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High-pressure investigation of the heavy-fermion antiferromagnet $U_3Ni_5Al_{19}$

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Measurements of magnetic susceptibility, specific heat, and electrical resistivity at applied pressures up to 55 kbar have been carried out on single crystals of the heavy-fermion antiferromagnet $U_3Ni_5Al_{19}$, which crystallizes in the $Gd_3Ni_5Al_{19}$ orthorhombic structure with two inequivalent U sites. At ambient pressure, an upturn of the specific heat and T-linear electrical resistivity below 5 K indicate non-Fermi liquid (NFL) behavior in the presence of bulk antiferromagnetic order at $T_N = 23$ K. Electrical resistivity measurements reveal a crossover from non-Fermi liquid to Fermi liquid behavior at intermediate pressures between 46 and 51 kbar, followed by a return to NFL $T^{3/2}$ behavior at higher pressures. These results provide evidence for an ambient pressure quantum critical point and an additional antiferromagnetic instability at $P_c \approx 60$ kbar.

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A considerable amount of experimental and theoretical effort has been devoted in recent years to the investigation of quantum criticality in f-electron heavy-fermion materials (for a recent review, see Ref. 1). Until now, most research has focused on the behavior of Ce-based heavy-fermion systems in the vicinity of an antiferromagnetic (AFM) quantum critical point (QCP) in which the Néel temperature T_N is tuned to absolute zero by an external control parameter such as composition x, pressure P, or magnetic field H. Measurements under applied pressure on these systems have been particularly useful in accessing the QCP and also exploring the unusual power law or logarithmic T-dependences of the physical properties, characteristic of non-Fermi liquid (NFL) behavior, found near the critical point. For instance, at a critical pressure $P_c = 28$ kbar necessary to completely suppress antiferromagnetism in $CePd_2Si_2$,[2] the electrical resistivity exhibits a power law, i.e., $\rho - \rho_0 = AT^n$, with $n = 1.2$ over an extended range in temperature, in contrast to the T^2 Fermi-liquid behavior expected for a simple metal. The antiferromagnetic Ce-based heavy-fermion compounds have proved unstable to the formation of superconductivity in the vicinity of the AFM QCP. The transition temperatures are often quite low, $T_c \sim 0.4$ K (0.2 K) for $CePd_2Si_2$ ($CeIn_3$) at $P = 28$ kbar (26 kbar);[2] more recently, superconductivity has been observed at $T_c \sim 2$ K in antiferromagnetic $CeRhIn_5$ above 15 kbar,[3] more than half the value of the Néel temperature at ambient pressure ($T_N = 3.8$ K). The occurrence of superconductivity near the AFM QCP where spin fluctuations are strongest is indicative of an unconventional magnetically mediated pairing mechanism in these compounds. Therefore it is of interest to investigate pressure-induced quantum criticality in antiferromagnetic U-based heavy-fermion materials as comparatively fewer such studies have been performed.[4] To this end, we present measurements of magnetic susceptibility, specific heat, as well as electrical resistivity up to 55 kbar of the heavy-fermion antiferromagnet $U_3Ni_5Al_{19}$.

$U_3Ni_5Al_{19}$ crystallizes in the orthorhombic $Gd_3Ni_5Al_{19}$ structure[5] (space group *Cmcm*, No. 63) with two inequivalent U sites (one U atom in 4c and two U atoms in 8f) which can be thought of as an intergrowth between the imaginary structures of $YbNiAl_4$ (with the orthorhombic $YNiAl_4$-type structure[6]) and that of $Yb_2Ni_4Al_{15}$ (monoclinic). A recent report[7] concluded that $U_3Ni_5Al_{19}$ orders antiferromagnetically at $T_N = 23$ K, due to a prominent feature in $\chi(T)$ for $H \| c$ involving one of the two distinct U sites, while the other site showed no sign of magnetic order down to 50 mk.

