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Non-Fermi liquid behavior in $U_{3-x}Ni_3Sn_{4-y}$ single crystals

L. Shlyk[a,b], J.C. Waerenborgh[c], P. Estrela[d], A. deVisser[d], L.E. DeLong[b,*], A.M. Gurevich[a], M. Almeida[c]

[a]*Institute of Low Temperature Physics and Engineering, Kharkov, Ukraine*
[b]*Department. Physics and Astronomy, University of Kentucky, 177 Chem. Phys. Building, Lexington, KY 40506-0055, USA*
[c]*Department of Quimica, Instituto Tecnologico e Nuclear, Sacavem, Portugal*
[d]*Van der Waals-Zeeman Institute, Amsterdam, The Netherlands*

Abstract

$U_3Ni_3Sn_4$ and $U_{2.9}Ni_{3.0}Sn_{3.9}$ single crystals exhibit a non-Fermi liquid susceptibility $\chi \propto T^{-0.3}$ below 10 K. The electronic heat capacity coefficient $\gamma(T)$ of $U_{2.9}Ni_{3.0}Sn_{3.9}$ varies as the square root of temperature between 0.3 and 5 K, consistent with fluctuations near an antiferromagnetic quantum critical point.

Keywords: Heavy fermions; Non-Fermi liquid; Uranium compounds

A number of paramagnetic 4f- and 5f-electron systems are known to exhibit an extended "non-Fermi-liquid" (NFL) regime in which critical fluctuations induce non-analytic temperature dependences of physical properties at $T < T_0$. T_0 is a characteristic temperature [1] that depends upon the degree of f-ligand hybridization, which depends on the cation occupancy in $U_3M_3X_4$ compounds (M-transition metal, X-metalloid) with the cubic $Y_3Au_3Sb_4$ structure (filled Th_3P_4-type) [2,3]. Therefore, we have investigated two single-crystal $U_3Ni_3Sn_4$ samples of slightly different composition, grown via slow-cooling in a levitated cold crucible using an induction furnace [4].

X-ray refinements were consistent with the $Y_3Au_3Sb_4$-type structure and estimated compositions, $U_{2.9}Ni_{3.0}Sn_{2.9}$ and $U_3Ni_3Sn_{\delta}$, having unit-cell parameters 9.3577(4) or 9.3524(5) Å, hereafter referred to as Sample 1 and Sample 2, respectively. The temperature dependences of the magnetic susceptibilities of Samples 1 and 2 follow a $T^{-0.3}$ dependence in the range 1.7–5 K (Fig. 1, lower inset). Above 25 K, χ^{-1} versus T data approximately obey a modified Curie–Weiss behavior with Curie–Weiss temperature $\theta_p \approx -50\,(-60)$ K, effective moment $\mu_{eff} \approx 2(1.8)\mu_B$/U, and $\chi_0 \approx 1.1(0.95) \times 10^{-3}$ emu/mol U for Sample 1 (2), in agreement with polycrystal results [3].

The heat capacity of Sample 1 was measured in a ^{3}He cryostat using the relaxation-time method with results similar to polycrystal data [3] above 1.5 K (see Fig. 2); but lower temperature single-crystal data reveal a rapid increase in C/T between 0.3–0.7 K. The heat capacity of Sample 1 can be fit (see Fig. 2) with the following expression:

$$C = (\gamma_0 - \alpha\sqrt{T})T + \beta T^3 + D/T^2, \quad (1)$$

where $C_E = (\gamma_0 - \alpha\sqrt{T})T$ is the electronic contribution, $C_L = \beta T^3$ is the lattice contribution, and $C_N = D/T^2$ represents the high-temperature form of a nuclear Schottky term [5–8]. The best-fit coefficients are $\gamma_0 = 0.124$ J/mol U–K^2, $\alpha = 0.0151$ J/mol U–K$^{2.5}$, $\beta = 2.071 \times 10^{-3}$ J/mol U–K^4, $D = 4.622 \times 10^{-4}$ J–K/mol U.

