

## Studies of the non-Fermi-liquid behavior in $U(\text{Pt}_{0.94}\text{Pd}_{0.06})_3$

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Coexistent magnetism and non-Fermi-liquid (nFl) behavior occur in only a few of the more than 100 known nFl systems. We have investigated the resistivity, dc magnetic susceptibility, and specific heat as a function of field (0–13 T) down to 0.4 K (0.09 K for resistivity) in high-purity polycrystalline  $U(\text{Pt}_{0.94}\text{Pd}_{0.06})_3$ , where antiferromagnetism at 6 K is known to precede non-Fermi-liquid behavior in the specific heat at lower temperatures in zero field. Unusually for a system that shows nFl behavior in the bulk specific heat, both the dc magnetic susceptibility  $\chi$  and the electrical resistivity  $\rho$  of  $U(\text{Pt}_{0.94}\text{Pd}_{0.06})_3$  show Fermi-liquid behavior down to our lowest temperature of measurement. Possible inferences about the  $q$  dependence of the non-Fermi-liquid fluctuation spectrum are discussed. As expected, magnetic field succeeds in driving the specific heat of the system back into a Fermi-liquid ground state with  $C/T$  saturating to a constant value below about 0.9 K.

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### I. INTRODUCTION

Experimental evidence for non-Fermi-liquid (nFl) behavior was first discovered<sup>1</sup> in  $U_{0.2}\text{Y}_{0.8}\text{Pd}_3$  in 1991. Since then, more than 100 additional nFl systems have been discovered,<sup>2,3</sup> ranging from systems where the “tuning” of such behavior is achieved through doping, pressure, or magnetic field to almost 20 systems where nFl behavior occurs in the as-prepared, stoichiometric compound. In general, nFl behavior is found in the part of the phase diagram where a sample is just at the border to magnetism, for example, when magnetic field suppresses a second-order antiferromagnetic phase transition to  $T=0$ .

Doping the heavy fermion superconductor  $UPt_3$  with Pd was found to induce antiferromagnetism;<sup>4,5</sup> for  $U(\text{Pt}_{0.95}\text{Pd}_{0.05})_3$   $T_N$  is around 6 K with an ordered moment of  $\sim 0.6 \mu_B$ .<sup>5</sup> After the discovery of nFl behavior in  $U_{0.2}\text{Y}_{0.8}\text{Pd}_3$ ,  $U(\text{Pt}_{0.94}\text{Pd}_{0.06})_3$  (with  $T_N$  also  $\sim 6$  K) was discovered<sup>6</sup> in 1992 to show nFl behavior ( $C/T \sim \log T$ ) via specific-heat measurements in *zero field*, i.e., coexistent with the antiferromagnetism, down to the lowest temperature of measurement, 0.4 K. Although unremarked upon in this context,  $\text{CeCu}_{5.7}\text{Au}_{0.3}$ ,  $T_N \sim 0.5$  K and with an ordered moment of  $0.4 \mu_B$ ,<sup>7</sup> was characterized in 1995 and also shows  $C/T \sim \log T$  down to 0.1 K (Ref. 7) in zero field, coexistent with the magnetic order albeit over a narrower temperature range. Five more such coexistent nFl/magnetic systems were discovered<sup>2,3</sup> starting in 2001. As the number of known nFl systems gradually increased after the discovery of  $U_{0.2}\text{Y}_{0.8}\text{Pd}_3$ ,  $U(\text{Pt}_{0.94}\text{Pd}_{0.06})_3$  appeared more and more unique for a reason besides the coexistence with magnetism. Namely, although the specific heat displayed<sup>6</sup> nFl behavior below 4.5 K, the magnetic susceptibility down to 2 K—measured by us (previously unpublished) or see data from van Spang<sup>8</sup> in the similar composition  $U(\text{Pt}_{0.95}\text{Pd}_{0.05})_3$ —showed *Fermi liquid*, or  $\chi \rightarrow \text{const}$ , behavior. Experimentally, as the overviews<sup>2,3</sup> of the measured properties of nFl systems show, finding nFl behavior in the

specific heat almost always (an exception is<sup>2</sup> MnSi) implies that  $\chi$  will also show such behavior.

