to a decrease in C/T (and of course in C itself). What are possible explanations for this crossover behavior?

Since we are dealing with U atoms much further apart (5.9 vs 3.4 Å) than where, unless hybridization prohibits, local magnetic moments form, our first thoughts turn to investigating the magnetic character of the anomalies in Fig. 3. First, is there some correlation between where the respective maxima of the anomalies occur and some feature in the magnetic susceptibility, Fig. 1? The χ data in Fig. 1 show no peaks in χ vs T, however the data do show an inflection

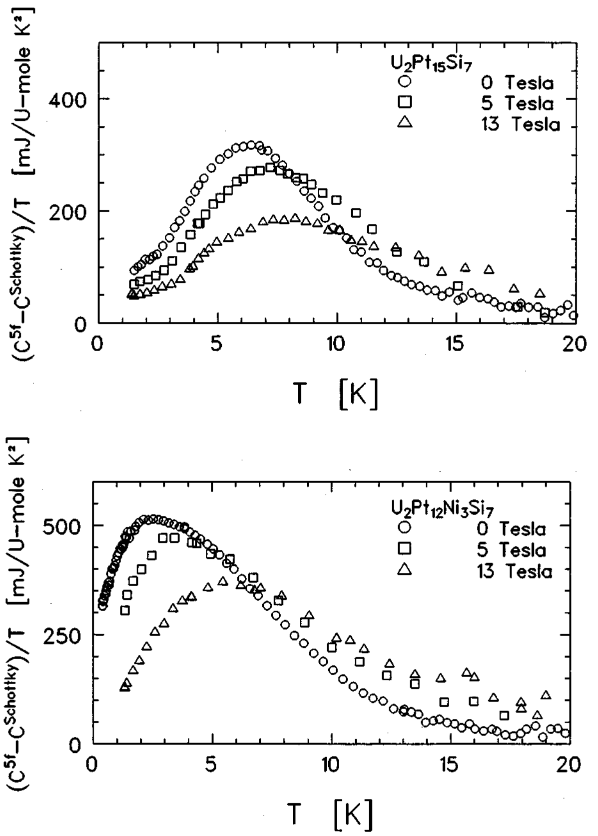

FIG. 4. The specific heat so normalized as in Fig. 3 ($C-C^{Th}-C^{Schottky}$, δ=55 K) divided by temperature vs temperature in 0, 5, and 13 T magnetic field for $U_2Pt_{15}Si_7$ and $U_2Pt_{12}Ni_3Si_7$.

point. (This might be possible evidence for a small peak in χ vs T overwhelmed by large contributions to χ from other sources.) The temperature at which the inflection point occurs matches fairly well the temperature of the maximum in C/T for a given $U_2Pt_{15-x}Ni_xSi_7$ sample. As discussed above and in Table I, both the magnitude of χ(1.8 K) and the amount of saturation in M vs H also decrease monotonically with increasing Ni doping. These three observations are then consistent with a lessening of the magnetic character of the $5f$ electrons with increasing x in $U_2Pt_{15-x}Ni_xSi_7$.

What is the field dependence of these specific-heat anomalies? Figure 4 shows the specific heat in 0, 5, and 13 T of $U_2Pt_{15}Si_7$ and $U_2Pt_{12}Ni_3Si_7$. The magnetic field strongly affects both anomalies, shifting them upwards in temperature and depressing C^{max}. The depression of the specific heat in 13 T is about 25% larger (expressed as $\Delta C/C^{max}$) in $U_2Pt_{15}Si_7$ as in $U_2Pt_{12}Ni_3Si_7$, i.e., these $C(H)$ data not only confirm at least a partial magnetic character of the specific-heat anomaly but also show that the anomaly for $x=3$ displays less field dependence than for pure $U_2Pt_{15}Si_7$. (It is interesting to note that the inflection point in χ vs T can still be seen in measurements of χ in 5 T for $U_2Pt_{15}Si_7$, and roughly corresponds in temperature with the observed position of the specific-heat peak in 5 T in Fig. 4.)