Single crystals of $U_3Ni_5Al_{19}$ were grown in Al flux. The elements were placed in the ratio U:Ni:Al=1:1:10 in an alumina crucible sealed under vacuum in a quartz tube. The sample was heated to 1100 °C and kept at that temperature for 4 h, then slowly cooled to 650 °C at 7 °C h^{-1}, at which point excess Al flux was removed in a centrifuge. The resulting crystals were needles with typical dimensions $1 \times 1 \times 5$ mm^3. The orthorhombic $Gd_3Ni_5Al_{19}$ structure was confirmed by single crystal x-ray diffraction with lattice parameters $a = 4.0850(2)$ Å, $b = 15.9305(8)$ Å, and $c = 26.959(1)$ Å, (further details of the structural refinement can be found in Ref. 8). Magnetic susceptibility measurements were performed in a commercial magnetometer from 2 to 350 K in a magnetic field $H = 0.1$ T both parallel and perpendicular to the long axis of a single crystal. It was then concluded on the basis of the measurements[7] that the long axis of the crystal was the b axis. Specific heat measurements were made using a commercial calorimeter from 0.4 to 50 K on a collection of single crystals using an adiabatic method. Electrical resistivity measurements under pressure were carried out using a profiled toroidal anvil clamped device with anvils supplied with a boron-epoxy gasket and Teflon capsule, containing pressure-transmitting liquid, sample, and a pressure sensor.[9] The pressure was determined from the variation of the superconducting transition of lead using the pressure scale of Eiling and Schilling.[10] A standard four-probe technique was performed using an LR-700 Linear Research bridge operating at a current of 1 mA applied along the b axis of the crystal.

The magnetic susceptibility $\chi \equiv M/H$ of $U_3Ni_5Al_{19}$ in a magnetic field $H = 0.1$ T along various crystallographic directions is shown Fig. 1. A clear signature of a magnetic transition, presumably antiferromagnetic, is found in χ_c at $T_N = 23$ K, while χ_b exhibits temperature independent paramag-

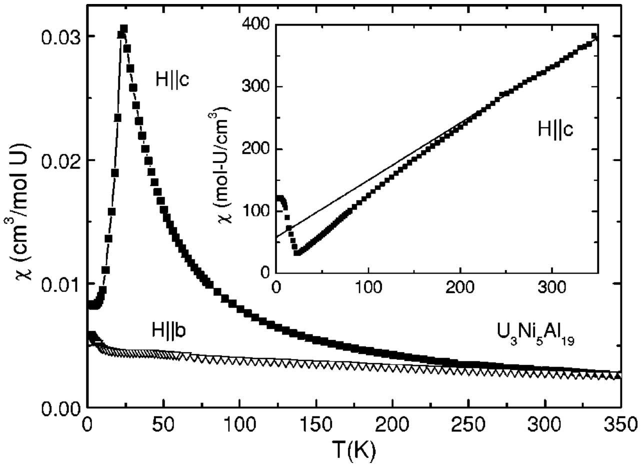

FIG. 1. Magnetic susceptibility $\chi(T)$ in a magnetic field $H = 0.1$ T along the *b* and *c* axes. Inset: Inverse magnetic susceptibility $\chi^{-1}(T)$ for $H \| c$. The solid line is a linear fit of the data.

netism, indicating marked magnetic anisotropy. The χ_c data can be fit to a Curie-Weiss law above 250 K yielding an effective moment $\mu_{eff} = 3.0\ \mu_B/\mathrm{U}$ atom and Curie-Weiss temperature $\theta_{CW} = -79$ K as shown in the inset of Fig. 1.

