Non-Fermi-liquid behavior and spin fluctuations in doped UAl_2

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Using the canonical spin-fluctuation system UAl_2 as a starting point, via negative chemical pressure (doping with Y) we have expanded $d_{U\text{-}U}$ in a system known to be near the Hill limit of f-electron localization, and characterized the samples via resistivity, magnetic susceptibility, and specific-heat measurements. All system parameters, including magnetic susceptibility, specific heat γ ($\equiv C/T$ $\lim_{T\to 0}$), and spin-fluctuation temperature, behave monotonically. For $U_{1-x}Y_xAl_2$, $0.30 \leqslant x \leqslant 0.70$, spin-glass behavior is found with $T_f \simeq 5.1 \pm 0.5$ K. This spin-glass behavior weakens (T_f sinks, smaller magnetic signature, no specific-heat anomaly) for $x \geqslant 0.75$ while, at the same time, the spin-fluctuation $T^3 \ln T$ term also gradually disappears from the specific heat. For $x \geqslant 0.875$, a non-Fermi-liquid (nFl) $\ln T$ term is found in the low temperature C/T. This new, perhaps equilibrium, ground state persists upon further dilution of the U ions with Y. Thus, we report on the evolution of nFl behavior in the neighborhood of a spin-glass ground state but, indeed, directly out of a yet weaker form of magnetism than heretofore reported, that of spin fluctuations. [S0163-1829(97)03102-0]

INTRODUCTION

The field of non-Fermi-liquid (nFl) systems has grown quite rapidly since the discovery[1,2] of nFl behavior in $U_{0.2}Y_{0.8}Pd_3$. In the intervening time, a number of further systems have been discovered which has led to the (essentially) general rule that this nFl behavior ($C/T \propto \ln T$, — i.e., C/T is divergent at low T, $\chi \propto \ln T$ or T^{-a}, $\rho \propto T$) is found in the vicinity of magnetism.[3] The type of magnetism that must be suppressed[4] below $T=0$ includes antiferromagnetism,[3,5,6] spin-glass behavior,[7] as well as (recently discovered) ferromagnetism.[8] Heavy fermion systems with their, by definition, already large low-temperature C/T values have been fruitful starting points for investigation.

The current work presents a search for nFl behavior in $U_{1-x}Y_xAl_2$ where the starting UAl_2 compound exhibits a yet weaker type of magnetic behavior, that of spin fluctuations, than previously found necessary to produce nFl behavior. In addition, in contrast to all the nFl U systems found to date, where the distance between U lattice sites has all been well over the Hill limit[9] of 3.4 Å, $d_{U\text{-}U}$ in UAl_2 is almost exactly equal to 3.4 Å.

UAl_2, and specifically doping with Y, were chosen based on several known experimental facts. UAl_2 has been studied extensively via doping studies, including work on $U_{1-x}M_xAl_2$, $M=$La,[10] Th,[11] and Pu,[12] and our recent work[13] on $M=$Ti, Zr, Hf, and Sc. Wire[14] has also done phase stability and magnetic-susceptibility studies on many $U_{1-x}M_xAl_2$ compounds. Doping on the non-f-site has been carried out somewhat less rigorously, see, e.g., Ref. 15 for Ga, Si, and Mn doping of the Al site. Two general facts useful for planning the current study may be summarized from the numerous previous studies. First, there exists a miscibility gap[14] in many $U_{1-x}M_xAl_2$ pseudobinary solutions such that, even though a given MAl_2 exists in the same cubic $cF24$ ($C-15$) structure as UAl_2, for approximately $0.3 < x < 0.7$ there is a two-phase region.

The inability to continuously trace the evolution of spin fluctuations with doping in $U_{1-x}La_xAl_2$, where there is a miscibility gap, led to the interesting result of Ref. 16 (that at $U_{0.1}La_{0.9}Al_2$ spin-glass and spin-fluctuation behavior apparently coexist[17]) evolving out of a miscibility gap region that is inaccessible to trying to trace the root causes.

Thus, based on the work of Wire,[14] $U_{1-x}Y_xAl_2$, for which no miscibility gap is found, was deemed a good candidate for trying to trace the suppression of the spin fluctuations with dilution.

The second conclusion that can be won from the existing results (see also Ref. 13) is that size seems to be a deciding factor[15] for determining the specific-heat enhancement due to spin fluctuations, with smaller dopants [e.g., Zr (Ref. 13) or Si (Ref. 15)] rapidly suppressing the spin fluctuations, while larger dopants, like La (Ref. 10) or Pu,[12] enhance the low-temperature specific heat and magnetic susceptibility substantially.

With the exception of several recently published γ values[18] for very dilute $U_{1-x}Y_xAl_2$, $x \geqslant 0.9$ ($\gamma \equiv \lim_{T\to 0} C/T$) (no curves of the specific-heat vs T were published), no specific heat data exist for Y-doped UAl_2. However, Y is larger than U in UAl_2 [$d_{M-M}(YAl_2) = 3.403$ Å vs 3.363 Å for UAl_2], and the low-temperature magnetic susceptibility, χ, is known[14,19] to grow larger than that of UAl_2 upon doping with Y. Thus, based on the considerations of whether a miscibility gap exists or not and the expectation, based on the increased χ, that Y doping will enhance the low-temperature spin fluctuations and specific heat, the present work was performed on $U_{1-x}Y_xAl_2$ in order to investigate the possibility of spin fluctuations as a basis for nFl behavior.