We turn now to a brief discussion of what was learned from the measurements of $U_2Pt_{13}Co_2Si_7$, $U_2Pt_{13}Ni_2Si_5Ge_2$, and $U_{2-x}Th_xPt_{12}Ni_3Si_7$ in order to present as complete a picture as possible. Surprisingly, Co, in contrast to Ni, totally

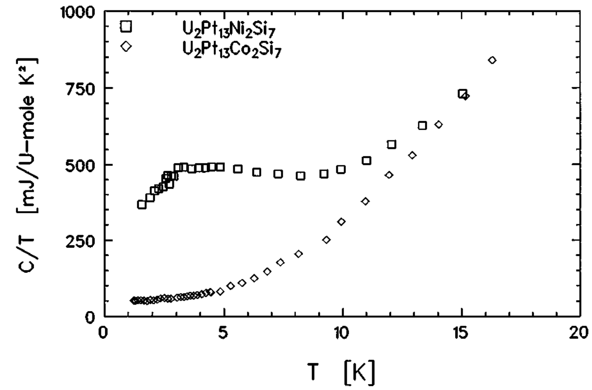

FIG. 5. Specific heat divided by temperature vs temperature for $U_2Pt_{13}Ni_2Si_7$ and $U_2Pt_{13}Co_2Si_7$.

suppresses the anomaly in the specific heat, see Fig. 5. (Susceptibility is not reported due to a slight ferromagnetic parasitic phase of, presumably, Co.) One sees that while Ni ($3d^8 4s^2$) replacement of Pt ($5d^9 6s^1$) in $U_2Pt_{15}Si_7$ continues to upturn in C/T to sin temperature (i.e., C^{max}/T increases), Co ($3d^7 4s^2$) doping destroys the anomaly entirely, leaving a γ of 45 mJ/U mol K^2 and a higher-temperature specific heat ($=C^{Schottky}+C^{phonon}$) that is similar to that of $U_2Pt_{13}Ni_2Si_7$. Thus, the smaller number of d electrons in Co vs Ni is clearly important in the hybridization with U $5f$ electrons. Doping with Ge leads to results for both C and χ equivalent to doping with Ni, i.e., the specific heat and susceptibility for $U_2Pt_{11}Ni_4Si_7$ are essentially the same as for $U_2Pt_{13}Ni_2Si_5Ge_2$. Finally our results for $U_2Pt_{12}Ni_3Si_7$ with the U diluted with Th show a slight (10–25%) decrease in χ(1.8 K)/U mol with increasing Th content, an essentially constant amount of saturation in M vs H (see Table I), and a shifting of the anomaly in C/T such that for $U_{2-x}Th_xPt_{12}Ni_3Si_7$, $x\geqslant1.6$, the specific heat per U mol is approximately that of pure $U_2Pt_{15}Si_7$, see Fig. 6. In other words, the continuation to lower temperatures of the upturn in C/T above 8 K in $U_2Pt_{15}Si_7$ caused by increasing Ni content, see Fig. 3, is a

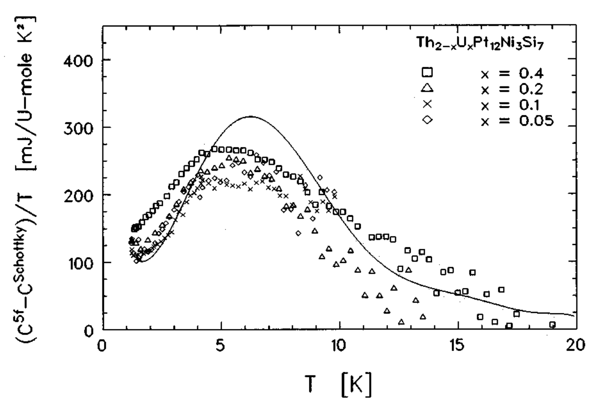

FIG. 6. Specific heat C of $U_{2-x}Th_xPt_{12}Si_7$ for $x\geqslant1.6$ minus $C(Th_2Pt_{12}Ni_3Si_7)$ minus $C^{Schottky}$, all divided by temperature vs temperature, i.e., the data are normalized as in Figs. 3 and 4. The solid line represents the data for pure $U_2Pt_{15}Si_7$ and is, within the scatter (25%) of the data themselves, similar to the dilute $U_{2-x}Th_xPt_{12}Ni_3Si_7$ data shown.

