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Strain-dependent magnetic anomalies in doped UPt_3

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An anomaly at 6 K in the magnetic susceptibility and the specific heat is found in UPt_3 doped both by Pd on the Pt sites and by Th on the U sites. The compositional behavior of the anomaly, with T_{anomaly} essentially constant for certain compositions ($0.04 \leq x \leq 0.08$ for Pd and $0.1 \leq x \leq 0.2$ for Th) and falling rapidly at higher or lower concentrations, is unusual. With the use of field measurement of the specific-heat and the susceptibility data, the anomaly appears to be due to antiferromagnetism. This long-range order is found to be quite strain sensitive, disappearing upon grinding, just as found for superconductivity in pure UPt_3. A simple model that encompasses all the data is proposed to explain the behavior of the anomaly.

I. INTRODUCTION

Since the discovery[1] of its strongly enhanced magnetic susceptibility, the compound UPt_3 has become of greater and greater interest. The finding[2] of bulk superconductivity in UPt_3 coupled with a $T^3\ln T$ term in the low-temperature specific heat, a sign of spin fluctuations, led to speculation[2,3] that UPt_3 may be the first example of a non—Bardeen-Cooper-Schrieffer superconductor. Ultrasonic attenuation data[4] and extensive theoretical work[5] have put this speculation on firmer footing. Additionally, the normal-state properties of UPt_3 show various interesting phenomena,[6] including some sort of field-induced transition at $20T$.

Since doping[7–9] has been proved to be a useful probe in understanding UBe_{13}, we have carried out studies of UPt_3 substituted with Th on the U sites and, separately, Pd on the Pt sites. Some of our preliminary work on $U(Pt_{0.8}Pd_{0.2})_3$ has already been reported.[10] Also, a specific-heat study between 1.2 and 30 K of $U(Pt_{1-x}Pd_x)_3$ for $x=0.01$, 0.02, 0.05, 0.10, 0.15, 0.20, and 0.30 by de Visser *et al.*[11] revealed the existence of an anomalous peak in the specific heat around 6 K for 5 at. %-Pd-substituted UPt_3 that had essentially vanished by the 2 and 10 at. % concentrations. We have therefore focused our Pd work in this area, with $x=0.04$, 0.05, 0.06, 0.08, and 0.10, and report both specific heat (0.6 to 10 K) and magnetic-susceptibility results which shed further light on this anomaly. These data, coupled with our results on $U_{1-x}Th_xPt_3$, for $x=0.1$, 0.15, 0.2, 0.25, 0.30, and 0.41, show that substitution on either site in UPt_3 produces qualitatively quite similar magnetic anomalies.

II. EXPERIMENTAL DETAILS

Small arc-melted buttons were prepared using pure starting materials and remelted four times. In order to make precisely the rather small changes in Pd concentration required, a larger button of $U(Pt_{0.95}Pd_{0.05})_3$ was prepared, and small amounts of Pd were added to make the compositions $x=0.06$, 0.08, and 0.10. Pure UPt_3 was used to dilute a piece of $U(Pt_{0.95}Pd_{0.05})_3$ in order to make $x=0.04$. (The question of possible second phases will be discussed below with the results.) As will be seen in the data below, rapid changes in properties accompany these quite small changes in Pd concentration, justifying the effort in obtaining a precise, internally consistent accuracy for x. The absolute accuracy of x, as distinct from the internal precision, is $\pm 0.5\%$. The Th-doped samples were prepared similarly, with the starting composition $x=0.1$ and further samples made by adding $ThPt_3$ except for a second sample of $x=0.2$ and the $x=0.15$ sample, made from new starting materials for reasons discussed below.