Theoretically, however, this need not be the case.<sup>9</sup> If the magnetic fluctuations responsible for the nFl behavior are not present at  $q=0$  but are at finite or even large  $q$ , then the uniform magnetic susceptibility measured in a susceptibility (which samples solely  $q=0$ ) will indeed see Fermi-liquid behavior while the resistivity  $\rho$  (which is strongly influenced by fluctuations with certain  $q$  vectors based on the Fermi surface) and the specific heat  $C$  (which samples all  $q$ ) can display nFl behavior. Thus, the current work’s measurement of the uniform magnetic susceptibility  $\chi$  of  $U(\text{Pt}_{0.94}\text{Pd}_{0.06})_3$  to lower temperatures will verify if indeed the nFl behavior in  $U(\text{Pt}_{0.94}\text{Pd}_{0.06})_3$  can be assigned solely to finite  $q$  fluctuations.

A second goal of this work is to measure the low-temperature resistivity of  $U(\text{Pt}_{0.94}\text{Pd}_{0.06})_3$  down to 0.09 K. The temperature dependence of  $\rho$  for  $U(\text{Pt}_{1-x}\text{Pd}_x)_3$  has not previously been reported below 1.4 K. A third goal is the further understanding of  $U(\text{Pt}_{0.94}\text{Pd}_{0.06})_3$  via the application of magnetic field, which has often<sup>2,3</sup> been used as an investigative tool in non-Fermi-liquid systems to suppress the magnetic fluctuations responsible for the nFl behavior and re-enter the Fermi-liquid ground state.

Thus, the present work uses measurements of low-temperature  $\rho$ ,  $\chi$ , and  $C/T$  down to 0.4 K (0.09 K for  $\rho$ ) in  $U(\text{Pt}_{0.94}\text{Pd}_{0.06})_3$  in fields between 0 and 13 T to investigate the dichotomy in the  $\chi$  vs  $C/T$  results to lower temperatures, to measure  $\rho$  for the first time to lower temperatures, to determine if magnetic field causes the specific heat to recover Fermi-liquid behavior in this coexistent magnetic and nFl system, and to further understand this unusual coexistence of long-range magnetism and nFl behavior.

### II. EXPERIMENT

The original<sup>6</sup> specific-heat result, that  $C/T \sim \log T$  below  $T_{\text{Neel}} \sim 6$  K, in  $U(\text{Pt}_{0.94}\text{Pd}_{0.06})_3$  was performed on polycrys-

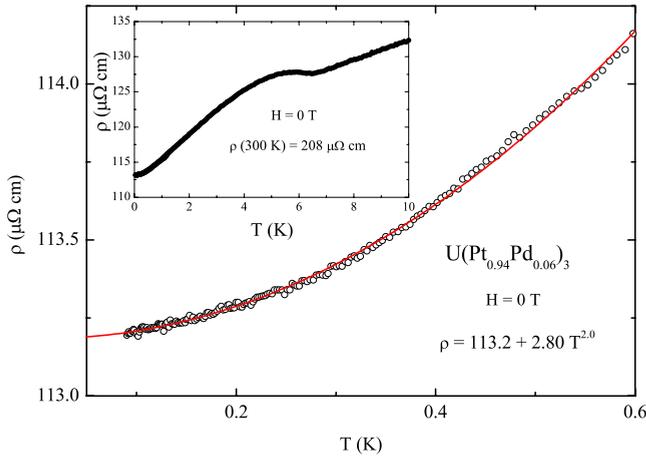


FIG. 1. (Color online) Electrical resistivity  $\rho$  vs temperature  $T$  for  $\text{U}(\text{Pt}_{0.94}\text{Pd}_{0.06})_3$ . Note the anomaly at  $T_N$  around 6 K in the inset showing the higher temperature data. The zero-field low-temperature data, together with a fit of the temperature dependence, are shown here on an expanded scale. The resultant temperature dependence is consistent with Fermi-liquid behavior. Field data  $H = 13$  T (not shown) showed considerably more noise for  $T < 0.25$  K, but again gave a temperature dependence consistent with  $\rho \sim T^2$ .