correlation effect which is destroyed by dilution of the U atoms. In order to persue this point for the pure compound, $U_2Pt_{15}Si_7$, we also prepared $U_{0.4}Th_{1.6}Pt_{15}Si_7$. The low-temperature specific heat shows that, normalized per U mol, the anomaly is essentially suppressed in the diluted sample, i.e., the peak in the specific heat for $U_2Pt_{15}Si_7$ appears also to be a correlation effect, as proposed in Ref. 2.

Thus, $U_2Pt_{15}Si_7$ has an anomaly in the specific heat that has at least some magnetic character (inflection point in χ, ΔC is field dependent). This anomaly is strongly dependent on U $5f$-electron hybridization with both the Pt and Si sites. (Ni shifts the peak in C/T to lower temperatures while extending the upturn in C/T above $T^{\max}$, Co destroys the specific-heat anomaly; Ge acts the same as Ni.) The anomaly in C in $U_2Pt_{15}Si_7$ as well as in Ni-doped samples appears to be a correlation effect. One possible way to relate all of these facts together is to, as proposed theoretically in Ref. 5, consider the heavy-fermion ground state as the result of the interplay between intersite spin correlations and on-site "Kondo-type" screening effects. Thus, a completely heavy-fermion ground state is a result of "a transformation in the course of which the entropy of disordered local magnetic moments is effectively transferred to the itinerant electrons" (Ref. 5), or a "spin-liquid" state.

Regardless of whether this theory is exactly correct, it is certainly the case in heavy-fermion systems that some mechanism (or mechanisms) converts the entropy ($=\int C/T dT$) present in the f-electron spin degrees of freedom into an increase in C/T ($\propto m^*$) at low temperatures. We propose that $U_2Pt_{15}Si_7$ is the first recognizable example of the "between" case of a system where the f-spin entropy is being converted as T is lowered into a high effective mass ground state (upturn in C/T for $8<T<15$ K, Fig. 3), but that the conversion process fails at a finite T, leading to a peak in C/T with a complex magnetic character. (One example of the unusual magnetic nature of the anomaly is that, although the high temperature $1/\chi$ vs T Curie-Weiss behavior exhibits antiferromagnetic correlations, magnetic field shifts the anomaly to *higher* temperatures, Fig. 4.)

Thus, within this picture Ni acts to support the conversion process to lower temperature, leading to a further monotonic increase in C/T from $\sim$300 mJ/U mol K^2 to almost 600 mJ/U mol K^2 for $U_2Pt_{11}Ni_4Si_7$. That this upturn in C/T, based on Th doping, is a correlation effect is perfectly consistent with known results on, e.g., UBe_{13}, where 60% of the low-temperature C/T value of 900 mJ/U mol K^2 (Ref. 6) is a correlation effect which is destroyed by dilution. That the creation of the upturn in C/T is sensitive to hybridization ($U_2Pt_{13}Co_2Si_7$ vs $U_2Pt_{13}Ni_2Si_7$, Fig. 5) is also consistent with known heavy-fermion systems, e.g., UPt_5 has[7,8] a $C/T(T\rightarrow 0)$ of $\sim$85 mJ/mol K^2 vs 700 mJ/mol K^2 for UPt_4Au. That f-spin degrees of freedom can create an anomaly in C which, upon doping, is converted into an increasing rise in C/T as temperature is lowered has been observed[9] in pure UBe_{13} (anomaly in C at 2.5 K) upon doping with, e.g., Th [$U_{0.97}Th_{0.03}Be_{13}$ has[9,10] no anomaly at 2.5 K, and a $C/T(T\rightarrow 0)$ of 2300 mJ/mol K^2].

