ARTICLES

U_2Co_2Sn: An undoped non-Fermi-liquid system with $C_e \approx \gamma - A\sqrt{T}$

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Investigation of the low-temperature specific heat, magnetic susceptibility, and resistivity of U_2Co_2Sn for expected paramagnon behavior led instead to the discovery of a non-Fermi-liquid system obeying the Millis/Moriya theory prediction of an electronic specific heat that varies approximately as γ-$A\sqrt{T}$ over an appreciable temperature range, in this case over the whole temperature range (0.3–10 K) of measurement. The temperature dependence of the low-temperature resistivity, however, follows $\rho = \rho_0 + AT^{\alpha}$, with $\alpha(\approx 1.8)$ lying below that predicted for a Fermi liquid (i.e., $\alpha = 2$) but above the $\rho = \rho_0 + AT^{1.5}$ predicted by the quantum phase transition, weakly interacting spin fluctuation theory of Millis/Moriya. Scaling of the specific heat with field indicates that the electron interactions responsible for the non-Fermi-liquid behavior are not single ion in nature. Several non-Fermi-liquid theories and their possible applicability to these results are discussed, as well as the possible influence of the relatively small U-U spacing in U_2Co_2Sn on the unusual non-Fermi-liquid behavior.

INTRODUCTION

The discovery of one example of interesting behavior in a compound system, for example, the finding by Steglich *et al.*[1] in 1979 of heavy-fermion superconductivity in $CeCu_2Si_2$ (a member of the $ThCr_2Si_2$ structure type) often leads to further discoveries in other members of the same compound system. Havela *et al.*[2] recently reported heavy-fermion behavior in several U_2T_2X compounds (T = transition metal and X = Sn or In), as well as antiferromagnetism in a number of other members of this system. The upturn in the specific heat C divided by temperature T that they observed in U_2Pt_2In was later tentatively identified[3] as obeying $C/T \sim \log T$—typical non-Fermi-liquid (NFL) behavior. This $C/T \sim -\log T$ dependence was then confirmed[4] between approximately 0.1 and 5 K in work on a single crystal.

The motivation for the present work was as follows. Havela *et al.* state that ''a strong upturn in C/T'' is also found in nonmagnetic U_2Co_2Sn (data not shown). The thesis of Nakotte[5] shows C/T vs T data down to 1.3 K (not fitted to any functional form) for U_2Co_2Sn with a rather shallow minimum ($\sim$ four times less pronounced than that for U_2Pt_2In) and a γ (defined as C/T as $T \rightarrow 0$) of 260 mJ/mol K^2 vs 850 mJ/mol K^2 for U_2Pt_2In. The shape of this C/T minimum in U_2Co_2Sn is reminiscent of noninteracting spin fluctuation or ''paramagnon'' behavior, where $C/T \sim T^2 \log T$, which is the next term in Fermi liquid theory after $C/T \sim \gamma$. This, coupled with the behavior of the resistivity (described[6] as resembling the resistivity of the known paramagnon system UPt_3) and the fact[5] that the U-U interatomic separation $d_{U\text{-}U}$ in U_2Co_2Sn is 3.5 Å (equal to the Hill limit above which itinerant $5f$-electron behavior becomes local) where $C/T \sim T^2 \log T$ has been found in UAl_2, inspired the investigation reported here.

EXPERIMENTAL RESULTS AND DISCUSSION

Samples of U_2Co_2Sn were prepared by arc melting together the pure elements under a purified argon atmosphere. X-ray diffractometry indicated single-phase material in the tetragonal U_3Si_2 structure, which as discussed in Ref. 7 for the general ternary U_2T_2Sn has distinct atomic positions for the transition metal (Co in this case) and the Sn. The high-angle x-ray linewidths indicated good lattice order, with a full width at half maximum for the (α_1-α_2 resolved) peak at 116.6°(2Θ) of 0.37°, or less than twice instrumental resolution. Site switching of the Co and Sn would in any case not be expected, due to the disparity ($\sim$ 30%) in atomic sizes. Resistivity measurements down to 0.3 K were made at distinct temperatures using a four-wire dc measurement technique; resistivity measurements between 0.1 and 1.3 K were made using a sensitive continuous ac lock-in technique. Susceptibility vs temperature and magnetization vs field measurements at various temperatures down to 1.8 K were made using a commercial Quantum Design magnetometer. Specific heat data down to 0.3 K and in fields up to 13 T were measured using established[8] techniques.

