

Non-Fermi Liquid Behavior in the $U_{1-x}Lu_xCu_5$ System

D. J. Burnette, J. S. Kim, T. Scott, and G. R. Stewart

University of Florida
Department of Physics
P.O. Box 118440
Gainesville, FL 32611 – 8840

(July 27, 2006)

Abstract

Possible non-Fermi liquid behavior was sought using Lu doping on the U site in UCu_5 . The cubic $AuBe_5$ crystal structure of the compounds was verified, the magnetic susceptibility and electrical resistivity were measured and curve fitting was performed. The electrical resistivity was fit to $\rho = \rho_0 - AT$ over a temperature range of 0.34 K to 18.5 K, while magnetic susceptibility was fit to the curve $\chi^{-1} = \chi_0^{-1} + AT^\alpha$ for $\alpha=0.16$. To determine where $T_N = 0$, magnetic susceptibility was measured as a function of temperature, from 1.8-40K for samples of $U_{1-x}Lu_xCu_5$ with $x= 0.00, 0.10, 0.30, 0.35, 0.50, 0.80, 0.90, \text{ and } 1.00$.

I. INTRODUCTION

In the mid to late 1950's, LD Landau predicted that the behavior of metallic compounds could be described by a gas of Fermi 'particles', or a collection of non-interacting electrons whose masses had been renormalized. He claimed that the particle-particle interactions were temperature independent at low temperatures and thus could be treated easily via this renormalization procedure [1]. In his theory, Landau determined that the Fermi particles and hole excitations carry the same quantum numbers as their electron counter-parts in the non-interacting Fermi gas, while the wavefunctions and energies of the particles are different [1]. The particles are thus referred to as electron-like "quasiparticles" formed from the electron gas. Despite recent failures that motivate the subject of non-Fermi liquids, Landau's Fermi-liquid theory has been remarkably successful at describing a wide range of metals including some, like UPt_3 , where the interactions between electrons have been known to play a significant role in determining the metal's measurable properties at low temperatures.

The Fermi-liquid model predicts certain temperature dependences in the low-temperature behavior of various physical properties [2]. For example, the specific heat C , divided by the temperature T , approaches a constant (called the Sommerfeld coefficient γ) as T tends to 0. The low-temperature magnetic susceptibility χ also remains temperature-independent, while the electrical resistivity ρ goes as $\rho(0) + AT^2$.

In 1991, Seaman *et al.* discovered that the particle-particle interactions in $Y_{1-x}U_xPd_3$ were too strong and long-ranged to obey the model predicted by Fermi-liquid theory [3], i.e., the interactions remained temperature *dependent* as $T \rightarrow 0$, causing a violation of the simple Fermi-liquid temperature dependences. This discovery has led to a

new field in condensed matter physics: non-Fermi liquids. Since this initial discovery, a variety of theoretical explanations for this non-Fermi liquid behavior have been suggested, but no single theory has thus far been successful at describing the behavior of all non-Fermi liquid (nFl) systems. In fact, nFl behavior is typically discovered experimentally by measuring the low-temperature properties of the system and then comparing them to known non-Fermi liquid systems.

One situation where non-Fermi liquid behavior often occurs is in metals close to a quantum critical point [1]. A quantum critical point (QCP) occurs when a second-order phase transition takes place at $T=0$, and the quasiparticles are subject to *quantum* (rather than classical) fluctuations and cease to behave in the way that Fermi-liquid theory would predict. It is often the case that a critical point (QCP) can be brought about in a magnetic system by variation of pressure, applied magnetic field or composition (doping) to suppress magnetic ordering temperatures to zero [4].