Renormalization group theory [9,17] predicts $\gamma \propto \gamma_0 - \alpha\sqrt{T}$ near a zero-temperature antiferromagnetic instability. The fitted value of $\alpha = (15/64)k_B N_A N [2/\pi T_0]^{3/2}\zeta(5/2)$ allows us to estimate the characteristic

*Corresponding author. Fax: 1-606-323-2846; e-mail: ldelong@pop.uky.edu.

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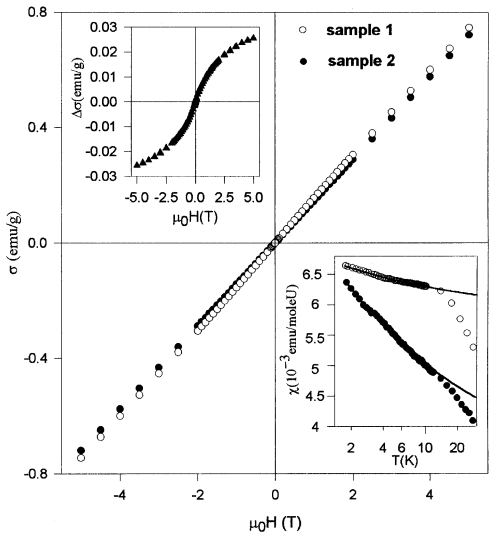

Fig. 1. Magnetization $\sigma(H)$ versus magnetic field H at $T = 1.8$ K for $U_{2.9}Ni_{3.0}Sn_{3.9}$ (Sample 1) and $U_3Ni_3Sn_4$ (Sample 2). Upper inset: Difference in magnetizations $\Delta\sigma$ of Samples 1 and 2 versus H. Lower inset: Low temperature susceptibilities; the lines are fits to $\chi(T) \propto T^{-0.3}$.

temperature $T_0 \approx (10\text{ K}) \times N^{2/3}$, assuming a N-dimensional bosonic order parameter, corresponding (e.g., $N = 1$) well with the onset temperature of the non-analytic behavior of $\chi(T)$ shown in the lower inset of Fig. 1. Alternatively, self-consistent renormalized spin fluctuation (SCR) theory [10] also predicts a square-root form of the electronic heat capacity at low temperatures that evolves into a logarithmic behavior, $C/T \sim \ln(T_0/T)$, and a good fit of the data is obtained using

$$C = \gamma_0 T \ln(T_0/T) + \beta T^3 + D/T^2, \quad (2)$$

with $\gamma_0 = 7.74 \times 10^{-3}$ J/mol U–K^2, $T_0 = 1.34 \times 10^6$ K, $\beta = 1.8 \times 10^{-3}$ J/mol U–K^4, and $D = 3.79 \times 10^{-4}$ J–K/mol U. Multi-channel Kondo [11] or Kondo disorder [12] models also predict a logarithmic divergence for $\gamma(T)$. The Kondo disorder model demands $\chi(T) \propto -\ln T$; whereas, the multichannel Kondo model can yield $\chi(T) \propto -T^{0.5}$ or $-\ln T$ at low temperatures. Fits to a $T^{-0.3}$ dependence yield slightly better results than the $-T^{0.5}$ or $-\ln T$ forms; however, the best-fit value of T_0 is extremely high, and probably does not correspond to a physically significant spin fluctuation energy. On the other hand, the $T \to 0$ behavior of $\gamma(T)$ is also predicted to be of square-root form as a consequence of a zero-temperature quantum transition from a paramagnetic metal to a spin-glass [13,14], but the predicted [13] low temperature susceptibility $\chi \propto -T^{3/4}$, in conflict with our data.