In addition to the above arguments, we would like to point out that the $U_2Pt_{15}Si_7$ data, whatever the correct explanation for the cause of the anomaly in C, are comparable to those of known heavy-fermion systems. In Fig. 7, we show the upturn

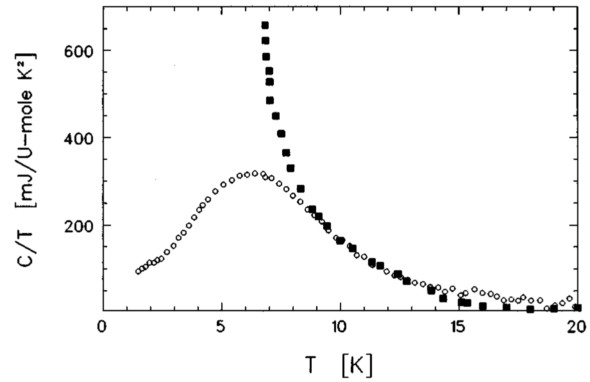

FIG. 7. Specific heat divided by temperature (normalized by subtracting C/T for $Th_2Pt_{15}Si_7$ and C/T for the Schottky anomaly, as in Fig. 3) for $U_2Pt_{15}Si_7$ and for $CeCu_2Si_2$. The latter data, which are essentially equivalent (Ref. 13) to those of UBe_{13}, have had the high-temperature ($T>20$ K) extrapolation of C/T (Ref. 13) subtracted in order to focus on the change with temperature in the low-temperature electronic contribution. Further, in order to bring the two data sets on top of one another for comparison, the $CeCu_2Si_2$ data were shifted upwards in temperature by 5 K.

in the C/T data for $CeCu_2Si_2$ shifted in temperature and plotted with the specific-heat of $U_2Pt_{15}Si_7$. As may be seen in the figure from this comparison, the temperature dependence of the increase in C/T with decreasing temperature in the known heavy-fermion system $CeCu_2Si_2$ is qualitatively similar to that seen above the peak in $U_2Pt_{15}Si_7$. This is at least supportive of the idea that the rise in C/T below 20 K in $U_2Pt_{15}Si_7$ (and the related $U_2Pt_{15-x}Ni_xSi_7$) may be a nascent creation of a heavy-fermion ground state which fails below 8 K. In Fig. 8, we show the susceptibility data of $U_2Pt_{11}Ni_4Si_7$ together with the data for $CeCu_2Si_2$. That scaling the temperature for the $CeCu_2Si_2$ χ data by a factor of 10 and the magnitude of the χ data by the inverse of the *same* factor ($\chi \propto 1/T_{\mathrm{characteristic}}$) gives such perfect agreement with the $U_2Pt_{11}Ni_4Si_7$ data is noteworthy. Based on this comparison,

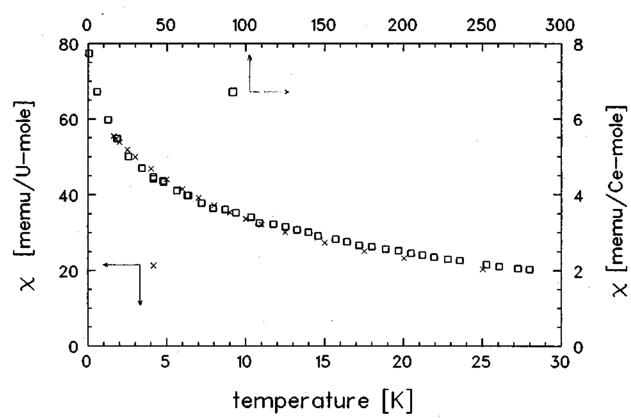

FIG. 8. Magnetic susceptibility vs temperature for $U_2Pt_{11}Ni_4Si_7$ ($\times$) and $CeCu_2Si_2$ (squares). The susceptibility of the latter is approximately a factor of 10 smaller at a given temperature T, than that for $U_2Pt_{11}Ni_4Si_7$ at $T/10$, i.e., the two sets of data scale quite well. $U_2Pt_{11}Ni_4Si_7$ was chosen for the comparison because the inflection point is depressed below the lowest temperature of measurement, see Fig. 1.