III. RESULTS

A. Pd

The low-temperature specific heat of UPt_3 and $U(Pt_{0.94}Pd_{0.06})_3$ are shown in Fig. 1, with arrows indicating the position of the peaks for $x=0.04$, 0.05, 0.08, and 0.10. In the work of de Visser *et al.*,[11] a small peak in C/T at $T=3.6$ K (with $C_{\max}/T=490$ mJ/mole K^2) was observed for $x=0.02$ and their data for $x=0.05$ and 0.10 match very well with our results shown in Fig. 1. The fact that the peak in C/T for $x=0.06$ is slightly higher in temperature than that observed for $x=0.05$ may not be significant, i.e., may be due to thermal history (e.g., rate of cooling on the copper hearth after melting). In any case, $T_{\max}$ is almost constant for $0.04 \leq x \leq 0.08$, while it falls in temperature by 40% (Ref. 11) for $x=0.02$ and 45% for $x=0.10$. Also, the value of $C_{\max}/T - C^{UPt_3}/T$ behaves similarly, with only a slight shoulder for $x=0.10$, as shown in Fig. 2.

What is the origin of this anomaly, which is absent[10] at higher Pd content? One method for distinguishing magnetic from nonmagnetic transitions is to measure the specific heat as a function of field. In $5.5T$, the values of

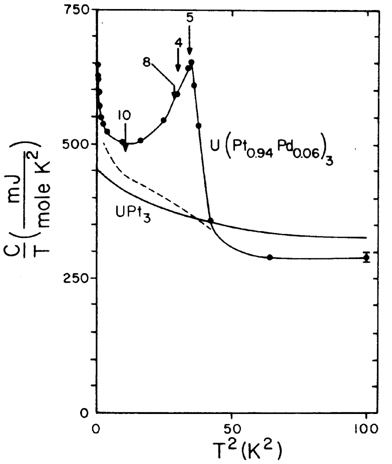

FIG. 1. Low-temperature specific heat C, divided by temperature T, versus T^2 for $U(Pt_{0.94}Pd_{0.06})_3$ (upper curve with points) and pure UPt_3 (lower curve). The arrows indicate the height and temperature of peaks in C/T for the respective values of x in $U(Pt_{1-x}Pd_x)_3$. The dashed line is explained in the text.

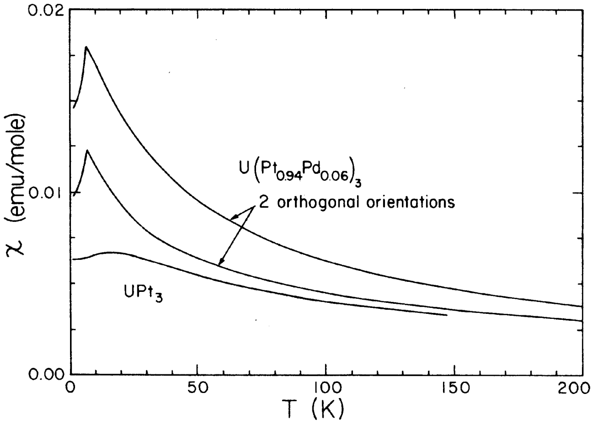

FIG. 3. Susceptibility χ versus temperature T for pure UPt_3 (lower curve) and two orthogonal orientations for the same bulk piece of $U(Pt_{0.94}Pd_{0.06})_3$. A plot of χ^{-1} versus T for the upper set of data for bulk $U(Pt_{0.94}Pd_{0.06})_3$ is parallel to such a plot of the pure UPt_3 data, unlike the data for a powdered sample shown in Fig. 4.

C_{max}/T for $x=0.05$ (these field data are not shown in Fig. 2 for the sake of clarity) falls 6% and T_{max} decreases from 5.83 to 5.26 K. These results are consistent with an antiferromagnetic transition. The small ($\sim 16\%\ R\ln 2$) entropy associated with the anomaly indicates that the antiferromagnetism is of itinerant nature.

The susceptibility, measured in a Faraday-balance magnetometer, of $x=0.06$ and pure UPt_3 is shown in Fig. 3. Ignoring the preferred orientation of our arc-melted $UPt_{0.94}Pd_{0.06}$ button for the moment, the χ data clearly show a peak at $T \sim 6$ K, in agreement with the specific-heat data. Furthermore, the shape and size of the peak in χ are indicative of antiferromagnetism, again in support of the field data for C. A χ^{-1} versus T plot (not shown) of the data in Fig. 3 shows approximately the same effective moment for pure UPt_3 and $U(Pt_{0.94}Pd_{0.06})_3$, with the (negative) intercepts being 43 K for the doped sample versus ~ 100 K for pure UPt_3.

