



Magneto-resistance measurements in $UCu_{4+x}Al_{8-x}$ compounds

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Abstract

The intermetallic $UCu_{4+x}Al_{8-x}$ compounds ($0.1 \leq x \leq 1.95$) have remarkable electronic and magnetic properties. The Cu-poor compositions within this x -range are antiferromagnetic (AF). The Néel temperature (T_N) drops gradually from ≈ 36 K for $x = 0.1$ until AF is completely suppressed near $x = 1.1$, while the Sommerfeld coefficient (γ) increases dramatically with x , reaching a maximum value of 750 mJ/mol K^2 for $x = 1.75$. Reported in this investigation are magneto-resistance (MR) and magnetic susceptibility (χ) measurements at low temperatures (T) and high magnetic fields (B). The isothermal MR data below 40 K crosses over from positive in the low- x to negative in the high- x range. These results are interpreted in the context of spin fluctuations, hybridization, and non-magnetic atomic disorder. © 2001 Elsevier Science B.V. All rights reserved.

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The $UCu_{4+x}Al_{8-x}$ compounds crystallize in the tetragonal $ThMn_{12}$ -type structure for $0.1 \leq x \leq 1.95$ [1] and their electronic and magnetic properties depend very strongly on the Cu/Al content ratio. The Cu-poor compounds within this x -range are antiferromagnetic (AF) with reported T_N near 35 K. However, the substitution of Cu for Al is accompanied by (1) a rapid suppression of AF in the $0.1 < x < 1.15$ range; and (2) an enormous increase γ from about 100 mJ/mol K^2 ($x \approx 0.25$) to 750 mJ/mol K^2 ($x = 1.75$) [2,3]. Neutron diffraction (ND) [3] and NMR [4] results confirm the suppression of AF near $x = 1.15$. ND also indicates that the substitution of Cu for Al favors one of two inequivalent Al sites, and that the U-ordered moments lie along the tetragonal c -axis. Since the ionic size of Cu is smaller than Al, it has

been suggested that the suppression of AF when x increases is due to an enhancement of c - f hybridization, favoring the intra-site Kondo over the inter-site RKKY groundstate [1]. The electrical resistivity (ρ) vs. T data for the Cu-rich compounds [1] as well as the MR [5] are consistent with the Kondo picture. However, an interpretation in terms of hybridization alone is oversimplified, and changes in the Fermi level need to be considered [2]. For example, the crossover from heavy fermion (HF) behavior to AF in the $CeCu_{5-x}Al_x$ system occurs for increasing Cu concentration [6], contrary to the $UCu_{4+x}Al_{8-x}$ system. It has been pointed out by Gschneidner et al. that false indications of HF behavior can occur as a result of non-magnetic atomic disorder, as for example in the Cu-Al compounds $CeCu_{6.5}Al_{6.5}$ (NaZn₁₃-type structure), $CeCu_4Al$, and $CeCu_3Al_2$ [7]. The origin of the false HF behavior is the formation of low-lying crystal field (CF) levels due to disorder, giving rise to a large Schottky anomaly contribution [7]. In order to probe further the effect of the substitution of Cu

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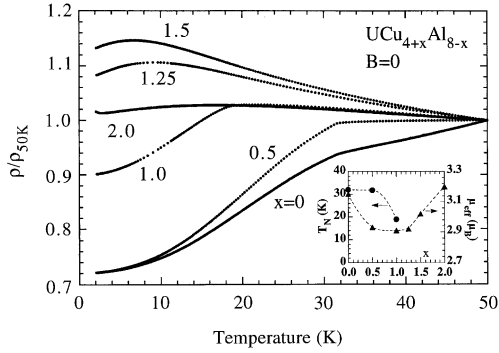


Fig. 1. Normalized electrical resistivity ρ/ρ_{50K} in $B = 0$ for $UCu_{4+x}Al_{8-x}$ compounds. The inset shows T_N vs. x and μ_{eff} vs. x . The lines are guides to the eye.

for Al in $UCu_{4+x}Al_{8-x}$, we performed a series of measurements of χ vs. T and MR in B up to 18 T.