The zero-field C/T data plotted versus temperature between 0.3 and 10 K are shown in Fig. 1, with the solid line a fit to the normal electronic and lattice specific heat plus the noninteracting spin fluctuation functional form[9] [$C \sim T^3 \log(T/T_{SF})$, where T_{SF} is the characteristic spin fluctuation temperature]. As is readily seen, the upturn in the specific heat divided by temperature is apparently not caused by noninteracting spin fluctuations, or paramagnons.[9] Since C/T as shown in Fig. 1 is more divergent at low temperatures than the fit, the data were then replotted as $\Delta C/T$ (where $\Delta C = C_{\text{measured}} - C_{\text{lattice}} = C_{\text{electronic}}$) vs $\log_{10} T$ as also shown in Fig. 1 to check— despite U_2Co_2Sn's smaller γ and shallower minimum— for the NFL behavior observed in the related compound U_2Pt_2In. Figure 1 shows that the electronic specific heat data for U_2Co_2Sn diverge less rapidly than

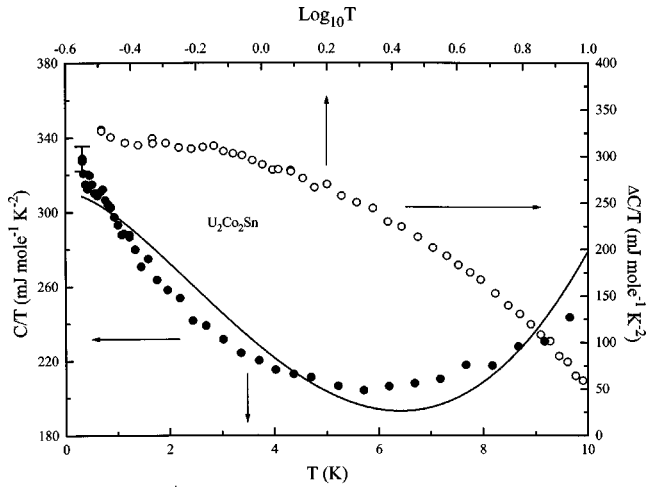

FIG. 1. Specific heat C divided by temperature T of U_2Co_2Sn between 0.3 and 10 K versus T (solid circles, lower axis). The solid line is a fit to $C=\gamma+\beta T^3+\delta T^3\log_{10}(T/T_{SF})$, where the last term in predicted (Ref. 9) for noninteracting spin fluctuations and T_{SF} is the spin fluctuation temperature. The fit uses only three parameters ($\gamma,\beta'=\beta-\delta\log_{10}(T_{SF})$, and δ); these are found to be 310 mJ/mol K^2, -13.5 mJ/mol K^4, and 13.2 mJ/mol k^4, respectively. (The fact that β' is negative is due to $\log_{10}T_{SF}$ being larger than the coefficient β of the lattice Debye specific heat contribution.) Even with a four-parameter fit (discussed later in the text), with $C_{lattice}=\beta T^3+\alpha T^5$, the fit does not convincingly reproduce the data. Note the divergence of C/T above the spin fluctuation fit below 1 K. The open circles (upper axis) show ΔC [$\equiv C_{measured}-C_{lattice}$, where $C_{lattice}=\beta T^3$, with $\beta=1.466$ mJ/mol K^4 ($\Rightarrow\theta_D=188$ K) determined from a separate measurement—not shown—of the specific heat of $UThCo_2Sn$] divided by temperature T vs $\log_{10}T$ of U_2Co_2Sn. Clearly, C/T is also not represented by $-\log_{10}T+C_{lattice}/T$ over any appreciable temperature range.

$C/T\sim-\log_{10}T$, following a monotonic-appearing convex curvature over the whole temperature range when plotted vs $\log T$.