RESULTS AND DISCUSSION

Samples of $U_{1-x}Y_xAl_2$, $0 \leqslant x \leqslant 1.0$, were prepared by arc-melting together the constituent metals. All samples were single phase as determined by x-ray-diffraction studies, with the cubic lattice parameter (see Table I) varying linearly

TABLE I. Parameters for $U_{1-x}Y_xAl_2$.

$x=$	0	0.1	0.2	0.3	0.35	0.4	0.5	0.7	0.75	0.8	0.85	0.875	0.9	0.95
a_0 (Å)	7.766	7.775	7.788	7.800	7.804	7.809	7.818	7.838	7.844	7.846	7.851	7.854	7.856	7.858
χ (1.7 K) (memu/U mole)	4.6	5.9	13	22	28	36	56	88	77	70	63	50	55	24
γ [a] (mJ/U mole K^2)	145	190		380	390	460	490	430	380	470				
	(140)	(185)		(365)	(345)	(355)	(355)	(365)	(380)	(460)	(445)	(460)	(460) [b]	(305) [b]
β [c] (mJ/mole K^4)	-3.80	-2.58		-6.65	-5.65	-6.55	-5.61	-1.65	-1.26	-1.92		0.096	0.091	0.056
ϵ [c] (10^{-3} mJ/mole K^6)	-1.87	-1.25		-2.35	-1.72	-2.04	-1.59	-0.21	-0.20	-0.53		0.13	0.11	0.10
δ [c] (mJ/mole K^4)	1.67	1.15		2.52	2.09	2.42	2.04	0.58	0.46	0.71		-17.8	- 15.3	- 3.36
T_{SF} (K)	10.5	10.3		14.5	15.5	16.2	16.2	19.0	17.8	15.7				
T_f [d] (K)			2.2	4.6	4.9	5.3	5.6	5.3	5.1	4.5	3.2	2.3	1.8	
T_f [e] (K)				(5.4)	5.9	6.1	6.6	6.4	(6.3)	(5.3)	(4.2)	(3.4)		

[a]Values taken from spin fluctuation fit; values in parentheses are as measured at 1.3 K.

[b]As mentioned in the introduction, Ref. 18 reported values for γ (no curves given) for $x=0.9$ and 0.95. These values are approximately consistent with our C/T values for $T\sim 1$ K (see Fig. 9).

[c]Parameters are from the spin fluctuation fit for $x\leqslant 0.8$ and from the non-Fermi-liquid fit for $x\geqslant 0.875$, where for the nFl δ is given in mJ mole^{-1} K^{-2}.

[d]Derived from χ_{fc} - χ_{zfc}.

[e]Derived from the temperature of the peak of the broad anomaly in $C(T)$. The values in parentheses are for those compositions where only a slight anomaly in C vs T, and no anomaly in C/T, is visible.

(obeying Vegard's law) between 7.766 Å for UAl_2 and 7.860 Å for YAl_2.

Magnetic measurements were made using an automated superconducting quantum interference device magnetometer from Quantum Design, an MPMS7. Resistivity measurements were made on cut bars using a standard four-lead dc method. Specific heat was measured using a small sample time constant method, as discussed in the literature.[20]

As seen in Figs. 1 and 2 and in Table I, previous magnetic-susceptibility measurements on this system included $U_{0.9}Y_{0.1}Al_2$,[14,19] $U_{0.5}Y_{0.5}Al_2$,[19] and $U_{1-x}Y_xAl_2$, $x\geqslant 0.95$,[18] which were not sufficient to describe the strong increase in χ with x at low temperatures that begins between 10% and 30% Y doping (see Fig. 1), nor the peak in χ(1.7 K) near 70% Y doping (Fig. 2 and Table I). Also, for $0.35\leqslant x\leqslant 0.75$ (see Fig. 2) there is a leveling off of χ below 5 K that may be indicative of a transition.

In order to further investigate this anomaly, we measured the low-temperature magnetic susceptibility of the $0.20\leqslant x\leqslant 0.95$ samples cooled in field (''fc'') as well as cooled in zero field (''zfc'') down to 1.7 K, with the field (200 G) then turned on and measurements made as a function of increasing temperature. These data, shown in Figs. 3 and 4, show classic spin glass behavior, with the spin freezing temperature, T_f (defined as the temperature where the zfc and fc curves deviate from one another, see Table I)