Since a Laue x-ray diffraction pattern of the as-prepared arc-cast piece of $U(Pt_{0.94}Pd_{0.06})_3$ did not give a clear idea of the orientation (although it was clearly preferentially aligned), the appropriate average of the χ results for the two orthogonal orientations shown in Fig. 3 to obtain χ_{av} for comparison to pure UPt_3 is uncertain. If one assumes that, like UPt_3, $\chi(C_{\parallel}) < \chi(C_{\perp})$, and that the arc-melted button grew with the c axis approximately along the button axis, then $\frac{2}{3}\chi^{upper} + \frac{1}{3}\chi^{lower}$ (from Fig. 3) is 13.0×10^{-3} emu/mole at 1.6 K compared to 6.7×10^{-3}

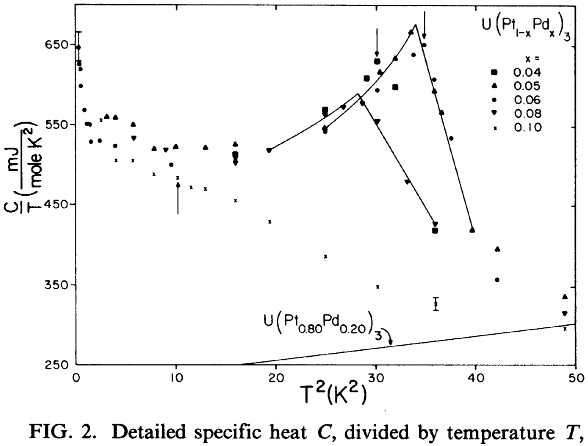

FIG. 2. Detailed specific heat C, divided by temperature T, versus T^2 plots of the data for $U(Pt_{1-x}Pd_x)_3$. Lines through the $x=0.05$ and 0.08 data are guides to the eye. Arrows mark the peaks in C/T for the other compositions. Our previous data for $x=0.20$ are shown as the straight line in the lower part of the figure. A field of 5.5 T shifts the $x=0.05$ data left and down slightly, coincident with the $x=0.08$ peak in temperature but between the $x=0.05$ and 0.08 data in amplitude.

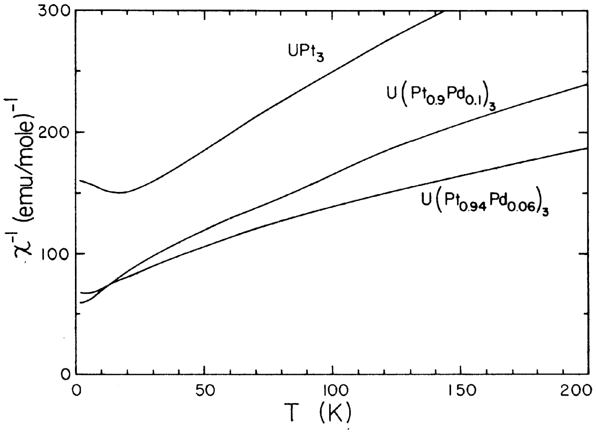

FIG. 4. Inverse susceptibility χ^{-1} versus temperature T for *powdered* specimens of $U(Pt_{0.94}Pd_{0.06})_3$ and $U(Pt_{0.90}Pd_{0.10})_3$. Note the absence of any strong minimum in the $x=0.06$ data, i.e., the peak seen in Fig. 3 is absent after grinding. Also, the increase in χ by adding Pd is approximately saturated by $x=0.10$.

emu/mole for pure UPt_3. (Such alignment assumptions are not central to the comparison, since $\frac{1}{3}\chi^{\text{upper}}+\frac{2}{3}\chi^{\text{lower}}$ gives 11.5×10^{-3} emu/mole for χ_{av}.) Therefore, the low-temperature magnetic susceptibility shows a strong (factor of 2) enhancement upon substitution of 6 at.% Pd in UPt_3, as well as an antiferromagnetic-like anomaly at 6 K.