The specific heat $C(T)$ of $U_3Ni_5Al_{19}$ and nonmagnetic $Th_3Ni_5Al_{19}$ is shown in Fig. 2(a). The specific heat of $Th_3Ni_5Al_{19}$ is characterized by a Sommerfeld coefficient $\gamma = 1\ \mathrm{mJ/mol\ Th\ K^2}$ and a Debye temperature $\theta_D = 370$ K. An anomaly in $U_3Ni_5Al_{19}$ is observed at $T_N = 23$ K confirming bulk AFM order. A second, much smaller feature is found at ~13 K and is attributed to an impurity phase as no such feature is observed in $\chi(T)$ or $\rho(T)$, although a spin-reorientation of the U moments cannot be ruled out. A volume fraction of 1% is estimated from the measured magnetic entropy of this small anomaly ($S_{mag} \sim 100\ \mathrm{mJ/mol\ U}$), assuming that the impurity phase contributes $R\ln(2)$ of entropy below the transition at ~13 K. Powder x-ray diffraction measurements reveal no evidence for impurity phases (with an upper limit of a few percent). Below 5 K, C_{el}/T exhibits an upturn as the temperature is lowered characteristic of non-Fermi liquid behavior, suggesting proximity to a quantum critical point. The electronic contribution C_{el} to the specific heat of $U_3Ni_5Al_{19}$, plotted as C_{el}/T vs T, is shown in Fig. 2(b), where the nonmagnetic contribution of $Th_3Ni_5Al_{19}$ has been subtracted from the $C(T)$ data. A significant quasiparticle mass enhancement is indicated by the large value of $C_{el}/T = 185\ \mathrm{mJ/mol\ U\ K^2}$ at $T = 0.4$ K. The electronic entropy $S_{el}(T) = \int (C_{el}/T) dT$ released below T_N amounts to $S_{el}(23\ \mathrm{K}) = 3.9\ \mathrm{J/mol\ U\ K}$ as shown in Fig. 2(b). This value amounts to $(0.67)R\ln(2)$, considerably less than $R\ln(9)$ or $R\ln(10)$ expected for a $5f^2$ ($J=4$) or $5f^3$ ($J=9/2$) electronic configuration, respectively. The C_{el}/T data below 5 K can be fit by the spin-fluctuation model of Moriya and Takimoto[11] describing the contribution of critical spin fluctuations to the specific heat. In this theory, the anomalous NFL temperature dependences of $C(T)$ and $\rho(T)$ are calculated as a function of reduced temperature T/T_0, with a control parameter y_0 denoting the distance from the QCP (i.e., $y_0 = 0$ at the QCP) that provides a measure of the inverse correlation length. Two additional parameters are needed for comparison to experiment, the first of which, T_0, is related to the exchange energy by $T_0 = \mathcal{J}/(2\pi^2)$; the second parameter is the contribution to the electronic specific heat of noncritical fermions γ_0. A fit of this spin-fluctuation theory is displayed in Fig. 2(b), yielding $y_0 = 0.001$, $T_0 = 3.2$ K, and $\gamma_0 = 80\ \mathrm{mJ/mol\ U\ K^2}$, suggesting $U_3Ni_5Al_{19}$ is close to a $P = 0$ quantum critical point. After subtracting this NFL contribution from the $C_{el}(T)$ data of $U_3Ni_5Al_{19}$, the magnetic contribution to the specific heat C_{mag} (not shown) is reasonably well-described by a T^3 power law in the antiferromagnetic state characteristic of antiferromagnetic spin-wave excitations.[12]

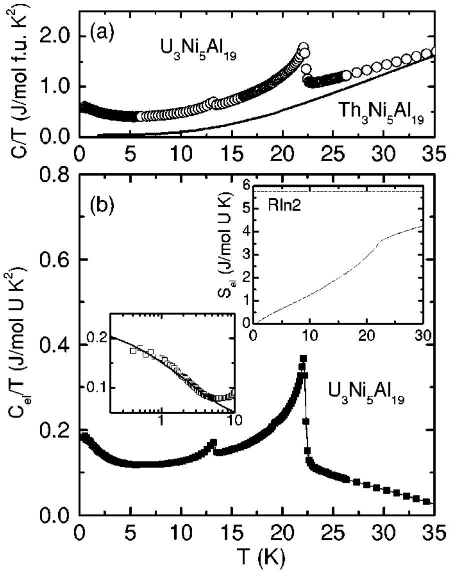

FIG. 2. (a) Specific heat $C(T)$ of $U_3Ni_5Al_{19}$ and nonmagnetic $Th_3Ni_5Al_{19}$. (b) Electronic contribution C_{el} to the specific heat of $U_3Ni_5Al_{19}$, plotted as C_{el}/T vs T. Lower inset: $C_{el}(T)/T$ below 10 K. The solid line is a fit to the spin-fluctuation theory discussed in the text. Upper inset: Electronic entropy $S_{el}(T)$.

The results of electrical resistivity measurements under pressure of $U_3Ni_5Al_{19}$ are presented in Fig. 3 at four different pressures. The $\rho(P,T)$ curves have "s-shaped" curvature typical of spin fluctuations systems. The magnetic phase transition is readily visible as a kink in $\rho(T)$ at $T_N = 23$ K at ambient pressure. Upon further cooling, the resistivity is linear in temperature below 5 K consistent with the NFL behavior observed in specific heat. The application of pressure suppresses the Néel temperature to $T_N = 2.4$ K at $P = 55.2$ kbar, the highest pressure reached in this experiment, as shown in the lower inset of Fig. 3. The derivative $d\rho/dT$ is characteristic of a second order phase transition[13] and reveals a similar suppression of the magnetic transition as displayed in the upper inset of Fig. 3.