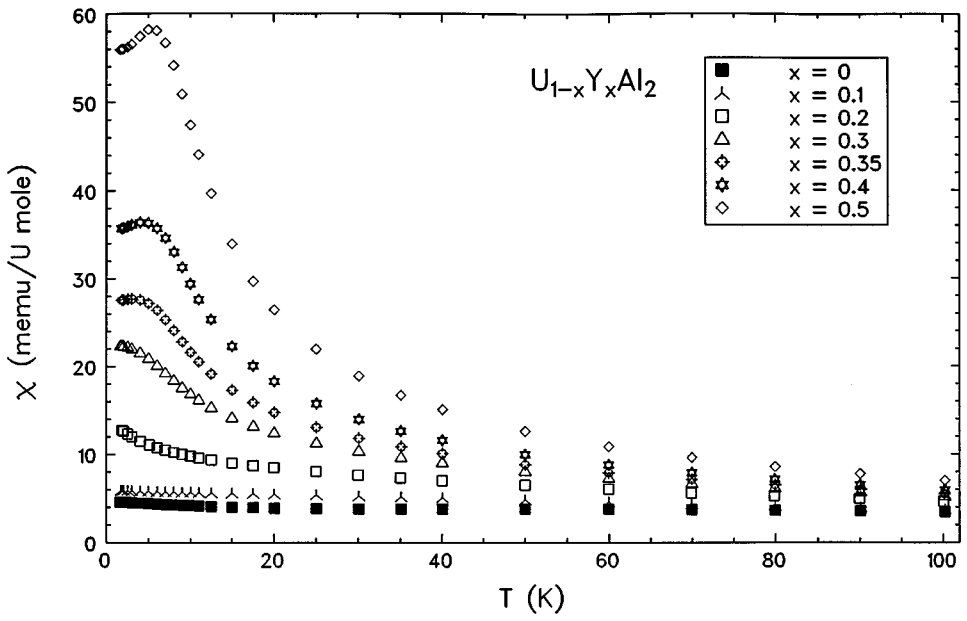

FIG. 1. Low-temperature magnetic susceptibility ($B=5000$ G) of $U_{1-x}Y_xAl_2$, $0\leqslant x\leqslant 0.5$, normalized per U mole. Note the rapid jump in $\chi(T\rightarrow 0)$ between $x=0.1$ and 0.3, and the peak around 6 K for $x=0.35$, 0.4, and 0.5.

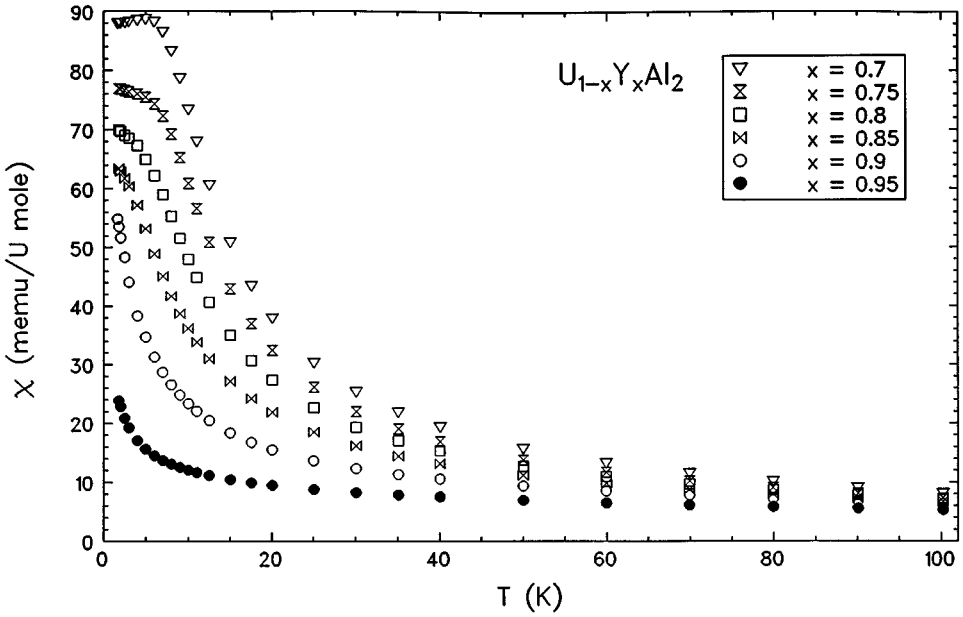

FIG. 2. Low-temperature magnetic susceptibility ($B=5000$ G) of $U_{1-x}Y_xAl_2$, $0.7\leqslant x\leqslant 0.95$, normalized per U mole. $\chi(T\rightarrow 0)$ is a maximum for $x=0.7$. Note the plateau in χ for $x=0.7$ and 0.75 below ~ 5 K.

being approximately constant (~ 5 K) for $0.35\leqslant x\leqslant 0.70$, and then falling to 2 K for $x=0.875$.