This divergent, non-Fermi-liquid behavior in C/T that nevertheless follows $\log_{10}T$ over no apparent temperature range shown in Fig. 1 has been reported until now in no other system to our knowledge. C/T in NFL systems discovered to date has one of the following behaviors. (a) It follows $\log T$ from $\sim$ 10 K down to the lowest temperature of measurement ($\sim$ 0.1 K), such as in $CeCu_{5.9}Au_{0.1}$,[10] i.e., there is no crossover region observed from one dependence to another. (b) It follows $\log T$ down to around 0.2 K and then shows greater divergence at lower temperatures, such as in $U_{0.2}Y_{0.8}Pd_3$.[11] (c) It follows $\log T$ down to some low temperature and then exhibits less divergent behavior over the remaining temperature range of measurement. Systems that belong to this last category include[12] $CeCu_2Si_2$ in 0.7 GPa and 2 T between 0.4 and 1.2 K and $CeNi_2Ge_2$ between 0.4 and 1 K. Such results are typically plotted with C/T fitted to $1-\sqrt{T}$ in this low-temperature regime, after the NFL theories of Millis[13] and Moriya[14] for a quantum phase transition with weakly interacting spin fluctuations, although a fit to a particular temperature dependence over less than a decade in temperature is not definitive.

The monotonicity of the curvature of the C/T vs $\log T$ plot of the data for U_2Co_2Sn in Fig. 1 led us to consider that

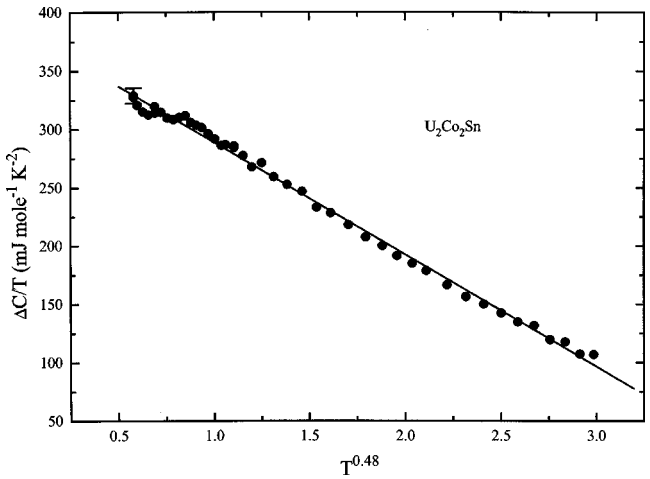

FIG. 2. $\Delta C(=C_{measured}-\beta T^3)/T$, where $\beta=1.466$ mJ/mol K^4 as discussed in Fig. 1, of U_2Co_2Sn is plotted vs T^α giving as a best fit $\Delta C/T=385-96T^\alpha$, with $\alpha=0.48\pm0.05$, where the error bar is from estimating a $\pm10\%$ uncertainty in the subtracted lattice contribution as discussed in the text. As may be readily seen, this three-parameter fit to the Millis/Moriya theory of weakly interacting spin-fluctuation-caused non-Fermi-liquid behavior passes through the data in the whole temperature range of measurement, with a much smaller standard deviation (by more than a factor of 2) than the three-parameter fit (see Fig. 1) to the noninteracting spin fluctuation theory (Ref. 9).

the data might follow such a $1-\sqrt{T}$ power law over some appreciable temperature range. Surprisingly, such a plot (see Fig. 2) results in the discovery that $\Delta C/T$ for U_2Co_2Sn obeys approximately $\gamma-A\sqrt{T}$ over the entire temperature range of 0.3 to 10 K, with only one fit parameter used for $C_{lattice}$ ($=\beta T^3$). (The standard deviation in the fit of the data to $\gamma-AT^\alpha$ for $\alpha=0.48$—the best fit—is only 0.6% smaller than for $\alpha=0.50$.) In order to make the excellence over the whole temperature range of this three-parameter fit ($C=\gamma T+\beta T^3-AT^\alpha$, $\alpha\approx0.5$) to the data even more apparent, a comparison of this fit to a four-parameter (where $C_{lattice}=\beta T^3+\alpha T^5$) noninteracting spin fluctuation fit (not shown) can be made. The four-parameter fit is still (slightly) worse than the three-parameter fit using the Millis/Moriya theory temperature dependence (either $\alpha=0.5$ or the best-fit exponent of 0.48 shown in Fig. 2). Further, the four-parameter fit is in fact unphysical in this case, since specific heat data (not shown) taken as part of this work on single-phase, U_3Si_2-structure $UThCo_2Sn$—with essentially the same mass and therefore presumably the same lattice behavior as U_2Co_2Sn—show that $C_{lattice}$ in fact follows the simple Debye law ($C=\beta T^3$) up to 10 K.