$\text{UCu}_{5-x}\text{Pd}_x$ was the first non-Fermi liquid system discovered without doping on the active f-atom site [5]. The nFl behavior was reported for doping Pd on the Cu sites, in UCu_5 , an antiferromagnet with Néel (ordering) temperature $T_N = 16$ K, and forms in the cubic AuBe_5 structure. The results for UCu_4Pd show that $\rho = \rho_0 - AT$ between 0.3 and 10K, a major deviation from the predicted $\rho_0 + AT^2$. Additionally, it has been shown that UCu_4Ni exhibits the same nFl $\rho = \rho_0 - AT$ between 2 K and 15K [6].

Motivated by these previous results, the present work involved a search for possible nFl behavior using Lu doping on the U site in UCu_5 . The cubic AuBe_5 crystal structure of the compounds was verified and the magnetic susceptibility was measured. The primary goals of the research were to determine the concentration at which $T_N = 0$, to

study the low temperature electronic properties at that concentration, and then to use those results to predict the behavior of similar systems.

To determine where $T_N = 0$, we measured magnetic susceptibility as a function of temperature, from 1.8-40K for samples of $U_{1-x}Lu_xCu_5$ with $x = 0.00, 0.10, 0.30, 0.35, 0.50, 0.80, 0.90,$ and 1.00. These measurements were performed using a Superconducting Quantum Interference Device (SQUID). In addition, we measured the electrical resistivity from 0.3-18.5 K. We then attempted to fit the data to various equations known to describe non-Fermi liquid behavior.

II. PREPARATION & EXPERIMENTATION

Sample Preparation

Before experimentation can begin, high quality samples had to be prepared. First, high purity elements were weighed out to the desired stoichiometric proportions to a precision of 0.1 milligrams. The target sample size ranged from 250 mg to 500 mg. It should be noted that uranium is a radioactive element and therefore **strict** safety guidelines were followed at all times. After weighing out the pieces, they were placed into an Edmund Bühler arc-melter where they were melted together to form a single bead. An arc-melter consists of a sealed chamber with an electrode shaft with a tungsten tip extending down the center. At the bottom, the elements sit on a pure-copper hearth just centimeters below the tungsten tip of the electrode. The hearth is water cooled from below to assure that the hearth does not melt along with the sample. Samples are melted in an ultra high purity argon environment, to ensure that no oxygen is present.

Successfully melting samples in an arc-melter is more of an art than merely routine. Factors such as each element's melting point and vapor pressure at various temperatures must be considered when preparing to melt. As a result, the approach to every sample must be treated differently. In the case of $U_{1-x}Lu_xCu_5$, this is especially important because of the large amounts of Cu needed for each sample. In general, Cu is difficult to work with because it sits on the Cu hearth, making the sample Cu and hearth practically indistinguishable. The samples used in our experiment were therefore melted very carefully on low heat. To ensure homogeneity, we melted the samples at least five times, flipping the sample after each melt. Specifically for the $U_{1-x}Lu_xCu_5$ system, samples with $x = 0.00, 0.10, 0.30, 0.35, 0.50, 0.80, 0.90,$ and 1.00 were prepared.

X-Ray Diffraction

Upon melting a sample, unwanted second phase compounds may form due to impurities, thermodynamics, or calculation errors. To verify that no second phase has formed in the sample, a simple x-ray diffraction pattern can be analyzed. To perform these measurements, we used the x-ray diffractometer (XRD) at the University of Florida's Major Analytical Instrumentation Center (MAIC).

Samples had to be prepared on a diffraction slide before XRD could be performed. The sample was first gently broken up into smaller pieces. Then approximately 100-150 mg of those pieces were ground up further into a fine powder and then glued onto the slide in an approximately 1 cm x 1cm square. The glue used for this process has a mixture of one part amyl acetate and seven parts collodion. This particular

mixture was used because it does not contribute significantly to the x-ray pattern due to the low scattering power of the atoms making up the organics.