In order to insure that this anomaly was not due to a spurious second phase, metallography was performed on an $x=0.05$ specimen. (At these small concentrations, and with the similarity in size of Pt and Pd leading to almost no change versus composition of a_0 and c_0 (5.747 and 4.895 Å, respectively), x-ray diffraction was not a good measure of second-phase content.) Based on the metallography work, which involved mechanical polishing down to 0.25-μm size grit and electro-etching in a HCl, NaCl, and water solution, no second phase (with a detectability limit of $\sim1-2\%$) was present. Without the Th work reported below, this low level of the possible second phase would be crucial to excluding the presence of a UPd_3 second phase as the cause of the anomaly at 6 K. (1% of UPd_3 would[12] give at most $\sim2-3\%$ of the anomaly actually observed in the specific heat shown in Figs. 1 and 2.) Since metallography was only performed on the $x=0.05$ $U(Pt_{1-x}Pd_x)_3$ sample, second phase could be present in the other specimens up to the lower detectability limit of the x rays (5%). However, except for UPd_3, none of the possible second phases (e.g., UPt_2, UPt_5) show[13] anomalies in their low-temperature specific heats, and the sample with the largest Pd content, $x=0.1$, in the present work ($x=0.2$ in previous work,[10] see Fig. 2) shows no anomaly at 6 K reminiscent of UPd_3 in any case.

In order to more accurately determine χ_{av} for the doped samples, we ground a specimen of $U(Pt_{0.94}Pd_{0.06})_3$ and pressed a pellet from the powder. As shown in Fig. 4, the peak structure in χ at 6 K (or minimum in χ^{-1}) is destroyed by grinding, and $\chi(1.6\ \text{K})=14.7\times10^{-3}$ emu/mole. Additionally, χ^{-1} for $x=0.06$ is no longer parallel to χ^{-1} for pure UPt_3, as it was before grinding.

These results raised the question: How does grinding effect the anomaly observed in the specific heat at 6 K, Figs. 1 and 2? A part of the $U(Pt_{0.94}Pd_{0.06})_3$ pellet used for the χ measurements was therefore measured in the calorimeter. The pellet was prepared using a 0.3-cm-diam pellet press operated at 1 400 000 kPa (200 000 psi), which was developed[14] for use with very hard materials. Excellent thermal contact between the approximately 200—300 mesh size particles was achieved at this rather high pressure such that no sign of inadequate internal thermal relaxation (a "τ_2") in the calorimeter was observed down to the lowest temperature of measurement, 1.6 K. Above about 7 K, the C/T data for the powder and the polycrystalline chunk are identical. However, below 7 K the specific heat of the powdered sample begins to fall significantly below that of the bulk specimen. These data are shown in Fig. 1 by the dashed line. Thus, the specific-heat anomaly at 6 K (believed, based on discussion above, to be due to antiferromagnetism) in $U(Pt_{0.94}Pd_{0.06})_3$ (and presumably also for the other compositions) is *totally suppressed* by grinding, just as we found[2] for superconductivity at 0.5 K in pure UPt_3. (Also, remelting the pressed pellet brings the specific heat for $x=0.06$ back to within a few percent of the original bulk result shown by the upper, solid curve in Fig. 1.)

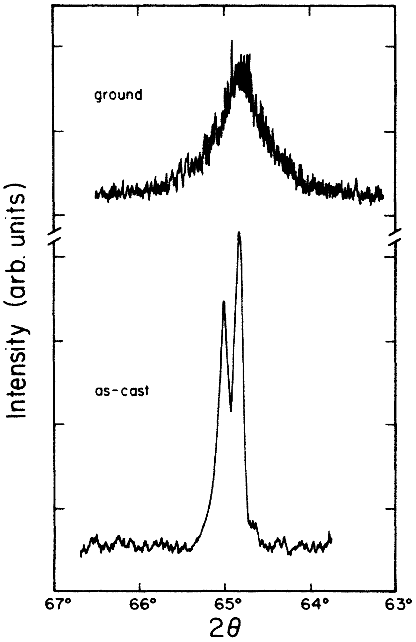

FIG. 5. Comparison of the (110) x-ray diffraction intensity at 64.8° 2θ for as-cast (lower curve) $U(Pt_{0.94}Pd_{0.06})_3$ and ground (upper curve) $U(Pt_{0.94}Pd_{0.06})_3$. Note the nicely resolved α_2 line at 65° 2θ in the as-cast specimen. The choice of the (110) line for comparison was dictated by the preferred orientation of the as-cast specimen.