In order to allow for possible errors in the temperature dependence of $\Delta C/T$ for U_2Co_2Sn shown in Fig. 2, and to investigate how critical the exact subtraction used for $C_{lattice}$ is to the temperature range where $\Delta C/T=\gamma_0-T^\alpha$, we have allowed the magnitude of $C_{lattice}$ to vary by $\pm10\%$ and replotted (not shown) these data as in Fig. 2. The result is that the exponent varies from 0.43 (smaller lattice contribution) to 0.53 (larger lattice contribution), with only a small ($\sim$10%) change in the quality (standard deviation) of the fit to the $\Delta C/T$ data. Thus, the specific heat data reported here for U_2Co_2Sn represent the first experimental example known

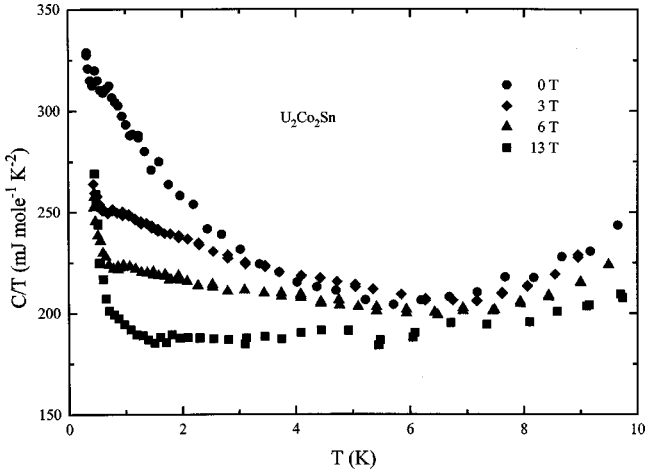

FIG. 3. Specific heat divided by temperature vs temperature of U_2Co_2Sn in magnetic fields up to 13 T. The upturns at the lowest temperatures, which become more pronounced and start at higher temperature with increasing field, are due to the nuclear moments (and accompanying level splitting) of both ^{59}Co (100% abundant) and three of the various Sn isotopes (total abundance 16.6%). The observed sizes of the upturns in C/T are consistent with the values calculated using the known nuclear moments of ^{59}Co and the Sn isotopes. The strong decrease of the specific heat with applied field (already 15% at 1 K in 3 T) is consistent with fairly weak magnetic interactions being responsible for the non-Fermi-liquid behavior.