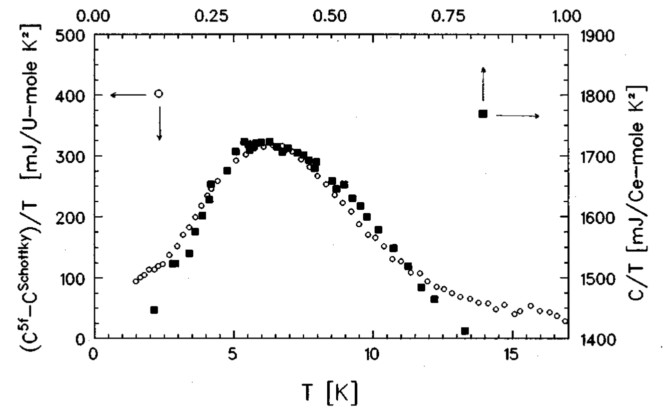

FIG. 9. Specific heat divided by temperature for $U_2Pt_{15}Si_7$, normalized as in Fig. 3, vs the data for the peak in C/T for $CeAl_3$. The peak in $CeAl_3$ sits on top of a large upturn in C/T corresponding to the creation of a heavy-fermion ground state. In order to qualitatively compare the two sets of data, we have simply shifted the y axis for the $CeAl_3$ data so that the maxima coincide, without any scaling of C. In order to bring the two peaks onto each other, the horizontal temperature scales for the two sets of data differ by the ratio of the respective $T(C^{max})$, i.e., by ~6.5/0.38.

the (unknown) mechanism for conversion of the f-spin degrees of freedom into an enhanced effective mass is more effective for $CeCu_2Si_2$ than for the $U_2Pt_{15-x}Ni_xSi_7$ compounds, leading to a less complete transformation of the spin entropy in the latter into m^*, or C/T entropy.

A second heavy-fermion system to consider is $CeAl_3$ where it has long been known that the increase in C/T as $T\rightarrow 0$ in this system is interrupted by a peak in C/T with $T^{max}=0.35$ K. Figure 9 shows this peak in C/T for $CeAl_3$ overlaid on our data for $U_2Pt_{15}Si_7$. Although this is only at best a qualitative comparison, the agreement of the data in Fig. 9 is not uncompelling. An additional important point that binds together the herein reported magnetic character of the anomaly in $U_2Pt_{15}Si_7$ with this anomaly in $CeAl_3$ is the recent discovery[11,12] that the anomaly in $CeAl_3$ is evidently incipient magnetism. That is, upon doping either on the Ce site[11] with 5–20% La, or on the Al site[12] with various dopants, the anomaly is monotonically shifted to higher T and becomes manifestly a bona fide fully magnetic transition. The speculation is[11,12] that if this incipient magnetism peak in C/T of $CeAl_3$ could be suppressed (as can[9] the small peak in C in UBe_{13} via Th doping), then C/T would continue rising as T is lowered just as we observe in Ni-doped $U_2\,Pt_{15}Si_7$.

In conclusion, the anomaly observed in the specific heat at 6.5 K in $U_2Pt_{15}Si_7$ may be argued to be the interrupted or incomplete precursor to a heavy-fermion ground state. Examples of such systems should exist, and further characterization of such "between" cases[14] could give further insight into the mechanism responsible for the transformation of f-electron spin entropy into large effective masses at low temperatures.

ACKNOWLEDGMENTS

Work at Florida was performed under the auspices of the U.S. Department of Energy, Grant No. DE-FG05-86ER45268.

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[14] Another possible example of the behavior reported on here for $U_2Pt_{15-x}Ni_xSi_7$ may possibly be found in UPd_2Sn [C. Rossel, M. B. Torikachvili, J. W. Chen, and M. B. Maple, Solid State Commun. **60**, 563 (1986)]. As pointed out in Ref. 2, the peak in C/T for $U_2Pt_{15}Si_7$ is similar to the one observed in UPd_2Sn centered at 10 K. In fact, we are able to scale the two peaks to fairly well coincide with one another. However, materials preparation in UPd_2Sn, as well as doping thereof appears in a preliminary study to be difficult.