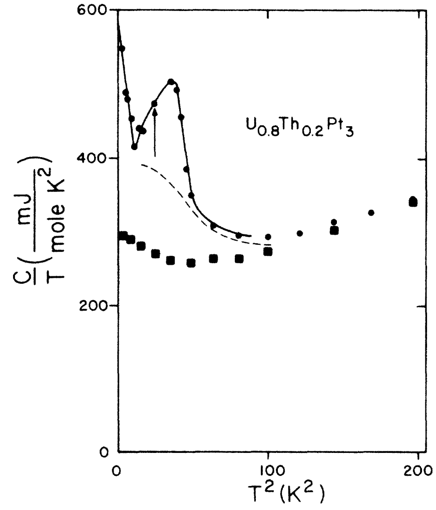

FIG. 6. Specific heat C divided by temperature T versus T^2 for two samples of $U_{0.8}Th_{0.2}Pt_3$ (dots and squares). The arrow marks the peak in $5.5T$ for the upper-curve sample. An $11T$ field essentially totally suppresses the peak, leaving a slight shoulder. The specific heat for $U_{0.9}Th_{0.1}Pt_3$ is essentially the same as for the upper curve. A pellet pressed from powder of $U_{0.9}Th_{0.1}Pt_3$, has the specific heat shown by the dashed line, i.e., the 6-K anomaly is destroyed by grinding. The lower squares are data for a second preparation of $U_{0.8}Th_{0.2}Pt_3$.

Therefore, the anomaly at 6 K is seen to be quite sensitive to strain in the lattice induced by grinding. A semiquantitative measure of the effect on the lattice of this grinding is the measured linewidths of the higher angle x-ray diffraction lines. Figure 5 compares the as-arc-melted versus after-grinding (110) linewidths for $U(Pt_{0.94}Pd_{0.06})_3$. The effect of grinding the UPt_3 lattice is seen to be quite severe, causing the α_1-α_2 doublet in the as-cast $x=0.06$ material to be completely smeared out.

B. Th

Our work on $U_{1-x}Th_xPt_3$, in fact, predated our Pd substitution work, but, as discussed below, its interpretation was unclear until the strain sensitivity of the anomaly in $U(Pt_{1-x}Pd_x)_3$ was discovered.

The specific heat of two samples of $U_{0.8}Th_{0.2}Pt_3$ is shown in Fig. 6. The results for $U_{0.9}Th_{0.1}Pt_3$ are essentially identical with those of the upper curve, while only slight shoulders in C/T starting at $T\sim 8$ K (7 K) are observed for $x=0.25$ (0.30), whereas for $x=0.41$ a drop-off below 5 K is seen with no shoulder. The $\gamma(\equiv C/T$ as $T\rightarrow 0)$ values for these three higher Th compositions are 160, 108, and 42 in mJ/mole K^2, respectively. From the results for $x=0.1$, 0.2 (upper curve), 0.25, 0.30, and 0.41, and remembering that Th significantly expands the lattice ($a_0=5.838$ Å, $c_0=4.849$ Å for $x=0.41$), $U_{1-x}Th_xPt_3$ seems quite similar to the Pd results: (1) the width of the transition is due to strains from the (Th-caused) lattice expansion; (2) the temperature and height of the anomaly in C/T decreases with increasing Th content; (3) γ at higher Th concentrations falls, just as seen[10] in $U(Pt_{0.8}Pd_{0.2})_3$. What was initially unclear, previous to our Pd strain results, was why a sample of $U_{0.85}Th_{0.15}Pt_3$ had a specific heat with *no* anomaly peaked at 6 K, just a smooth rise below 8 K in C/T with $\gamma=400$ mJ/mole K^2. Since, as discussed in Sec. II above, this sample was made from a new set of starting materials due to depletion of the $x=0.1$ ingot, we initially suspected impurities as the cause of the peak in C/T at 6 K, Fig. 6. A sample of $U_{0.8}Th_{0.2}Pt_3$ was prepared from the $x=0.15$ ingot and gave the results shown in Fig. 6, lower set of data, i.e., no anomaly was present. This also seemed consistent with the impurity hypothesis. Metallography, however, indicated that the first $x=0.20$ sample had at most a 2% impurity phase, while the second sample had $\sim 0.5\%$ impurity. Since the entropy difference between the two sets of data shown in Fig. 6 for $U_{0.8}Th_{0.2}Pt_3$ is $\sim 25\%$ $R\ln 2$, the impurity picture was essentially ruled out, since such a small amount of impurities would have to have $\sim 12R\ln 2$ entropy per mole impurity, an unphysical amount.