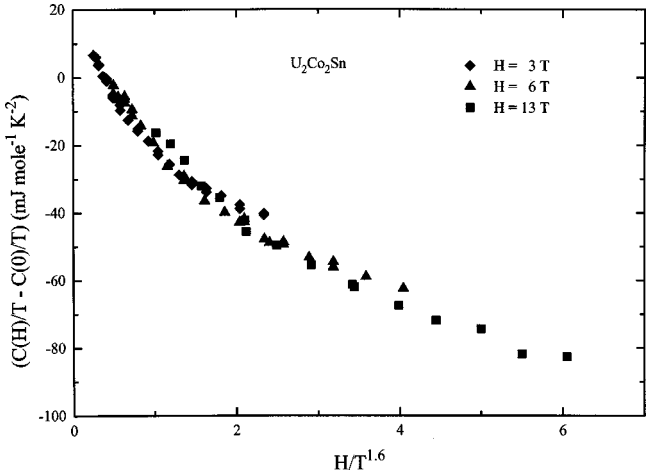

FIG. 4. The change in C with applied field H divided by temperature (data from Fig. 3) is plotted vs $H/T^{1.6}$, which brings the various curves rather well onto one universal scaled curve. (The upturns in C/T caused by nuclear Schottky level splitting seen at low temperature in Fig. 3 are excluded from this scaling.) Using a scaling exponent β of 1.4 or 1.8 gives significantly greater deviation from a common curve. A similar scaling of the magnetization vs field up to fields of 5 T and at temperatures of 2, 4, and 10 K (not shown) is complicated by an apparent small metamagnetic transition at ~2800 G. The scaling exponent that is obtained from the higher-field data is approximately 1.3, still in the regime ($\beta > 1.0$) where single-ion magnetic excitations are excluded. (Ref. 16).

to the authors of the Millis and Moriya weakly divergent, spin-fluctuation-interaction-induced non-Fermi-liquid behavior in other than a crossover, limited temperature regime. Of course, the data must be extended to lower temperature where deviations from this behavior may occur, as seen, for example, in the $C/T \sim -\log T$ behavior of $U_{0.2}Y_{0.8}Pd_3$ below 0.2 K.[11]

As a method of further characterizing these underlying weak spin fluctuations, we have measured the response of the specific heat to a magnetic field, Fig. 3. Clearly, the interactions responsible for the upturn in C/T at low temperatures are strongly affected by an applied field. As discussed by Tsvelik and Reizer,[15] calculating details of a particular model at or near a quantum critical point at $T=0$, where quantum fluctuations prevent entry into the Fermi-liquid ground state, is very difficult, whereas insight into the underlying physics may be obtained by examining the scaling behavior at finite temperatures, where a parameter (e.g., C) measured for various fields and temperatures is plotted vs H/T^{β}. Thus, Fig. 4 shows that plotting the change with field in C divided by temperature vs $H/T^{1.6}$ collapses the disparate curves from Fig. 3 onto one. The fact that $\beta > 1.0$ implies, according to Tsvelik and Reizer (see also Ref. 16 for work on $U_{0.2}Y_{0.8}Pd_3$), that the underlying magnetic interactions are of a correlated nature. Thus, single-ion models such as the multichannel Kondo model[17] or the quadrupolar Kondo model[18] are apparently ruled out. However, there is a slight (~10–15%), faster than linear increase in the magnetization as a function of field (not shown) at 0.28 T, which may be a sign of a weak metamagnetic transition. Since the specific heat and magnetization data (see Fig. 4) for the scaling are all at fields greater than 0.28 T, there could possibly be a different scaling behavior for low fields. Thus, until more work is done on scaling behavior above and below B_{metamag} (see Ref. 19 for partial work on UPt_3, where $B_{\text{metamag}} = 20$ T), this scaling result for U_2Co_2Sn should be considered as indicative but not conclusive of the nature of the correlations.

The low-temperature magnetic susceptibility of U_2Co_2Sn is shown in Fig. 5 plotted vs $\log_{10} T$, and obeys this temperature dependence between approximately 4 and 40 K, with a deviation at low temperatures. If the data are instead plotted to obtain a power law $\chi \sim \chi_0 T^{-\eta}$ (not shown), η is found to be 0.17, with the data obeying this form between the lowest temperature of measurement (1.8 K) and 5 K. Since the theories of Millis and Moriya do not address[20] the analytic temperature dependence of the magnetic susceptibility of (pre-

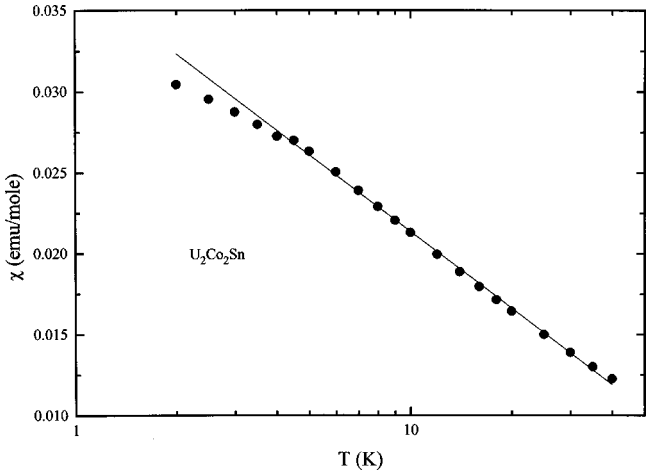

FIG. 5. Low-temperature magnetic susceptibility χ of U_2Co_2Sn plotted vs $\log_{10} T$. Below 4 K the behavior becomes less divergent.