The assumed slight increase in defect concentration of Sample 1 evidently leads to a small non-linear field dependence to the difference in magnetization $\Delta\sigma$ of the two samples (see upper inset to Fig. 1), which begins to saturate above $H_{sat} = 6$ T. This is consistent with a characteristic temperature $T_0 \approx \mu_B H_{sat}/k_B \approx 4$ K, and agrees with $\chi(T)$ and $C(T)$ data.

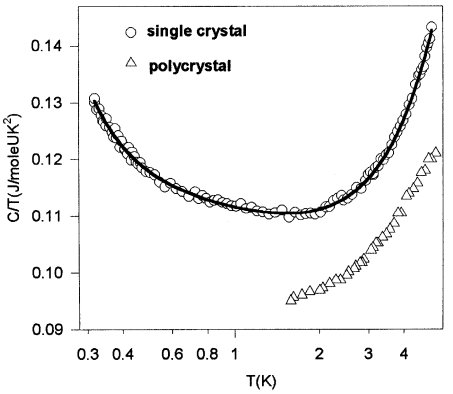

Fig. 2. (a) Heat capacity C divided by temperature T for $U_{2.9}Ni_{3.0}Sn_{3.9}$ (Sample 1) and polycrystalline sample data [3], versus $\log(T)$. The solid line is a fit of the data to Eq. (1).

A logarithmic divergence of γ has been observed for the majority of uranium NFL compounds [1]; our results identify $U_3Ni_3Sn_4$ as the first U material found to display the novel square-root dependence [6]. The temperature dependence of the susceptibilty and unrealistically high values of T_0 derived from logarithmic fits to the heat capacity appear to rule out the applicability of known theories to $U_3Ni_3Sn_4$, except the model of Millis [11] (which gives no predictions of $\chi(T)$).

Very recent experimental [15] and theoretical [16] work proposes NFL behavior is caused by competition between RKKY and Kondo interactions in the presence of atomic disorder, leading to a Griffiths phase (large magnetic clusters) close to a quantum critical point. This picture requires $C/T \propto \chi(T) \propto T^{-1+\lambda}$, which is consistent with our data for $U_3Ni_3Sn_4$ with $\lambda \approx 0.5$, and suggests the low-temperature Schottky term of the heat capacity may be due to quantum tunneling of antiferromagnetic clusters.

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References

[1] M.B. Maple et al., J. Phys.: Condens. Matter 8 (1996) 9773.
[2] T. Endstra et al., J. Magn. Magn. Mat. 89 (1990) L273.
[3] T. Takabatake et al., J. Soc. Japan 59 (1990) L16.
[4] L. Shlyk et al, Proc. 26ièmes Journées des Actinides, Szklarska Poreba, Poland, 1996, 214.

[5] O.V. Lounasmaa, in: A.J. Freeman, R.B. Frankel (Eds.), Hyperfine Interactions, Academic Press, New York, 1967.
[6] F. Steglich et al., J. Phys.: Condens. Matter 8 (1996) 9909.
[7] B. Chevalier et al., Proc. 26ièmes Journées des Actinides, Szklarska Poreba, Poland, 1996, p. 66.
[8] M.A. Polikarpov et al., Physica B 199/200 (1994) 45.
[9] A.J. Millis, Phys. Rev. B 48 (1993) 7183.
[10] T. Moriya, T. Takimoto, J. Phys. Soc. Japan 64 (1995) 960.
[11] D.L. Cox, M. Jarrell, J. Phys.: Condens. Matter 8 (1996) 9825.
[12] E. Miranda et al., J. Phys.: Condens. Matter 8 (1996) 9871.
[13] A.M. Sengupta, A. Georges, Phys. Rev. B 52 (1995) 10295.
[14] S. Sachdev et al., Phys. Rev. B 52 (1995) 10286.
[15] M.C. de Andrade et al., cond-mat/9802081, unpublished.
[16] A.H. Castro Neto, G. Castilla, B.A. Jones, cond-mat/9710123, unpublished.
[17] A.J. Millis, Phys. Rev. B 51 (1995) 8996.