The physics underlying XRD is straightforward. High-energy photons with wavelength 1.54056 \AA barrage the sample and some are diffracted by the crystal structure of the sample. These diffracted photons are then counted by a collector at various angles, and at certain incident angles, large intensities of x-rays are diffracted. Using these data, combined with Bragg's Law,

$$n\lambda = 2d \sin \theta \quad (1)$$

where λ is the photon wavelength, d is the distance between atoms in the lattice, and θ is the angle of reflection of the photons, we can identify the structure of the sample. UCu_5 , which forms in the AuBe_5 cubic structure, has only one lattice parameter, a , which can be determined by

$$\frac{1}{d^2} = \frac{(h^2 + k^2 + l^2)}{a^2} \quad (2)$$

where d is the distance between atomic planes and h , k , and l , are the Miller Indices.

Magnetic Susceptibility

In order to track the antiferromagnetic transition in $\text{U}_{1-x}\text{Lu}_x\text{Cu}_5$, magnetic susceptibility (χ) was measured. Magnetic susceptibility is defined as the magnetization of a material in response to an applied magnetic field at a fixed temperature and pressure, or

$$\chi_{ij} = \frac{\partial M_j}{\partial H_i} \quad (3)$$

Here i and j refer to the directions (e.g., x , y and z in Cartesian coordinates) of the applied field and magnetization, respectively. The tensor is thus rank 2, dimension (3,3) describing the response of the magnetization in the j -th direction from an incremental change in the i -th direction of the applied field. This measurement was performed using a Superconducting Quantum Interference Device (SQUID). We measured magnetic susceptibility as a function of temperature with an applied external magnetic field. The measurements were performed in a field of 1000 G over a temperature range of 1.8-40 K. Measured values were normalized per U mole, thus the reported units for χ are emu per mole.

Resistivity

Electrical resistivity (ρ) is a very important property that can be used to classify a compound as to whether it is a non-Fermi liquid. The process for measuring resistivity at low temperatures was complicated, but it starts by cutting portions of the sample into small and precisely shaped bars. We began by placing the sample onto a dressing stick using Duco cement; then the stick was mounted onto a low-speed saw equipped with a thin diamond coated blade, where the sample was cut into a series of slabs approximately 0.020 cm in thickness. The slabs were removed by boiling the stick in a beaker of acetone, which dissolved the cement. The slabs were carefully inspected under a microscope to ensure no cracks were present, as even a small crack in the resistivity bar would have had a significant influence on the measured resistivity. The slabs were then repositioned onto a new dressing stick in such a way that thin bars could be cut out

around any defects. The process with the saw was repeated, resulting in fairly precisely shaped bars approximately 0.020 x 0.020 x 1 cm in size.

Though achieving temperatures down to 0.3 K is a complicated process, the physics used to measure resistivity is as elementary as Ohm's Law,

$$V = IR \quad (4)$$

where V is the voltage, I is the current, and R is the resistance. One can simply apply a current through the ρ -bar, measure the voltage generated, and then calculate the resistance. While resistance is fundamentally an electronic property, resistivity is a function of geometry as well. The equation for resistivity,

$$\rho = R \frac{A}{l} \quad (5)$$

where A is the cross sectional area, and l is the length of the ρ -bar, shows that ρ is directly proportional to R .

After a ρ -bar was successfully cut, two thin platinum wires were welded onto each end of the bar using a spot welder. The bar was then placed onto a clean piece of silicon and glued down by electrically insulating GE 7031 varnish. Finally, the platinum wires are looped and attached to the silicon piece using highly conductive silver epoxy making sure that it was not possible for any of the wires to accidentally touch, as this would have caused a short circuit and ruined the measurement. The silicon platform is then placed on a microscope slide and heated at 120 °C for approximately half an hour.

Once the varnish and silver epoxy had had time to set, the sample was ready to be mounted onto the final probe, which was done using special insulating thermal grease. Finally, the loose ends of the platinum wires were connected to the probe, and the sample

was ready for measurement. In our experiment, resistivity was measured for $U_{.5}Lu_{.5}Cu_5$ from 0.3-18.5 K.