A series of six polycrystalline $UCu_{4+x}Al_{8-x}$ samples for this study with $0 \leq x \leq 2.0$ was synthesized by arc melting, followed by annealing at 800 °C for 5 days. The phase purity of the compounds with $0.5 \leq x \leq 1.5$ was confirmed by X-ray diffraction. The $x = 0$ and 2.0 compositions showed traces of extra phases. Measurements of χ vs. T , and M vs. B up to $B = 7$ T were performed with a SQUID magnetometer. Transverse MR measurements to 7 T (2–40 K) were performed at USP, and to 18 T (2–15 K) at the NHMFL, Los Alamos Facility. The ρ vs. T data of Fig. 1 for $0 \leq x \leq 1.0$ show noticeable discontinuities at T_N , which are probably due to the suppression of spin-disorder scattering. The composition dependence of T_N extracted from the ρ vs. T data is shown in the inset of Fig. 1. These values of T_N are close to the values obtained from the χ vs. T measurements. The χ vs. T data (not shown) yields the high T effective moments μ_{eff} shown in the inset of Fig. 1. The value of μ_{eff} for $x = 0.5$ –1.25 is about $2.9 \mu_B$, increasing slightly to about $3.0 \mu_B$ for $x = 1.5$. These values are somewhat reduced from the 3.58 or $3.62 \mu_B$ values expected for U^{4+} and U^{3+} , respectively, which suggests that the $5f$ orbitals are hybridized to a certain extent. The behavior of the transverse MR vs. B at $T = 2.1$ K for the six samples of this study is displayed in Fig. 2. The MR of the multiphase compound UCu_4Al_8 is positive. However, as more Cu is substituted for Al forming single-phase materials, the MR becomes negative, and its magnitude grows from near zero for $x = 0.5$ to a maximum near $x = 1.5$, diminishing again for multiphase $x = 2.0$. The MR for the $0.5 \leq x \leq 2.0$ compositions follows closely a B^2 dependence. The behavior of $\Delta\rho/\rho$ vs. B at higher T is qualitatively very similar, albeit reduced in magnitude. The resistivity in strongly correlated systems at low T can usually be described by $\rho(B, T) = \rho_0(B) +$

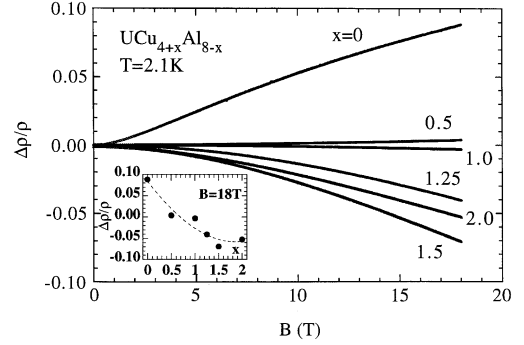


Fig. 2. Transverse magnetoresistance $\Delta\rho/\rho$ vs. B for $UCu_{4+x}Al_{8-x}$ compounds at 2.1 K. The inset shows $\Delta\rho/\rho$ vs. x at $B = 18$ T. The line is a guide to the eye.

$\rho_{\text{ep}} + A(B)T^2$, where $\rho_0(B)$ is the residual ρ , including the effect of imperfections and spin fluctuations; ρ_{ep} is due to the electron–phonon scattering; and the T^2 term includes both the interband electron–electron scattering, and spin fluctuations [8]. The MR can then be described by

$$\Delta\rho(B, T) = \Delta\rho_c(B, T) + \Delta\rho_{\text{sf}}(B, T), \quad (1)$$

where the first term is a positive contribution due to the cyclotron motion of the charge carriers, and the second term is a negative contribution due to the suppression of spin fluctuations with B . The $\Delta\rho/\rho$ vs. B data of Fig. 2 suggest that the cyclotron contribution from the impurity phase is larger than the spin-fluctuation term in UCu_4Al_8 . Upon the substitution of more Cu for Al the compounds become single phase, and the MR becomes progressively more negative, suggesting that spin fluctuation scattering becomes more relevant when T_N is reduced or suppressed.

In conclusion, the MR data on $UCu_{4+x}Al_{8-x}$ both below T_N as well as in the non-ordered compositions is negative due to the suppression of spin fluctuations. The negative value of $\Delta\rho/\rho$, the quadratic behavior with B , and the drop in magnitude with T are consistent with the spin fluctuation theory developed by Ueda [9]. The μ_{eff} values extracted from the χ vs. T data is reduced from the full U moment as expected in hybridized materials. However, μ_{eff} does not depend very much on x . The important question of whether these materials are real or false HF still remains. Although no CF excitations could be detected with neutron scattering (NS) in the high γ compositions [10], this does not preclude false HF behavior, considering the difficulty in detecting CF excitations in U compounds using NS [11]. Although the study of the properties of this series remains a challenging problem, a better understanding of the effect of non-magnetic disorder on the band scheme, and the CF excitations are in order.

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