C. Comparison of Th and Pd results

What later became apparent was that, based on the sensitivity of this anomaly in Pd-doped UPt_3 to strain induced by grinding, a working hypothesis to explain the sample dependence of the anomaly in Th-doped UPt_3 was a strain sensitivity made more severe by the Th-caused lattice expansion. A measurement of the specific heat of a pellet pressed from powder from the $U_{0.9}Th_{0.1}Pt_3$ sample,[15] whose specific heat showed an anomaly essentially the same as shown for $x=0.20$ in Fig. 6, has been made to check this hypothesis. Indeed, the anomaly in the ground sample is again essentially absent, as indicated by the dashed line for the data in Fig. 6.

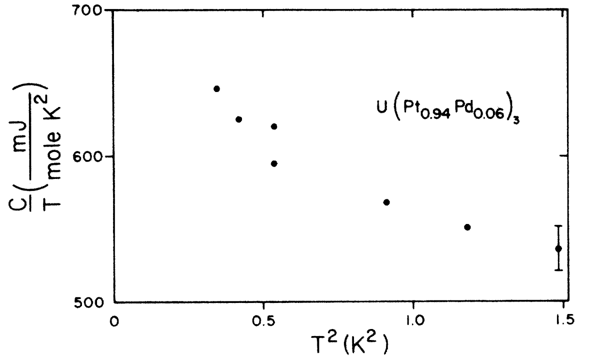

FIG. 7. Low-temperature specific heat C, divided by temperature T, versus T^2 for $U(Pt_{0.94}Pd_{0.06})_3$ between 0.6 and 1.2 K.

In addition to the similarity of T_{max}, size of the anomaly, strain sensitivity behavior, and (rough) compositional dependence, the susceptibility and field dependence of C of $U_{0.8}Th_{0.2}Pt_3$ are also comparable to that measured for $U(Pt_{1-x}Pd_x)_3$. In $5.5T$, the peak in C/T is shifted to lower temperature and decreased in size as indicated by the arrow in Fig. 6. By $11T$, the peak in C/T for $U_{0.8}Th_{0.2}Pt_3$ has vanished and been replaced by a slight shoulder at 3.6 K almost coincident with the zero-field data. The susceptibility, measured by a vibrating sample magnetometer, shows a rounded peak at 6.3 K (coincident with the specific-heat anomaly) which falls by 6.5% by 3 K, compared to $\sim 10\%$ in the same temperature range for $U(Pt_{0.94}Pd_{0.06})_3$.

A further point of comparison is the upturn in C/T below 3 K in both the Th-doped and Pd-doped systems, see Figs. 1 and 6. [This upturn is absent for $x\geq 0.20$ (0.25) for Pd (Th) doping.] From Figs. 1 and 6, strain appears not to strongly alter this lower upturn in C/T. As shown clearly in Fig. 1, this rise in C/T is much more rapid than that observed in pure UPt_3. In order to further investigate this anomaly, we measured the specific heat of the bulk sample of $U(Pt_{0.94}Pd_{0.06})_3$ down to 0.6 K, with the results shown in Fig. 7. The temperature dependence of this upturn in C/T, with the peak subtracted away,[16] is $\sim 1/T^{0.75}$, i.e., C is almost temperature independent. Thus, while this second, lower-temperature upturn in the doped samples could be the onset of a second magnetic transition (with entropy $\sim 6\%$ $R\ln 2$) as in Cr (where strain, however, increases[17] T_{order}), it does not appear to be a Schottky anomaly or to obey the spin fluctuation behavior, $C\sim T^3\ln T$. Also, the $5.5T$ and $11T$ results for C/T of $U_{0.8}Th_{0.2}Pt_3$ show a suppression of the upper anomaly but little change in the lower upturn in C/T, arguing against magnetism also as the cause of the anomaly. With superconductivity ruled out, by both our measurements and by those[11] of de Visser *et al.* for their Pd-doped samples, perhaps this second anomaly is structural.