III. RESULTS

Upon adding Lu to UCu_5 , the cubic lattice parameter of the samples decreased linearly (Vegard's Law) with increasing Lu concentration (see Fig. 1). This verifies that our samples formed in the correct $AuBe_5$ cubic structure. By comparing the x-ray diffraction patterns of various Lu concentrations, we see that there are no second phase lines, which serves to strengthen our argument that our samples were single phase, and the correct structure was obtained.

Magnetic susceptibility vs. temperature is plotted for four Lu concentrations. In Fig. 2, T_N drops from 11K for $x = 0.10$ to 8.5 K for $x = 0.30$. The $x = 0.50$ sample shows no feature indicative of magnetic ordering. A blown-up graph (Fig. 3) shows that $U_{0.65}Lu_{0.35}Cu_5$ appears to exhibit paramagnetic behavior with the exception of a slight kink around 5 K, which could represent a trace of T_N despite the presence of paramagnetic behavior below 5K. According to Ref. 6, the antiferromagnetic ordering temperature can, in some cases, be verified by a *plateau* in the graph of C/T . Systems in which this plateau behavior of C/T is observed at T_N include $CeCu_{6-x}Ag_x$ [7], $CeCu_{6-x}Au_x$ [8], $UCu_{5-x}Pd_x$ [9], and $UCu_{5-x}Ni_x$ [6]. Future work should consist of measuring C/T for $U_{0.65}Lu_{0.35}Cu_5$ to determine whether or not this characteristic plateau is present.

We were able to fit the magnetic susceptibility $U_{0.65}Lu_{0.35}Cu_5$ to an equation of the form,

$$\chi^{-1} = \chi_0^{-1} + AT^a \quad (6)$$

over the full measurement temperature range of 1.8 K to 40 K (Fig. 4) with $\chi^{-1} = 0.14 \text{ mol} \cdot \text{memu}^{-1}$, $A = 0.15 \text{ mol} \cdot \text{memu}^{-1} \cdot \text{K}^{-0.16}$, and $\alpha = 0.16$. This equation has previously been used to describe many non-Fermi liquids [2], thus there is evidence of non-Fermi liquid behavior in $\text{U}_{0.65}\text{Lu}_{0.35}\text{Cu}_5$.

The electrical resistivity was measured for $\text{U}_{0.5}\text{Lu}_{0.5}\text{Cu}_5$ over a range of 0.34 K to 18.5 K. We anticipated that the plot of resistivity vs. temperature of $\text{U}_{0.5}\text{Lu}_{0.5}\text{Cu}_5$ would closely follow the linear behavior evident in UCu_4Pd , and UCu_4Ni , and Fig. 5 demonstrates that the data fit well to the relation,

$$\rho = 210 - 0.58T \quad (7)$$

over the entire temperature range of 0.34 K to 18.5 K. The fit indicates that ρ_0 for $\text{U}_{0.5}\text{Lu}_{0.5}\text{Cu}_5$ is lower than those of UCu_4Pd and UCu_4Ni , $\rho_0 = 258 \mu\Omega \text{ cm}$, and $446 \mu\Omega \text{ cm}$, respectively. Additionally, the slopes of ρ for UCu_4Pd and UCu_4Ni are $-4.1 \mu\Omega \text{ cm T}^{-1}$ and $-1.22 \mu\Omega \text{ cm T}^{-1}$, respectively, suggesting that temperature dependence plays less of a role in the behavior of $\text{U}_{0.5}\text{Lu}_{0.5}\text{Cu}_5$ than that of UCu_4Pd and UCu_4Ni . Despite these distinctions, $\text{U}_{0.5}\text{Lu}_{0.5}\text{Cu}_5$ is indeed exhibiting nFl behavior.