Let us now consider the specific heat. Based on results for $U_{0.85}La_{0.15}Al_2$,[10] where a larger low-temperature χ ($\sim$ factor of 2 vs pure UAl_2) was accompanied by almost a factor of 2 increase in γ ($\equiv C/T$ as $T\rightarrow 0$) and an increase in the spin-fluctuation temperature, one would expect increases in γ per U mole also in the present work. Specific-heat data for $U_{1-x}Y_xAl_2$, $0\leqslant x\leqslant 0.3$, are shown in Fig. 5. At 10% doping, we see approximately the same relative increase in γ ($\sim 30\%$) vs pure UAl_2 as seen in Fig. 1 in the magnetic susceptibility. However, the upturn in C/T below the minimum, which is due to spin fluctuations (spin fluctuations give an extra $T^3\ln T/T_{SF}$ term in C, see, e.g., Ref. 21), is partially suppressed by 10% Y doping, as may be seen visually in Fig. 5 (the difference between C/T at the minimum and at 1.3 K is a factor of 2 smaller in $U_{0.9}Y_{0.1}Al_2$ vs UAl_2) and quantitatively from the size of the $T^3\ln T$ term coefficient, δ, in Table I. In contrast, 30% Y doping in UAl_2 gives (Fig. 5) a substantial increase in the upturn in C/T below the minimum, implying stronger spin fluctuations, as well as a large ($\sim$ factor 2.7) increase in γ. We have, following the method of Ref. 22, calculated T_{SF} (see Table I) and find — as qualitatively seen in Fig. 5 — a 40% increase in $U_{0.7}Y_{0.3}Al_2$ vs pure UAl_2. It is further worth noting that the $\chi(1.7\text{K})/\gamma$ ratio (proportional to the Wilson ratio[23]) is almost a factor of 2 larger in $U_{0.7}Y_{0.3}Al_2$ than in UAl_2, indicating a yet stronger magnetic character. This is naturally consistent with the above-mentioned observation that spin-glass behavior starts for $x\geqslant 0.2$.

Considering now the specific-heat data for the region $0.35\leqslant x\leqslant 0.7$ (see Fig. 6), we see that the spin-fluctuation caused upturn in C/T continues to higher Y concentrations and, in fact (see Table I) the calculated T_{SF} continues to climb in this concentration regime. However, at the T_f spin-glass transition (see Figs. 3 and 4 and Table I), the C/T data

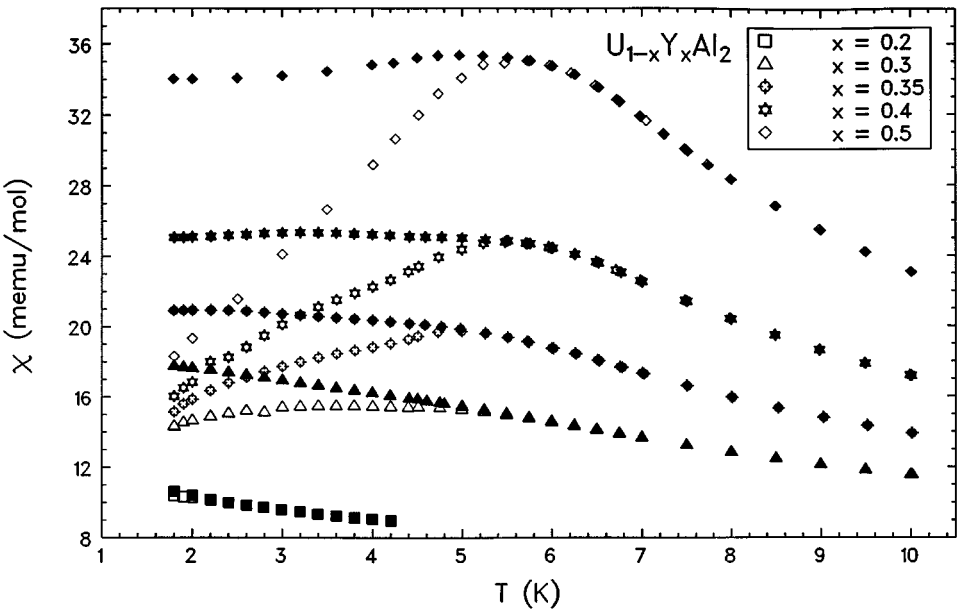

FIG. 3. Zero-field-cooled (open symbols) and field-cooled ($B=200$ G, closed symbols) dc magnetic susceptibility, normalized per formula unit $U_{1-x}Y_xAl_2$ (i.e., *not* per U mole), for $0.2\leqslant x\leqslant 0.5$. Note the increasing deviation between zfc and fc curves with increasing x.