talline material. It is known<sup>10</sup> that  $\text{UPt}_3$ , which occurs in the DO19 hexagonal structure, has approximately a factor of two anisotropy in the magnetic susceptibility [ $\chi(H_{ab \text{ plane}}) > \chi(H_{c \text{ axis}})$ ] as well as a field induced metamagnetic transition<sup>4,11</sup> for  $H$  in the  $ab$  plane. However, polycrystalline  $\text{UPt}_3$  and  $\text{UPt}_{3-x}\text{Pd}_x$  also show strong preferential orientation. For these reasons, we prepared, using arc melting under a purified Ar atmosphere, buttons of high-purity polycrystalline  $\text{U}(\text{Pt}_{0.94}\text{Pd}_{0.06})_3$  (Ames Laboratory 99.99% purity electrotransport-refined U and Johnson Matthey 99.9985% purity Pt) and aligned them for measurements in a field using single-crystal  $\chi$  results,<sup>8</sup> such that the samples were primarily oriented in the basal plane (high susceptibility direction). As will be seen in our data below, we achieved a value for  $\chi(T \rightarrow 0)$  for  $H_{-ab \text{ plane}} = 0.5$  T of 13.3 memu/mole, comparable with the value<sup>8</sup> for single-crystal  $\text{U}(\text{Pt}_{0.95}\text{Pd}_{0.05})_3$ ,  $H$  in the  $ab$  plane, from van Spang<sup>8</sup> of 13.6 memu/mole ( $\chi \perp$  basal plane is about 5 memu/mole). Thus, the use of magnetic susceptibility allows the good alignment of the preferentially oriented sample in the  $ab$  plane for measurements in field. The  $ab$ -plane alignment of field is the direction in pure<sup>11</sup> as well as Pd-doped<sup>8</sup>  $\text{UPt}_3$  where the metamagnetic transition is observed.

The specific heat,<sup>12</sup> resistivity,<sup>12</sup> and magnetic susceptibility<sup>13</sup> were measured using established techniques.

### III. RESULTS

#### A. Resistivity

The resistivity of  $\text{U}(\text{Pt}_{0.94}\text{Pd}_{0.06})_3$  between 0.09 and 10 K is shown as an inset in Fig. 1. The antiferromagnetism at 6 K is clearly seen as an anomaly in  $\rho$ . The residual resistivity for our sample, 113  $\mu\Omega$  cm, has approximately a  $\pm 5\%$  error

bar due to uncertainties in the geometrical factors, while for the residual resistivity ratio [ $\text{RRR} = \rho(300 \text{ K}) / \rho(T \rightarrow 0 \text{ K})$ ] of 1.86, these uncertainties cancel. These values are comparable to those of Verhoef *et al.*<sup>14</sup> on a similar (but not identical) composition of polycrystalline  $\text{U}(\text{Pt}_{0.95}\text{Pd}_{0.05})_3$  ( $\rho_0 = 101 \mu\Omega$  cm and  $\text{RRR} = 2.49$ ), particularly when one considers the trend with increasing Pd concentration (Verhoef *et al.*'s 10% Pd sample has  $\rho_0 = 633 \mu\Omega$  cm and  $\text{RRR} = 0.85$ ).