IV. CONCLUSIONS

Based on the x-ray diffraction data (not shown), as well as the lattice parameters (Fig. 1), we concluded that the $\text{U}_{1-x}\text{Lu}_x\text{Cu}_5$ samples formed single phase compounds in the correct cubic AuBe_5 structure. We were successful in suppressing antiferromagnetism at a concentration between $x = 0.35$ and $x = 0.5$, which suggests that a QCP exists. Future work should include specific heat measurements on $\text{U}_{0.65}\text{Lu}_{0.35}\text{Cu}_5$ to look for a plateau in C/T around 5 K, to test the existence of an antiferromagnetic ordering in $\text{U}_{0.65}\text{Lu}_{0.35}\text{Cu}_5$.

In summary, both $U_{0.65}Lu_{0.35}Cu_5$ and $U_{0.5}Lu_{0.5}Cu_5$ shows signs of non-Fermi liquid behavior, but further work is needed to pinpoint the concentration at which $T_N \rightarrow 0$.

ACKNOWLEDGEMENTS

D. J. Burnette would like to give a special thanks to Drs. G. Stewart and J. Kim for all of their time, patience, guidance, and extensive knowledge put forth to make this project possible. The authors would also like to thank NSF and the University of Florida Physics Department for providing the opportunity to participate in this REU program. Thanks to Dr. K. Ingersent, Dr. S. Hershfield and Kristin Nichola for all of their time and effort put in to making this program such a success.

References

- [1] A. J. Schofield, *Contemporary Physics*, **40**, 95 (1999).
- [2] G. R. Stewart, *Rev. Mod. Phys.* **73**, 797–855 (2001).
- [3] C. L. Seaman, M. B. Maple, B. W. Lee, S. Ghamaty, M. S. Torikachvili, J.-S. Kang, L. Z. Liu, J. W. Allen, and D. L. Cox, *Phys. Rev. Lett.* **67**, 2882–2885 (1991).
- [4] W.W. Kim, J. S. Kim, B. Andraka, and G.R. Stewart, *Phys. Rev. B* **47**, 12403 (1993).
- [5] B. Andraka and G. R. Stewart, *Phys. Rev. B* **47**, 3208–3212 (1993).
- [6] D. J. Mixson, J. S. Kim, M. Swick, T. Jones, and G. R. Stewart, *Phys. Rev. B* **73**, 125106 (2006).
- [7] K. Heuser, E.-W. Scheidt, T. Schreiner, and G. R. Stewart, *Phys. Rev. B* **57**, R4198 (1998).
- [8] B. Bogenberger and H. von Loehneysen, *Phys. Rev. Lett.* **74**, 1016 (1995).
- [9] S. Koerner, A. Weber, J. Hemberger, E.-W. Scheidt, and G. R. Stewart, *J. Low Temp. Phys.* **121**, 105 (2001).

