Unusual temperature dependence in the low-temperature specific heat of $U_3Ni_5Al_{19}$

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Specific heat has been measured down to 0.053 K on a single crystal of the heavy-fermion antiferromagnet $U_3Ni_5Al_{19}$ that orders at $T_N=23$ K. As has been previously reported, these data can be fitted between 0.4 and 4 K by the spin-fluctuation model of Moriya and Takimoto, which describes the contribution of weakly interacting critical spin fluctuations to the specific heat, C, where, as $T\to 0$, $C/T=\gamma_0-a\sqrt{T}$. However, below 0.35 K a noticeable divergence in $C/T\sim\log T$ dependence, consistent with the existence of strongly interacting fluctuations, is observed. This increase in the divergence of C/T at the lowest temperatures—which is contrary to the self-consistent renormalization theory of Moriya and Takimoto, which predicts $\sqrt{T}$ dependence for C/T as $T\to 0$ and $\log T$ dependence at *higher* temperatures—has been measured as a function of magnetic field to further understand its origin. The field data in the low-temperature regime, where $C/T\sim\log T$ exhibit scaling with $\Delta B/T^{1.9}$, further evidence that there exist strongly interacting fluctuations below 0.35 K in $U_3Ni_5Al_{19}$.

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Intense experimental and theoretical efforts have been dedicated to understanding non-Fermi liquid (nFl) systems in recent years (for reviews, see Ref. 1) and the unusual temperature dependences in their low-temperature properties. Many systems have been discovered which exhibit quantum criticality. Among those systems the typical type of nFl system has been a magnetically ordered Ce- or U-heavy Fermion system tuned to the proximity of an antiferromagnetic quantum critical point in the phase diagram using doping, pressure, or magnetic field. Recently $U_3Ni_5Al_{19}$ has been reported[2] as one of the rare uranium systems with *intrinsic* nFl behavior, joining the limited number of other such undoped U systems such as U_2Pt_2In,[3,4] U_2Co_2Sn,[5] and $U_3Ni_3Sn_4$.[6] In such "intrinsic" nFl (U or Ce) compounds the critical point in the phase diagram occurs in the pure system under normal ($P=H=0$) conditions. However the nFl behavior of some of these pure nFl compounds is limited in temperature range; also some compounds exhibit saturation in C/T at lower temperatures.[1]

Previously specific-heat measurements have been conducted[2] on single crystals of the heavy-fermion antiferromagnet, $T_N=23$ K, $U_3Ni_5Al_{19}$, down to 0.4 K. The spin-fluctuation theory of Moriya and Takimoto[7] describes those specific-heat data reasonably well between 0.4 and 4 K. Also, additional indications of nFl behavior have been noticed in the experimental results of a linear temperature dependence of the electrical resistivity (i.e., the electrical resistivity deviates from the Fermi liquid $\rho\sim T^2$ behavior) below $T=5$ K. This unusual behavior appears to occur in spite of the presence of bulk antiferromagnetic order at 23 K.

This work presents measurements of the specific heat below 0.4 K in order to further investigate the extent of the Moriya-Takimoto fit over a broader temperature range and to study the coexistent nFl and antiferromagnetic behavior upon further cooling.

The procedure of single crystal growth of $U_3Ni_5Al_{19}$ in Al flux has been previously reported,[2] and x-ray powder diffraction has confirmed no evidence of impurity phases. Low-temperature specific-heat measurements of $U_3Ni_5Al_{19}$ below 0.4 K in zero and applied magnetic field were carried out on a small single crystal (1.5 mg) using a thermal relaxation method[8] in the SCM-1 dilution refrigerator at the National High Magnet Field Laboratory in Tallahassee. Measurements on this crystal at temperatures ranging from 0.4 K up to 3.5 K were performed with a He^3 calorimeter using techniques described previously[9] and agree within the relative error bars ($\pm 3\%$) with the published[2] data.

The electronic contribution to the specific heat of $U_3Ni_5Al_{19}$ is shown in Fig. 1 plotted as $\Delta C/T$ vs $\log(T)$. The electronic contribution to the specific heat of $U_3Ni_5Al_{19}$, $\Delta C(T)$, is evaluated from the measured specific heat $[\Delta C(T)=C_{\text{measured}}(T)-C_{\text{ph}}(T)]$, where $C_{\text{ph}}(T)$ is the phonon contribution using results[2] ($\theta_D=370$ K) for nonmagnetic $Th_3Ni_5Al_{19}$. The Moriya-Takimoto fit to the spin-fluctuation theory was made and depicted as the solid line in Fig. 1. Overall this Moriya-Takomoto fit works quite well over the decade of temperature range above $T=0.35$ K. However, below 0.35 K, the $\Delta C/T$ data deviate markedly from this fit, following close to logarithmic form down to the lowest temperature (0.053 K).

In the Moriya-Takimoto theory,[7] the anomalous nFl temperature dependence of $C(T)$ is calculated as a function of temperature scaled by T_0, with another parameter y_0 ($y_0=0$ at the magnetic instability) denoting the distance from the QCP. The fit in the low-temperature limit converges to a finite value and starts to show $T^{1/2}$ dependence and then logarithmic behavior as temperature is increased. The parameters from the fit of our data above 0.35 K are $T_0=15.7$ K, $y_0=0.014$, and this fit matches the data up to about 4 K. This application of the theory for weakly interacting spin fluctuations indicates that the system is near its antiferromagnetic instability.