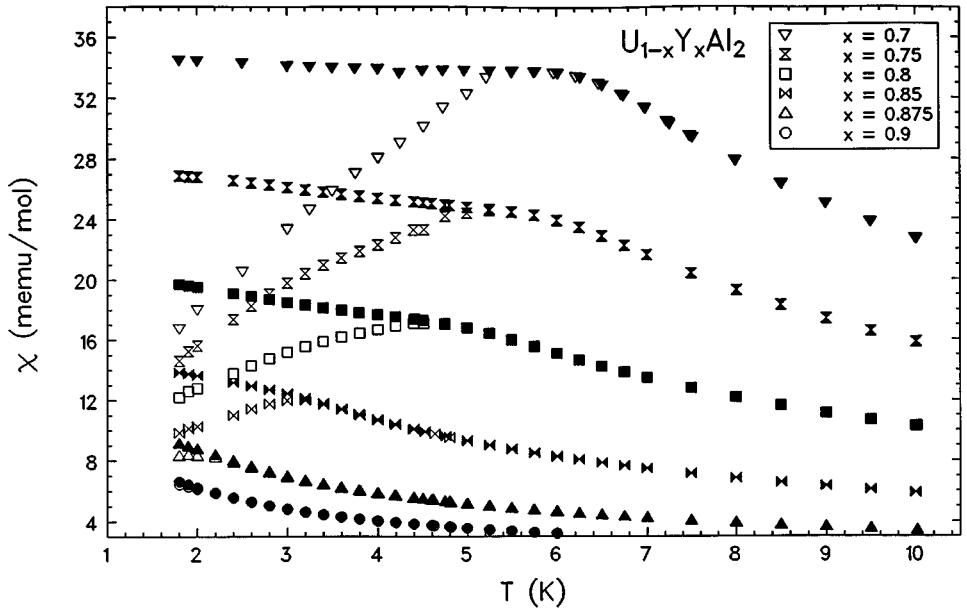

FIG. 4. Zero-field-cooled (open symbols) and field-cooled ($B=200$ G, closed symbols) dc magnetic susceptibility for $U_{1-x}Y_xAl_2$, normalized per formula unit, for $0.7\leqslant x\leqslant 0.9$. Note the strong decrease in the difference between zfc and fc curves, as well as the decrease in the temperature where the two curves diverge, with increasing x. A measurement for $x=0.95$ revealed no deviation between zfc and fc curves.

deviate below the fit to the higher temperature data. When plotted as C vs T (not shown), a broad spin-glass peak in C is easily recognizable for $0.35\leqslant x\leqslant 0.7$, with a peak temperature that scales with T_f approximately as 1.2 T_f (see Table I) — as is typical[24] of spin glasses. Thus, $U_{1-x}Y_xAl_2$, $0.35\leqslant x\leqslant 0.7$, is a system where spin fluctuations and relatively strong spin-glass behavior coexist. Presumably, the dopant atoms defeat the moment compensation mechanism locally, giving a frustrated U $5f$ spin that interacts with other defect lattice sites until, below T_f, the spins freeze due to insufficient thermal excitation in a zfc condition to align themselves in low fields. It is interesting that the spin fluctuations[25] persist after the spin glass forms — this is then a sign that these fluctuations possess a somewhat more local character, since long-wavelength paramagnons would likely be affected (scattered) by the local spins present in a spin glass.

In the concentration region $x>0.7$, where the spin-glass behavior in χ weakens (Fig. 4), the specific heat γ per U mole remains strongly enhanced and essentially constant up to $x=0.9$ (Fig. 7 and Table I), T_{SF} (Table I) falls somewhat, and χ (Fig. 2 and Table I) recedes from its maximum value at $x=0.7$. (See Fig. 8, where a plot of T_{SF} and T_f vs x gives an overview.) Further, there is no obvious spin-glass anomaly in C/T for $x>0.7$, as is in Fig. 6 for $0.35\leqslant x\leqslant 0.7$, although a weak anomaly can be seen in a plot of C vs T. Thus, the smaller deviation between the fc and zfc χ data (Figs. 3 and 4), the decrease in T_f and the weak anomaly in C (not shown), are consistent with a weakening of spin-glass behavior. It is noticeable in Fig. 7 that, for $x=0.85$, the fit for the specific heat using $T^3\ln T$ as an additional term to the normal electronic (γT) and lattice ($\beta T^3+\epsilon T^5$) terms does not describe the data well. Shown in