Figure 4 shows the low temperature power law fits ($\rho - \rho_0 = AT^n$) to the $\rho(P,T)$ data of $U_3Ni_5Al_{19}$. A linear T-dependence describes the data for $P \leqslant 25.6$ kbar, whereas an exponent $n = 1.3-1.7$ is needed to fit the $\rho(T)$ curves between $P = 31.3$ and 42.4 kbar over nearly a decade in temperature below 10 K. At higher pressures of $P = 46.4$ and 51.3 kbar, a Fermi-liquid ground state is revealed ($n=2$); however, a NFL-like exponent $n = 1.5$ is again found at the

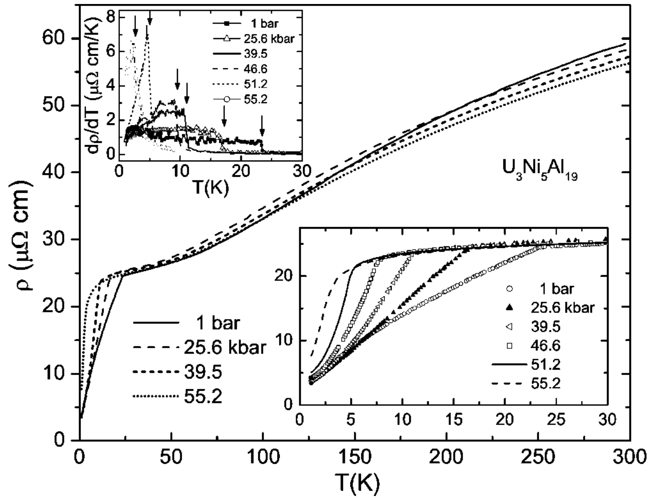

FIG. 3. Electrical resistivity $\rho(T)$ of $U_3Ni_5Al_{19}$ at various pressures P. Lower inset: $\rho(P,T)$ below 30 K. Upper inset: $d\rho/dT$ vs T at various pressures. The arrows indicate the Néel temperature T_N.

highest pressure $P=55.2$ kbar where the Néel temperature ($T_N=2.4$ K) is almost completely suppressed to $T=0$ K, indicating the influence of critical spin fluctuations at a QCP of $P_c\sim 60$ kbar.

Figure 5 provides a summary of our electrical resistivity measurements under pressure on $U_3Ni_5Al_{19}$. The power law T-dependence of $\rho(T)$ for $P\leqslant 42.4$ kbar indicates a NFL ground state in the presence of long-range magnetic order. A Fermi-liquid ground state is stabilized at intermediate pressures between 42.4 and 51.3 kbar; at the highest pressure $P=55.2$ kbar, critical fluctuations associated with the assumed AFM QCP at $P_c\sim 60$ kbar once again lead to a NFL exponent of the electrical resistivity of $n=1.5$ [Fig. 5(a)]. Both the A coefficient obtained from the power law fits and the residual resistivity (at $T=1$ K) increase dramatically upon approaching the AFM QCP (Fig. 5); such critical scattering arising from proximity to a QCP is typical of many heavy fermion systems such as $CeAgSb_2$,[14] UGe_2,[15,16] and $CeIn_3$.[17] A fit of the $T_N(P)$ data to a power law of the form $T_N\propto|P-P_c|^{\alpha}$ yields a critical pressure $P_c=58$ kbar and exponent $\alpha=0.7$, although a linear variation of $T_N(P)$ is found for $P\geqslant 40$ kbar.