The limiting  $T \rightarrow 0$  temperature dependence of  $\rho$  is clearly difficult to determine from previously published<sup>8,14</sup> data only down to 1.4 K, since as shown in the inset in Fig. 1, the data appear to be crossing over to another region of temperature dependence at lower temperatures. Thus, Fig. 1 shows  $\rho$  data between 0.09 and 0.6 K on an expanded scale. Since the variation of  $\rho$  from 0 to 0.6 K is less than 1% of  $\rho_0$ , rather good precision and a large number of data points are necessary in order to accurately determine the temperature dependence. These data fit  $\rho = \rho_0 + AT^\alpha$ , where  $\alpha = 2.0 \pm 0.1$ . Clearly, within our error bar, this is Fermi-liquid,  $\rho \sim T^2$ , behavior. Data (not shown) were taken on the same sample in 13 T applied field at the National High Magnetic Field Laboratory (NHMFL) and, within a somewhat larger error bar due to electrical and vibrational noises in the magnetic field causing greater scatter in the data below 0.25 K, also gave  $\alpha \approx 2 \pm 0.2$ . Thus, the resistivity, with its Fermi-liquid behavior, seems dominated by normal quasiparticle scattering processes giving  $\rho \sim T^2$  behavior. Since  $\Delta\rho$  in this low-temperature fit range (0.09–0.6 K) is indeed small when expressed as a percentage of  $\rho_0$ , one might ask about other possible contributions, e.g., as seen<sup>15</sup> in very pure elemental metals at low temperatures, that  $\rho$  could be better fit to both a  $T^2$  and an electron-phonon Bloch  $T^5$  dependence. Based on an estimate—using specific-heat data<sup>16</sup> for  $\text{U}(\text{Pt}_{0.8}\text{Pd}_{0.2})_3$ —of 214 K (Ref. 17) for the Debye temperature (proportional to lattice stiffness and an indicator of the degree to which phonons are excited at low temperature), the expected<sup>15</sup> Bloch  $T^5$  contribution would be at least three orders of magnitude smaller than the  $\sim 1 \mu\Omega$  cm contribution to  $\rho$  at 0.6 K seen here in our data from the quasiparticle scattering.

#### B. Magnetic susceptibility

The susceptibility data from 0.4 to 40 K of  $\text{U}(\text{Pt}_{0.94}\text{Pd}_{0.06})_3$  are shown in Fig. 2. The antiferromagnetic anomaly around 6 K is clearly seen. Consistent with the data down to 2 K as discussed above, the extension of the lowest temperature data to 0.4 K (see inset of Fig. 2 for an expanded view) shows no signs of a nFl divergence as  $T \rightarrow 0$ . Rather, the data approach a constant value approximately quadratically with temperature ( $\chi \sim a + bT^\alpha$ , where  $\alpha = 1.9 - 2.3$ , dependent on field and temperature ranges of the fit) as is consistent with the behavior of a Fermi liquid and, as shown in the inset to Fig. 2, similar to the temperature dependence of  $\chi$  of the heavy Fermion antiferromagnet  $\text{U}_2\text{Zn}_{17}$  (Ref. 18) below its  $T_N = 9.7$  K. This is strongly in contrast to the behavior<sup>2,3</sup> of most non-Fermi-liquid systems that show  $C/T \sim \log T$ , where  $\chi$  shows divergent (e.g.  $\chi \sim T^{-1+\lambda}$  or  $\log T$ ) behavior or at least increasing behavior as  $T \rightarrow 0$ .