The substantial deviation from this fit *below* 0.35 K is clearly inconsistent with the saturation anticipated from the Moriya-Takimoto theory. In order to investigate the low-temperature region where $C/T\sim\log T$, the possibility was

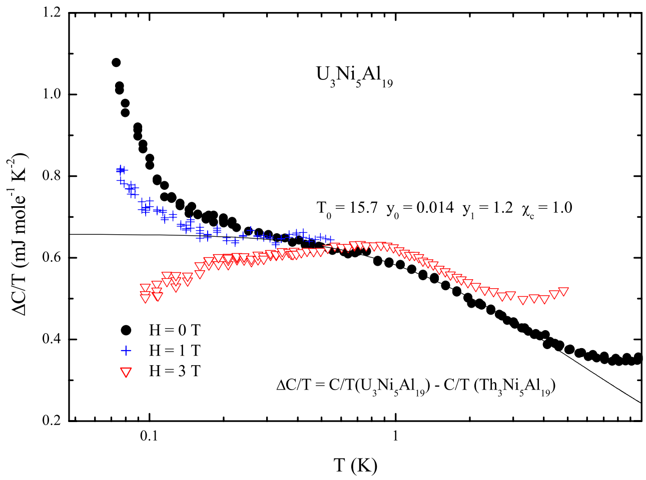

FIG. 1. (Color online) Electronic specific heat ΔC (with the lattice contribution from $Th_3Ni_5Al_{19}$ subtracted) in zero and applied field divided by temperature, T, of $U_3Ni_5Al_{19}$. The solid line is a fit of the data between 0.35 and 4 K to the weakly interacting spin-fluctuation theory of Moriya-Takimoto; the fit parameters from the theory are shown. Data above 0.4 K are from Ref. 2; data taken in the present work agree with these published data to within 5% between 0.4 and 3.5 K. There may be a slight anomaly at around 0.8 K in the zero-field data due to a minor ($<1\%$) impurity phase like seen in the discovery work (Ref. 2) at 13 K.

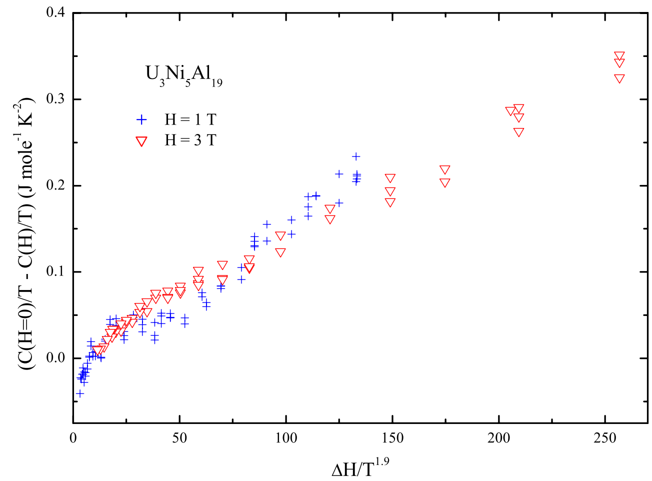

FIG. 2. (Color online) Change in the electronic specific heat of $U_3Ni_5Al_{19}$ with field plotted vs $\Delta H/T^{1.9}$. Such scaling behavior is only consistent (Ref. 10) with strongly interacting fluctuations.

considered that the upturn might be due to the internal field in the antiferromagnetic $U_3Ni_5Al_{19}$ splitting the nuclear levels of ^{27}Al (100% natural abundance) and causing a Schottky anomaly. If the electronic specific-heat data shown in Fig. 1, ΔC, are plotted (not shown) as the difference $\Delta C - C^{\text{Moriya Fit}}$, there is indeed an upturn in this difference at low temperature that over $\sim$0.25 K could be fit approximately to $C_{\text{Schottky}} \propto 1/T^2$. However, as clear from Fig. 1, the amplitude of this upturn is much too small to be explained as field splitting of the ^{27}Al nuclear spin 5/2 levels since the size of the upturn is less than 2% of that which would be expected from the 100% isotopic abundance.

Another method to investigate the $C/T \sim \log T$ divergence below 0.35 K is to measure the field dependence of the specific heat. These data are also shown in Fig. 1. Clearly they are also inconsistent with field splitting of nuclear levels being the cause of the upturn in C/T since an increasing field would increase the size of the splitting and thus the magnitude of the upturn. Instead, these field data appear to behave similarly to data taken on other strong fluctuation non-Fermi liquid systems,[1] where $C/T \sim \log T$ and $C(H)$ data have been shown to scale with $\Delta H/T^{\beta}$. The field data from Fig. 1 were scaled for various values of β; the result of the best scaling (β=1.9) is shown in Fig. 2. Following the discussion of Ref. 10, such scaling of $C(H)$ data implies unequivocally that there are strongly interacting fluctuations in $U_3Ni_5Al_{19}$ below 0.35 K, inconsistent with the weakly interacting fluctuations of the Moriya-Takimoto theory.

In summary, measurements of specific heat down to 0.053 K have been carried out on a single crystal of the heavy-fermion antiferromagnet $U_3Ni_5Al_{19}$. The results contrast with the Moriya-Takimoto theory, which works quite well down to T=0.35 K (with a value y_0=0.014), but then the C/T data begin to diverge more rapidly as $T \rightarrow 0$ and exhibit scaling of the specific-heat data in field proportional to $\Delta H/T^{1.9}$. Both the $C/T \sim \log T$ dependence and the scaling behavior are consistent with strongly interacting fluctuations being present below 0.35 K in $U_3Ni_5Al_{19}$. Such a crossover, from weakly interacting behavior describable by the Moriya-Takimoto theory to strongly interacting behavior at lower temperatures, has not previously been observed.

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