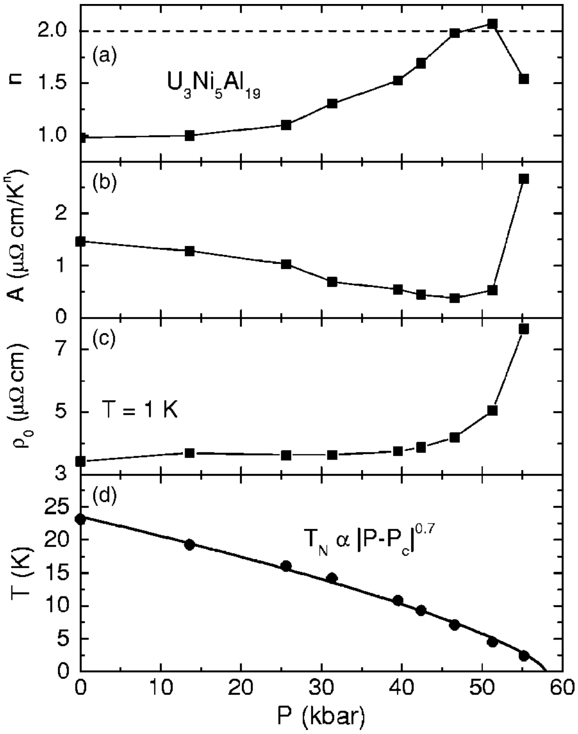

FIG. 5. Power law exponent n, coefficient A, residual resistivity ρ_0, and Néel temperature T_N vs P in (a)–(d), respectively. The lines are guides to the eye, except in (d) where the line is a power law fit of the data.

Our results on $U_3Ni_5Al_{19}$ provide evidence for both a non-Fermi liquid state at ambient pressure and pressure-induced quantum critical point at $P_c\simeq 60$ kbar. It is expected that for temperatures sufficiently below the Néel tempera-

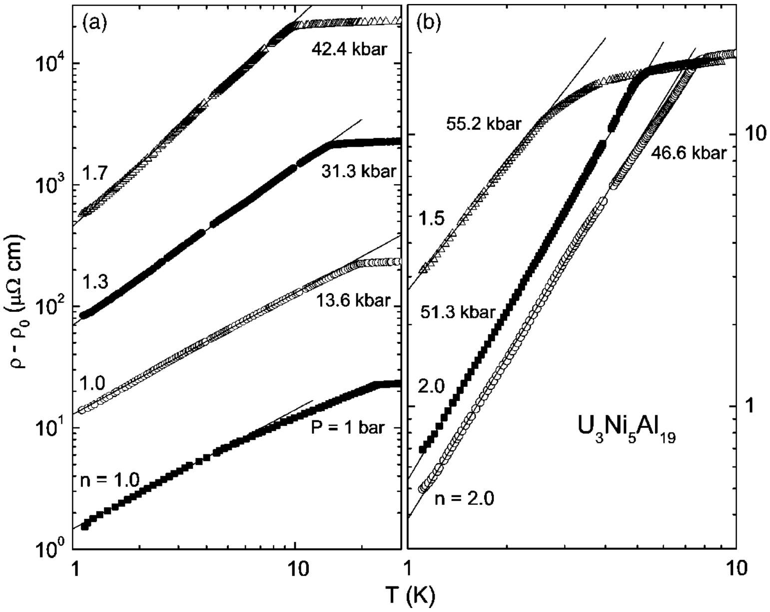

FIG. 4. (a) $\rho-\rho_0$ vs T for $P\leqslant 42.4$ kbar on a log-log scale. Each of the curves have been shifted vertically by one decade from the curve below it for clarity. (b) $\rho-\rho_0$ vs T for 46.6 kbar $\leqslant P\leqslant 55.2$ kbar (the curves have not been shifted vertically). The solid lines in (a) and (b) are power law fits of the data to $\rho-\rho_0=AT^n$, where the exponent n is indicated next to each curve.