#### C. Specific heat

The specific-heat data of high-purity polycrystalline  $\text{U}(\text{Pt}_{0.94}\text{Pd}_{0.06})_3$  plotted vs  $\log T$  in 0 and 13 T are shown in

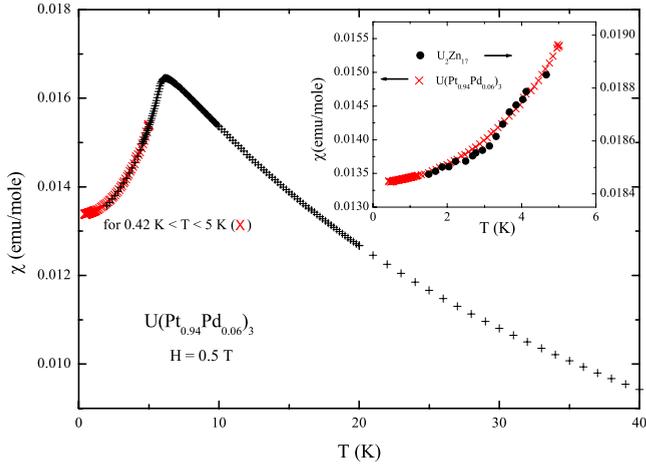


FIG. 2. (Color online) Magnetic susceptibility in 0.5 T from 0.4 to 5 K of  $U(\text{Pt}_{0.94}\text{Pd}_{0.06})_3$  measured in a Faraday magnetometer, together with data from 2–10 K taken on an adjacent piece of sample in a Quantum Design MPMS system. Note the anomaly at  $T_N \sim 6$  K. In the expanded view in the insert, data (Ref. 18) for the heavy Fermion antiferromagnet  $U_2\text{Zn}_{17}$ ,  $T_N = 9.7$  K, are also shown for comparison down to 1.4 K.

Fig. 3. Below the peak in  $C/T$  at the antiferromagnetic transition, the data follow  $\log T$  below 2 K even without the  $T < 6$  K higher temperature background contribution—with its strong magnon temperature dependence—subtracted off. With a straightforward fit to the data just below  $T_N$  subtracted, Ref. 6 showed that  $\Delta C/T \sim \log T$  over more than a decade in temperature. However, rather than provide such a fit here, our high density 13 T  $C/T$  data presented in Fig. 3—with the majority of the magnon contribution to  $C/T(B=0)$  below  $T_N$  suppressed by the field—show that  $C/T \sim \log T$  extends to 4 K without the necessity of any fitting

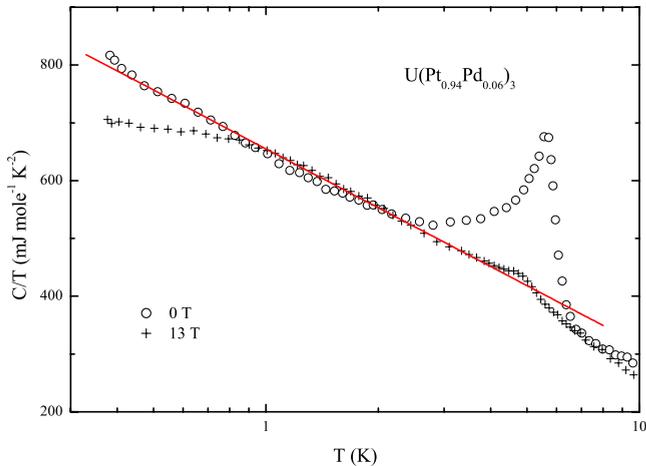


FIG. 3. (Color online) Specific heat divided by temperature  $C/T$  vs  $\log T$  in 0 and 13 T applied magnetic field. The red solid line is a fit of the low-temperature zero-field data to  $\log T$ . Note that with the suppression of the magnon contribution to  $C$  caused by the 13 T applied field that  $C/T(13 \text{ T})$  varies as  $\log T$  up to about 4 K. Note further the saturation behavior in  $C/T(13 \text{ T})$  below about 0.9 K, implying that field suppresses the nFI behavior in  $U(\text{Pt}_{0.94}\text{Pd}_{0.06})_3$  just as it does<sup>2,3</sup> in other nonmagnetic nFI systems.