IV. CONCLUSIONS

We have presented a great deal of data on both Th- and Pd-doped UPt$_3$. Both samples show a strain-sensitive anomaly at 6 K in the specific heat and magnetic susceptibility for intermediate compositions, which, based on the measurement of C in field and on the shape of the peak in χ, is consistent with antiferromagnetism. Aeppli *et al.*[18] report that they have observed antiferromagnetic spin fluctuations at low temperatures in pure UPt$_3$. The question is why Pd (Th) substitutions on the Pt (U) sites create an almost composition-independent anomaly, both in size and temperature, for $0.04 \leq x \leq 0.08$ ($0.10 \lesssim x \lesssim 0.20$), which is extremely composition dependent outside this narrow range?

Due to the large (4.1 Å) U-U separation in UPt$_3$, hybridization of the U $5f$ electrons with the intervening Pt electrons is clearly important. The (almost) equivalent efficacy of Th substitution on the U site and Pd substitution on the Pt site in creating the anomaly at 6 K is new evidence of just how important hybridization with the Pt site electrons is to the electronic properties in UPt$_3$. One possible explanation of the composition dependence observed for the anomaly is as follows. If one assumes[18] that UPt$_3$ has long-range (extending between at least next-nearest neighbors) antiferromagnetic spin fluctuations, then the effect of breaking U-Pt hybridization (through replacement of either member) is just tending to localize these fluctuations. The tendency toward localization (narrowing of the f band) would explain the observed increase in γ and the twofold enhancement of χ observed in U(Pt$_{0.94}$Pd$_{0.06}$)$_3$. At higher concentrations of Pd (or Th), the U-U order, coupled through the Pt sites and intervening U sites, necessary for the spin-ordered state is destroyed by too many site substitutions. This model is obviously a simple one, and would predict behavior similar to that observed here, for Th and Pd substitutions, for other substitutions, e.g., $[\mathrm{U(Pt}_{1-x}[\mathrm{Ir,Au,Rh,Ag}]_x)_3]$. As discussed in Ref. 19, subtleties of f ligand hybridization (i.e., U f—Pt site s,p,d electron hybridization) may well add additional affects in these other substitutions. However, beginning from a breaking of U-Pt hybridization causing increasing spin-fluctuation localizations, as proposed from the data presented here, may well remain a valid starting point. Because of the similar effect of Th doping to that observed for Pd, the present work certainly calls into question any explanation of the U(Pt$_{1-x}$Pd$_x$)$_3$ results as simply moving, as Pd replaces Pt, in the direction of pure UPd$_3$ with its quadrupolar transition at 6 K. Also, as pointed out in Ref. 19, structure is very important for f-ligand hybridization and the resulting range (itinerant—heavy fermion—local moment) of f-electron behavior, and pure UPd$_3$ has a different structure than the U(Pt$_{1-x}$Pd$_x$)$_3$ studied here.

Neutron diffraction studies of these alloys are planned to check the validity of this explanation and will also perhaps shed some light on the lower-temperature anomaly observed in the specific heat. Preliminary resistivity measurements on transforming samples of U$_{0.8}$Th$_{0.2}$Pt$_3$ and U$_{0.9}$Th$_{0.1}$Pt$_3$ show noticeable changes of slope (an increase in the positive slope characteristic in UPt$_3$ upon cooling of almost a factor of 3) in R versus T starting at ~6 K, but no evidence down to 1.4 K of any further anomaly.

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