ture, the electrical resistivity should exhibit a T^5 temperature dependence due to electron-magnon scattering.[18] Instead, $\rho(T)$ follows a T-linear dependence below ~ 5 K, indicating (along with the upturn in specific heat) NFL behavior coexisting with antiferromagnetism at $P=0$ perhaps associated with an AFM QCP. The spin-fluctuation theory of Moriya-Takimoto describes the specific heat at ambient pressure reasonably well; but this model predicts a $T^{3/2}$ variation of the electrical resistivity that is not observed experimentally. Similar apparently contradictory results have been obtained for the heavy-fermion antiferromagnet $\mathrm{Ce_7Ni_3}$, which has three inequivalent Ce sites. In this compound, AFM order is suppressed by modest pressures of $P_c \simeq 4$ kbar. Non-Fermi liquid behavior [i.e., $C/T \sim -\ln(T/T_0)$] is found within the AFM state,[19] possibly associated with one or more of the Ce sites. We consider two alternate mechanisms that might give rise to the NFL behavior observed at $P=0$. The first is the Kondo disorder model in which a distribution of Kondo temperatures due to chemical or structural disorder leads to non-Fermi liquid behavior, i.e., $\rho(T) \propto T$ and $C/T \propto -\log(T)$,[20,21] consistent with the properties of $\mathrm{U_3Ni_5Al_{19}}$, assuming C/T exhibits a $-\log(T)$ dependence. However, disorder effects do not appear to play a major role as single crystals of stoichiometric $\mathrm{U_3Ni_5Al_{19}}$ used for this investigation have small values of both the overall electrical resistivity [$\rho(300\ \mathrm{K}) \sim 50\ \mu\Omega\,\mathrm{cm}$] and residual impurity scattering ($\rho_0 \sim 3\ \mu\Omega$ cm). A second scenario in which the NFL behavior is associated with the possible impurity phase that may produce the anomaly in specific heat at ~ 13 K can be ruled out as no such anomaly is observed in $\rho(T)$ at 13 K, while a linear T-dependence is found below 5 K. Therefore both the upturn in $C(T)$ and linear T-dependence of $\rho(T)$ are intrinsic properties of $\mathrm{U_3Ni_5Al_{19}}$, and do not derive from disorder or impurities. The application of pressure drives the system out of the $P=0$ NFL state, inducing Fermi liquid behavior between $P=42.2-51.3$ kbar, and at the same time suppressing the magnetic transition. A presumed second pressure-induced quantum critical point at $P_c \sim 60$ kbar leads to the anomalous behavior of ρ_0, A, and n above 51.3 kbar. Behaviors such as the linear variation of T_N with control parameter $|P-P_c|$ [for $P \geqslant 40$ kbar], the $T^{3/2}$-dependence of $\rho(T)$ near P_c, and the steep increase of A close to P_c are consistent with the predictions for a two-dimensional AFM quantum critical point.[22] Clearly, additional measurements are necessary to determine the nature of the possible QCP at ambient pressure and the second one located at the antiferromagnetic instability at $P_c \sim 60$ kbar.

An important issue that remains unresolved is whether one or both of the distinct U sites is involved in the quantum criticality and antiferromagnetic order at ambient pressure in $\mathrm{U_3Ni_5Al_{19}}$. Three possibilities exist: (1) one U site is responsible for both the NFL behavior and the AFM order; (2) the NFL behavior involves one U site while antiferromagnetism is associated with the remaining U site; or (3) both U sites are responsible for the NFL behavior and the AFM order. Unfortunately, the current measurements are indecisive. Scenarios (1) and (3) are at least plausible as the coexistence of magnetism and NFL characteristics has been observed before, both in antiferromagnets[23] and ferromagnets.[24] Of these two possibilities, the reduced electronic entropy in $\mathrm{U_3Ni_5Al_{19}}$ tends to favor scenario (1) in which the singly occupied U site is associated with both NFL and AFM phenomena and the remaining doubly occupied U site exhibits temperature independent paramagnetism. It is interesting to note that the reduced electronic entropy in $\mathrm{U_3Ni_5Al_{19}}$ [$S_{el}(T_N) \sim 3.9\ \mathrm{mJ/mol\,U\,K}$] is similar to U-based antiferromagnets such as $\mathrm{U_2Zn_{17}}$ [$S_{el}(T_N) \sim 3.8\ \mathrm{mJ/mol\,U\,K}$] and $\mathrm{UCd_{11}}$ [$S_{el}(T_N) \sim 6.5\ \mathrm{mJ/mol\,U\,K}$], although a sizable fraction of $R\ln(9)$ is released below T_N in other compounds (e.g., $\mathrm{UCu_5}$, $\mathrm{UAgCu_4}$).[25] The reduced entropy also favors model (2) which provides a natural explanation for the occurrence of AFM order and NFL behavior at ambient pressure and has been suggested previously.[7] Neutron scattering measurements on $\mathrm{U_3Ni_5Al_{19}}$ would be invaluable for determining which U site(s) is (are) responsible for the magnetic ordering.

In summary, measurements of magnetic susceptibility, specific heat, and electrical resistivity at applied pressures up to 55 kbar have been carried out on single crystals of the heavy-fermion antiferromagnet $\mathrm{U_3Ni_5Al_{19}}$. The low-T upturn of the specific heat and T-linear electrical resistivity below 5 K indicates non-Fermi liquid behavior at ambient pressure in the presence of bulk antiferromagnetic order at $T_N=23$ K. Electrical resistivity measurements reveal a crossover from NFL to FL behavior at intermediate pressures between 46 and 51 kbar. The pressure dependence of the physical properties suggest proximity to an AFM QCP at a critical pressure $P_c \approx 60$ kbar.

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