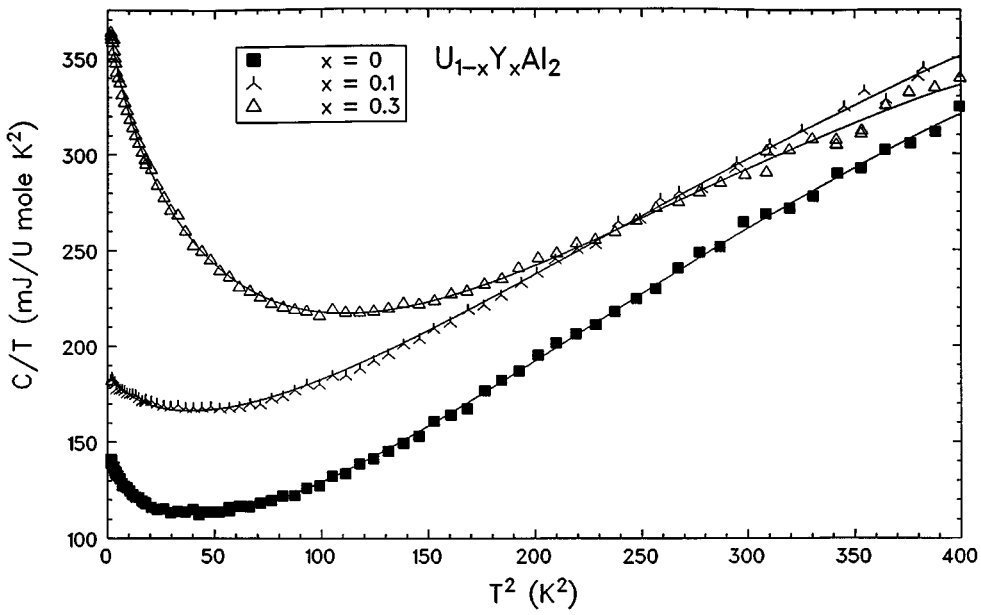

FIG. 5. Low-temperature specific heat divided by temperature, 1.3 K$\leqslant T\leqslant$20 K, normalized per U mole, versus temperature squared. The lines through the data are fits to $C/T=\gamma+\beta T^2+\epsilon T^4+\delta T^2\ln T$. The strong increase in γ ($\lim_{T\to 0}C/T$) coupled with the steep increase of C/T below 10 K for $x=0.3$ are qualitative signs of a strong increase in the influence of the spin fluctuations, which cause the $T^2\ln T$ term in C/T.

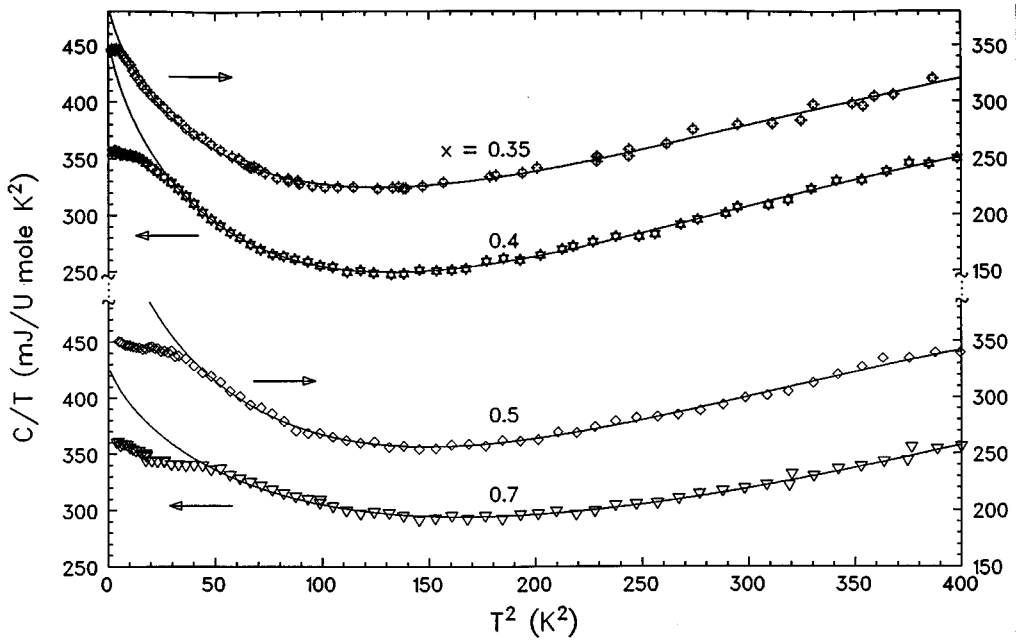

FIG. 6. Low-temperature specific heat divided by temperature versus temperature squared for $U_{1-x}Y_xAl_2$, $x=0.35$, 0.4, 0.5, and 0.7. The solid line is a fit of C/T to $\gamma+\beta T^2+\epsilon T^4+\delta T^2\ln T$ (i.e., spin-fluctuation behavior). These fits deviate from the data at low temperature at (roughly) 1 K above the temperature where the zfc and fc curves deviate from one another in Fig. 3, with the exception of $x=0.35$, where the specific heat deviates from the spin-fluctuation fit ~ 1.5 K lower than T_f determined from χ. When plotted as C vs T, the data below $T_{\text{deviation}}$ show a broad peak as is characteristic of spin glasses; this peak in C vs T (see Table I) — in contrast to $T_{\text{deviation}}$ from the C/T curves — lies consistently at about $1.2T_f$ derived from χ. The coinciding of spin-glass and spin-fluctuation specific heat in one and the same sample in so clear a fashion is unique.

Fig. 7 is a fit to $C/T=\gamma+\beta T^2+\epsilon T^4+\delta\ln T$, i.e., a non-Fermi-liquid behavior. This is carried further in Fig. 9 where, for $x\geqslant 0.875$, the specific-heat difference C/T(measured) $-(\beta T^2+\epsilon T^4)$ is shown vs $\ln T$. The logarithmic behavior over a decade and a half of temperature is convincing evidence that dilute U in YAl_2 achieves the non-Fermi-liquid (nFl) ground state.