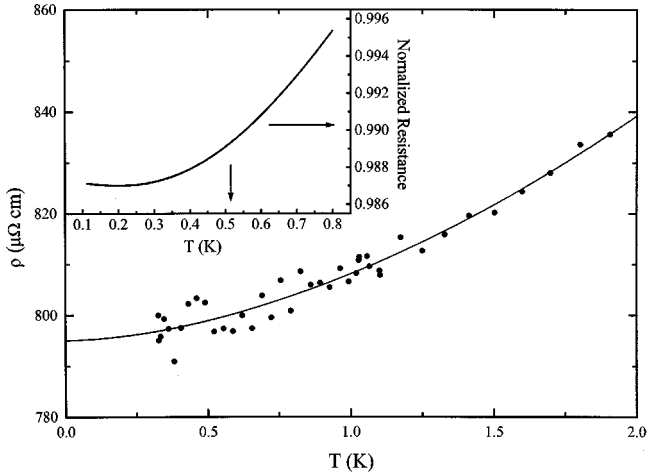

FIG. 6. The dc resistivity in absolute units of U_2Co_2Sn is plotted vs temperature as solid points in the main figure. The solid line shown through these points is a fit to $\rho=\rho_0+AT^{\alpha}$, where the best fit gives $\rho_0=795\ \mu\Omega$ cm, $A=13\ \mu\Omega\ cm/K^{\alpha}$, and $\alpha=1.76$. As discussed in the text, it has been reported (Ref. 6) that U_2Co_2Sn suffers from microcracks and that this unphysically very large (for a metal) ρ_0 is not intrinsic. In the inset, low-temperature ac resistivity, normalized to the 1 K value, of U_2Co_2Sn vs temperature was regressively fitted to a four-term polynomial to smooth the data. This representation of the data is shown as the solid line. If the data are fitted to $\rho=\rho+AT^{\alpha}$ (not shown) from 0.4 to 1.3 K, α is found to be 1.85 ± 0.05. The apparent flattening out of the curve at lowest temperatures is believed to be not an experimental artifact, e.g., heating due to too much measurement current was carefully ruled out.

sumably) three-dimensional U_2Co_2Sn, these fitted temperature dependences serve primarily for comparison to data for other known NFL systems ($\chi\sim\chi_0-A\sqrt{T}$ for $U_{0.2}Y_{0.8}Pd_3$,[21] $\chi\sim-\log T$ for $Ce_{0.1}La_{0.9}Cu_2Si_2$,[22] $\chi\sim\chi_0T^{-0.27}$ for $UCu_{3.5}Pd_{1.5}$,[23] $\chi\sim\chi_0T^{-0.13}$ for[24] $CeNi_2Ge_2$) and to other theories (the quadrupolar Kondo model[18] predicts $\chi\sim\chi_0-A\sqrt{T}$, the Griffiths phase-disorder model[25] predicts $\chi\sim\chi_0T^{\lambda-1}$). A recent experimental work,[26] inspired by the latter theory, has replotted χ data for a number of systems, including $U_{0.2}Y_{0.8}Pd_3$ where[21] good agreement to $\chi_0-A\sqrt{T}$ was found, to show agreement with power-law, $T^{\lambda-1}$, behavior. This illustrates—as do the two temperature dependences fitted to the U_2Co_2Sn χ data reported here—the ambivalence of fitting the susceptibility data over a limited temperature range, as well as the inconclusive status of current NFL theories as regards the temperature dependence of the low-temperature magnetic susceptibility. Certainly, the rather limited temperature range (1.8–5 K) where χ for U_2Co_2Sn obeys the disorder-model $\chi_0T^{\lambda-1}$ dependence [a plot of $\log(C-C_{\text{lattice}})$ vs $\log T$—not shown—obeys the disorder theory[25] only between 0.3 and 1 K] coupled with the apparent good order deduced from the high-angle x-ray linewidths argues against the disorder scenario in this undoped NFL system.

