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<th>MAGNETORESISTANCE MEASUREMENT IN URANIUM COPPER (4+X)ALUMINUM(8-X)</th>
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Magnetoresistance Measurements in UCu$_{4+x}$Al$_{8-x}$ Compounds

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The intermetallic UCu$_{4+x}$Al$_{8-x}$ compounds (0.1 ≤ x ≤ 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/mole·K$^2$ for x = 1.75. Reported in this investigation are magnetoresistance (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.

Keywords: UCu$_{4+x}$Al$_{8-x}$; Magnetoresistance; Antiferromagnetism; Heavy-fermion.
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 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 $\gamma$ from about 100 mJ/mole-K ($x = 0.25$) to 750 mJ/mole-K ($x = 1.75$).[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's 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 ($\rho$) versus $T$ 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$_{6.5}$ (NaZn$_{13}$-type structure), CeCu$_4$Al, and CeCu$_3$Al$_2$.[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 for Al in UCu$_{4+x}$Al$_{8-x}$, we performed a series of measurements of $\chi$ vs $T$ and MR in $B$ up to 18 T.

A series of 6 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 $\chi$ 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 \( \rho \) 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 \( \rho \) vs \( T \) data is shown in the inset of Fig. 1. These values of \( T_N \) are close to the values obtained from the \( \chi \) vs \( T \) measurements. The \( \chi \) vs \( T \) data (not shown) yields the high \( T \) effective moments \( \mu_{\text{eff}} \) shown in the inset of Fig. 1. The value of \( \mu_{\text{eff}} \) 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 6 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 \( \rho(B,T) = \rho_0(B) + \rho_{\text{ep}} + \lambda(B)T^2 \), where \( \rho_0(B) \) is the residual \( \rho \), including the effect of imperfections and spin fluctuations; \( \rho_{\text{ep}} \) 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

\[
\Delta \rho(B,T) = \Delta \rho_c(B,T) + \Delta \rho_{\text{sf}}(B,T)
\]

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 $\text{UCu}_{4+x}\text{Al}_{8-x}$ 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 $\mu_{\text{eff}}$ values extracted from the $\chi$ vs $T$ data is reduced from the full $U$ moment as expected in hybridized materials. However, $\mu_{\text{eff}}$ does not depend very much with $x$. 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 $\gamma$ 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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References


Figure Captions

Fig. 1 - Normalized electrical resistivity $\rho/\rho_{50K}$ in $B = 0$ for UCu$_{4+x}$Al$_{8-x}$ compounds. The inset shows $T_N$ vs $x$ and $\mu_{eff}$ vs $x$. The lines are guides to the eye.

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.
Figure 1 - M. S. Torikachvili et al. - 1P-26

Graph showing the temperature dependence of resistivity for different compositions of UCu$_{4+x}$Al$_{8-x}$ with $B=0$. The x-axis represents temperature in Kelvin (K), and the y-axis represents resistivity normalized to $\rho_{50K}$. The inset graph illustrates the variation of $T_N$ (K) with $x$. The main graph includes several curves for different compositions, indicating changes in resistivity with varying temperatures.
Figure 2 - M. S. Torikachvili et al. - 1P-26

\[ \Delta \rho / \rho \]

\[ \text{UCu}_{4+x} \text{Al}_{8-x} \]

\[ T=2.1 \text{K} \]