In addition to the specific heat, nFl systems typically[5–7] show a linear behavior in the low-temperature resistivity, as well as either a power law or $\ln T$ dependence in the magnetic susceptibility — in so far as pure temperature dependences can be found.[26] We observe[27] (not shown) $\rho\propto A-BT$ be-

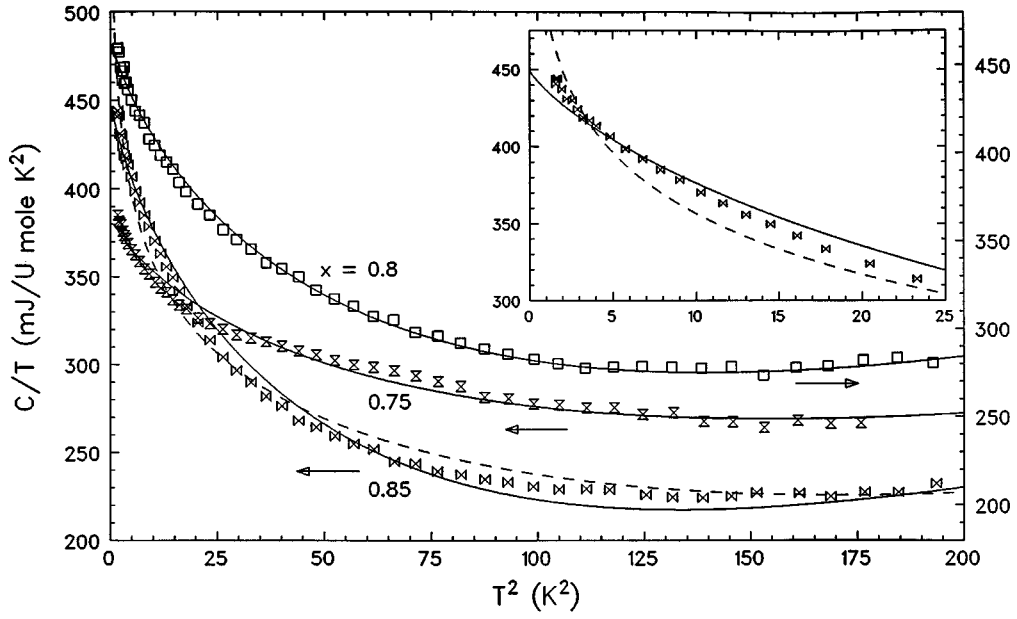

FIG. 7. Low-temperature specific heat divided by temperature vs temperature squared for $U_{1-x}Y_xAl_2$, $x=0.75$, 0.8, and 0.85 (also in the inset). Note in the inset that neither a spin-fluctuation fit (solid line) nor a nFl fit (dashed curve) fits the data. The solid lines are all spin-fluctuation fits as described in the text.

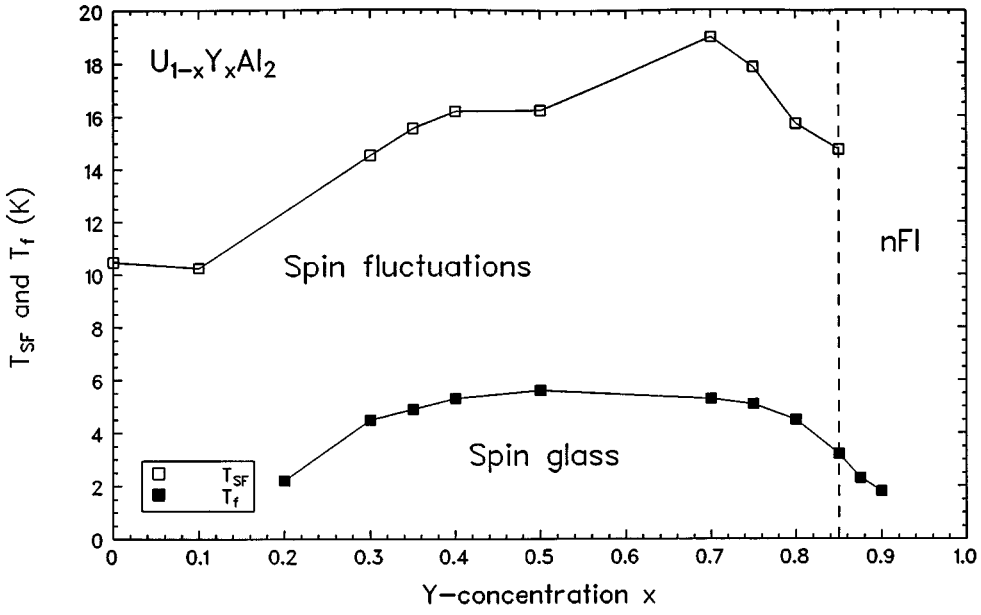

FIG. 8. Spin-fluctuation temperature, T_{SF}, and spin-glass temperature, T_f, as a function of x for $U_{1-x}Y_xAl_2$, with the boundary for non-Fermi-liquid (nFl) behavior shown by a vertical dashed line. This overview ''phase diagram'' summarizes the various trends discussed in the text.

tween 1 and 10 K in $U_{0.1}Y_{0.9}Al_2$, while $\chi \propto 1/\sqrt{T}$ between 3 and 200 K. At temperatures below these two measurements regimes, both ρ and χ rise with decreasing temperature less rapidly, i.e., there appears to be crossover behavior as observed in Ref. 26 — perhaps in the present case connected with the remanent weak spin-glass behavior.