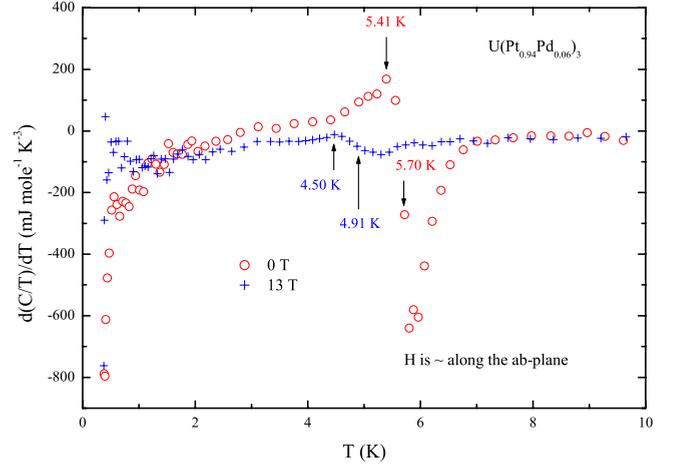


FIG. 4. (Color online) The first derivative of  $C/T$  vs  $T$  to delineate more accurately the suppression of  $T_N$  with 13 T magnetic field. Two temperatures in each field are marked with an arrow, the midpoint and the low-temperature-end finish of the jump in  $d(C/T)/dT$ . Thus, the suppression of the antiferromagnetic ordering anomaly in 13 T is approximately 5.70–4.91 K (0.79 K) or 5.41–4.50 K (0.91 K).

procedure. Thus, the field data in 13 T shown in Fig. 3, where the magnon contribution is much reduced and  $C_{\text{measured}}/T(13 \text{ T})$  behaves as  $\log T$  up to above 4 K, support the original<sup>6</sup> claim that  $C/T \sim \log T$  over more than a decade in temperature *below* the magnetic ordering temperature in  $U(\text{Pt}_{0.94}\text{Pd}_{0.06})_3$ .

Note that the 13 T specific-heat data show a tendency toward saturation ( $\Leftrightarrow$ Fermi liquid) behavior below 0.9 K as  $C/T$  deviates below the higher temperature  $\log T$  divergence, as is typical<sup>2</sup> of non-Fermi-liquid systems when sufficient field is applied.

As a last point of interest, Fig. 4 shows  $d(C/T)/dT$  for 0 and 13 T in order to precisely determine the field dependence of the antiferromagnetic ordering temperature. 13 T suppresses  $T_N(H=0)$  by 0.8–0.9 K, compared to<sup>4</sup> 5 T suppressing  $T_N$  in  $U(\text{Pt}_{0.95}\text{Pd}_{0.05})_3$  by  $\sim 0.4$  K.

#### IV. CONCLUSIONS

Based on the zero-field specific heat,  $U(\text{Pt}_{0.94}\text{Pd}_{0.06})_3$ —as first discussed in Ref. 6—clearly shows non-Fermi-liquid behavior, as well as the expected<sup>2</sup> re-entry into the Fermi-liquid state with applied field. Thus,  $U(\text{Pt}_{0.94}\text{Pd}_{0.06})_3$  seems to be one of only approximately seven examples<sup>2,3</sup> of coexistent magnetic and nFI behavior. Now with magnetic-susceptibility data down to 0.4 K and resistivity data down to 0.09 K,  $U(\text{Pt}_{0.94}\text{Pd}_{0.06})_3$  seems to be a rare example of a nFI system where the fluctuations responsible for the nFI behavior in the specific heat are both weak at  $q=0$  (where the uniform magnetic-susceptibility behavior is determined) and at  $q$  vectors that would cause significant scattering in the resistivity, since both  $\chi$  and  $\rho$  exhibit Fermi-liquid behavior. Although neutron-scattering measurements of nFI system are lengthy and difficult to perform, existing<sup>2,3,7</sup> only for a few well-studied nFI materials, such measurements for

$U(\text{Pt}_{0.94}\text{Pd}_{0.06})_3$  to investigate the nFI fluctuations as a function of  $q$  would be of interest.

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