The theories of Millis and Moriya do, however, make a firm prediction for the temperature dependence of the electrical resistivity of systems whose specific heat obeys $C/T\sim\gamma-\sqrt{T}$, namely, $\rho\sim\rho_0+AT^{1.5}$. Resistivity data for U_2Co_2Sn taken by a point-by-point dc method down to 0.3 K are shown in Fig. 6, with data taken by a continuous ac method down to 0.08 K shown as an inset in Fig. 6. (The large residual resistivity ρ_0 for U_2Co_2Sn has been observed before[6] and is thought[6] to be due to a tendency for microcracks to form in this material.) As may be seen in Fig. 6, down to 0.3 K $\rho=\rho_0+AT^{1.76\pm0.1}$, i.e., the data appear to have a temperature dependence somewhere between Fermi-liquid behavior ($\rho=\rho_0+AT^2$) and the predicted[13,14] NFL temperature dependence, with the accuracy of the fitted exponent negatively affected by the large ρ_0. The lowest-temperature resistivity data, shown in the inset in Fig. 6, flatten out and go through a slight minimum at 0.2 K. A fit of these data between 0.4 and 1.3 K results in $\rho\sim\rho_0+AT^{1.85\pm0.05}$. In order to further investigate the increase in the scattering rate below 0.2 K, which causes the minimum in ρ, magnetoresistance measurements are planned.

SUMMARY AND CONCLUSIONS

This deviation from monotonic behavior of the resistivity of U_2Co_2Sn below 0.3 K is a caveat for all research into NFL behavior, where it is the lowest-temperature behavior that should theoretically be the simplest but is observed experimentally to be dominated by deviations. Such behavior has been seen, for example, in the resistivity (specific heat) of $CeNi_2Ge_2$ below 0.1 (0.3) K, where a peak (flattening out) occurs,[24] as well as in the increase above a $\log T$ divergence observed below 0.2 K in C/T in $U_{0.2}Y_{0.8}Pd_3$,[11] mentioned above. Thus, our observation of $C/T\sim\gamma-A\sqrt{T}$ between 0.3 and 10 K, coupled with the non-Fermi-liquid behavior of χ and deviation from the Fermi-liquid $\rho\sim\rho_0+AT^2$ of the resistivity, is clearly cause for identifying U_2Co_2Sn as an undoped NFL system—one of only three known at ambient pressure. Comparison with current theories gives good agreement with Millis[13]/Moriya[14] as regards the specific heat, but shows a clear deviation from their predicted temperature dependence for the resistivity. Measurements to lower temperature are of critical importance, especially for C, in order to determine the extent of the Millis/Moriya weakly interacting spin fluctuation temperature dependence and to probe the expected deviations therefrom to provide information for the needed further development of the theories. It is interesting to speculate that the large temperature range (unique among currently known NFL systems) over which $C_c\approx\gamma-A\sqrt{T}$ (consistent with weak, ''Gaussian'' interactions) in U_2Co_2Sn may be related to relatively weak U $5f$-electron hybridization with the d electrons present. This hybridization need not be especially strong in U_2Co_2Sn to prevent $5f$ localized behavior, since $d_{\text{U-U}}$ is exactly at the Hill limit of 3.5 Å, whereas—due to the significantly larger[27] $d_{\text{U-U}}$ in the other known U NFL systems (3.58 Å for U_2Pt_2In, 4.07 Å for $U_{0.2}Y_{0.8}Pd_3$, 4.09 Å for $U_{0.1}Th_{0.9}Cu_2Si_2$, 4.19 Å for $U_{1-x}Th_xPd_2Al_3$, and 4.99 Å for $UCu_{5-x}Pd_x$)—the other known U systems definitely have strong $5f$ hybridization or localized (magnetic) behavior would be observed.

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