What conclusions can be drawn from the above results? First, Y doping clearly drives the system to a weak form of magnetic behavior, i.e. to a spin glass. In a separate work[28] on the effects of Sc (isoelectronic to Y) doping on UAl_2, we observe no sign of spin-glass behavior: neither a zfc vs fc difference in χ nor a peak in C vs T as seen in Figs. 3, 4, and 6 for $U_{1-x}Y_xAl_2$. Thus, since Sc contracts the UAl_2 lattice vs the expansion caused by Y, we conclude that this spin-glass behavior in $U_{1-x}Y_xAl_2$ may come from the expansion of the UAl_2 lattice beyond the Hill limit, rather than from either electronic effects or destruction of the U-sublattice order. At, however, the same time the spin-glass $U_{1-x}Y_xAl_2$ samples are still showing definite signs of spin-fluctuations in the specific heat and, actually, a further strengthening of the spin fluctuation temperature, see Fig. 8. Also in this region the large specific heat γ/U mole is clearly enhanced, i.e., the entropy involved in the spin-glass transition does not subtract from the entropy contained in the increased γ. After the spin-glass transition begins to weaken ($x > 0.7$) the spin fluctuations remain over a certain doping range ($x = 0.75$ and 0.8), albeit with a slightly decreasing T_{SF}, and then transform into a non-Fermi-liquid state with C/T diverging as $\ln T$. Thus, since — as often observed[3] for nFl systems — magnetism must first be suppressed nearby in the phase diagram in order for nFl behavior to first appear, we infer that the direct predecessor to the nFl behavior here is therefore a spin-fluctuation ground state. In fact, for $x = 0.875$ and 0.9, spin-glass behavior is *still present* in χ, while the specific heat is showing $C \propto T \ln T$ nFl behavior. Thus, if one understands[3] nFl behavior as occurring first when magnetism

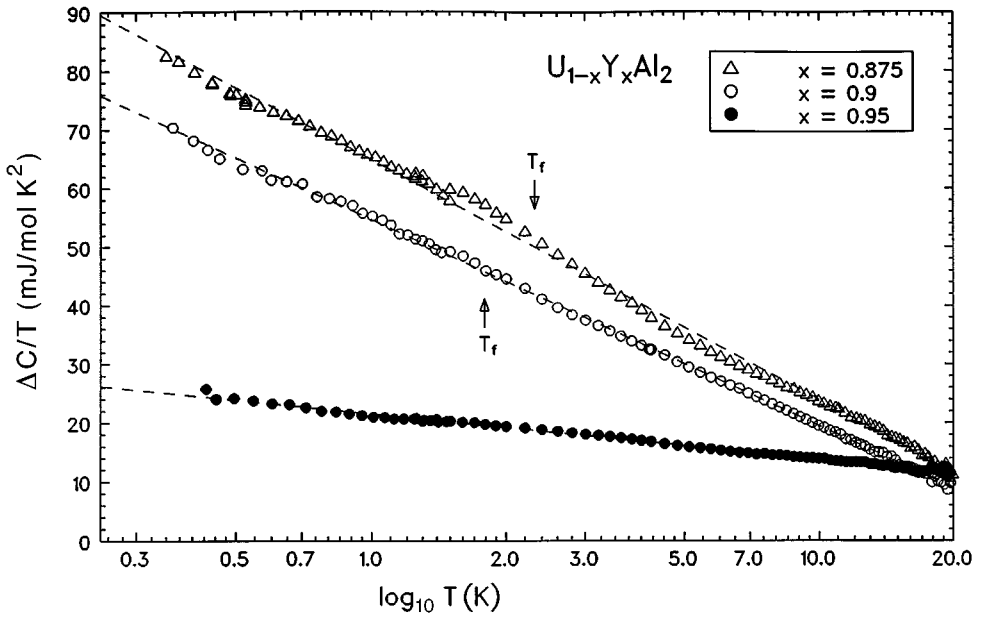

FIG. 9. $\Delta C (= C_{\text{measured}} - \beta T^3 - \epsilon T^5)$ divided by temperature vs $\ln T$ for $U_{1-x}Y_xAl_2$, $0.875 \leq x \leq 0.95$, normalized per formula unit. The linear behavior (dashed lines) of the data over a decade and a half in temperature is strong evidence for non-Fermi-liquid behavior. No substantial trace of a spin-glass anomaly is seen here in the specific-heat data, although a small deviation in zfc vs fc χ_{dc} data is still visible (Fig. 4) for $x = 0.875$.

is suppressed to $T=0$, then the present results would indicate that the suppression of the spin fluctuations is the harbinger of the nFl ground state.

The type of nFl behavior observed here, that past a certain dilution all samples show $C/T \propto \ln T$, is not unique but is in contrast to, e.g., the behavior observed in $CeCu_{6-x}Au_x$ (Ref. 5) and $CePtSi_{1-x}Ge_x$,[29] where this divergence in C/T is only observed at one certain concentration, with different (nondivergent) behavior in either direction of x. Thus, rather than a critical-point model perhaps the present results are more representative of a true equilibrium ground state, where we take this to mean a state insensitive to small excursions in parameter space, be it doping or magnetic field or pressure or some other parameter.

ACKNOWLEDGMENTS

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