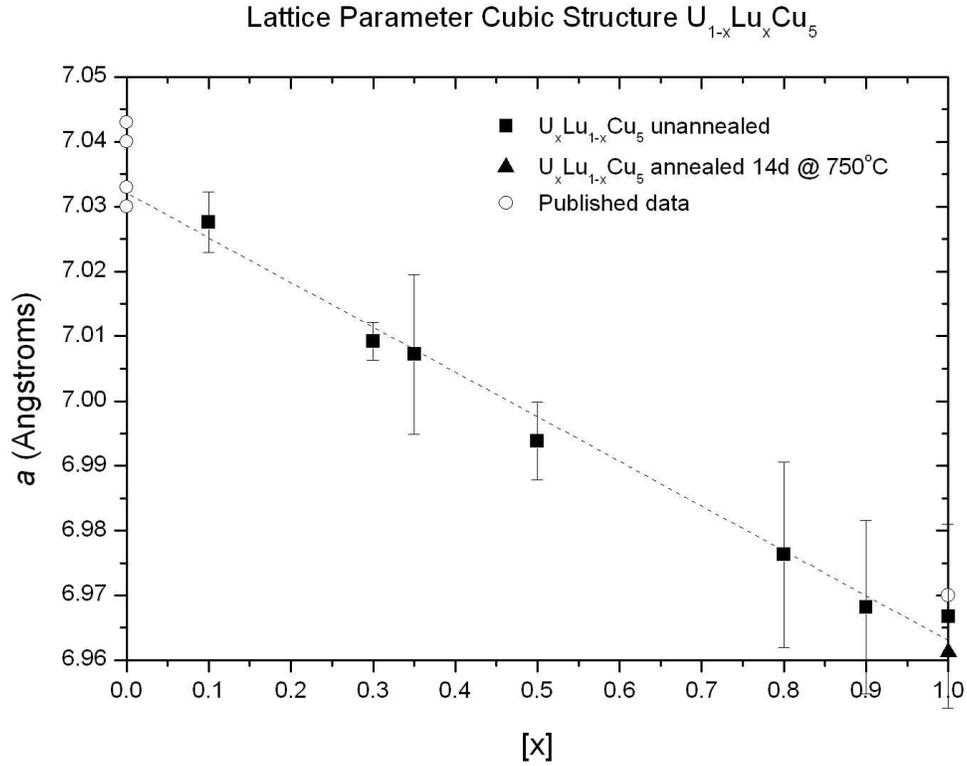


Fig. 1: Lattice parameters as a function of Lu doping, clearly showing a linear relationship.

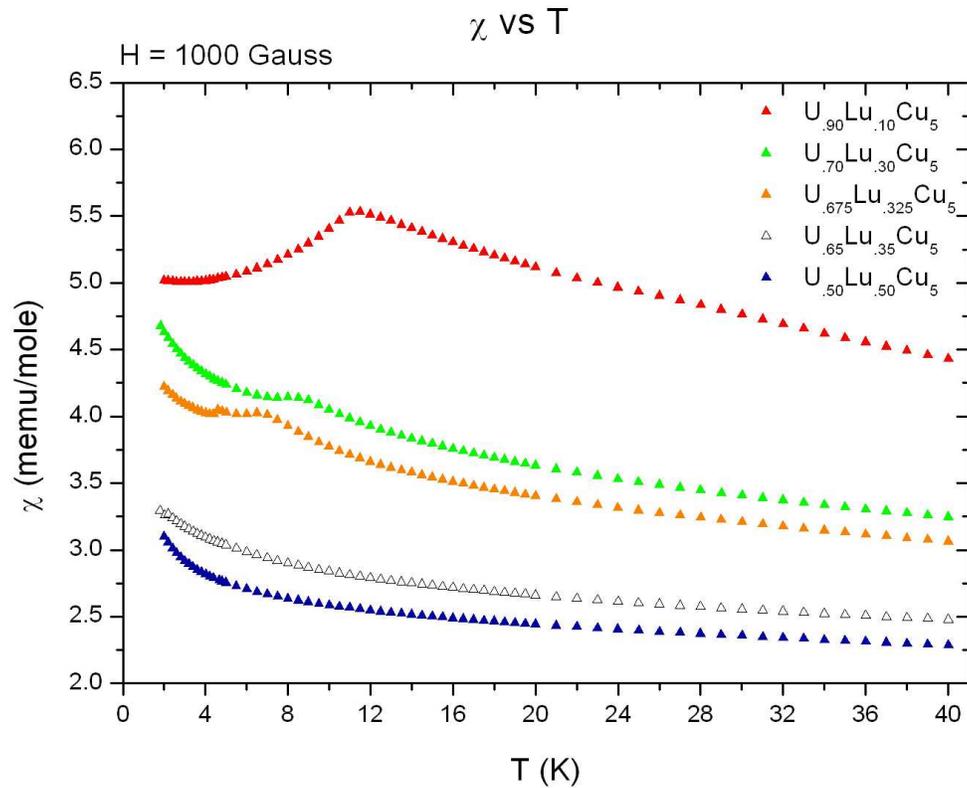


Fig. 2: Magnetic susceptibility vs. temperature for various Lu concentrations x . The Néel temperature T_N , is indicated by a kink in the curve.

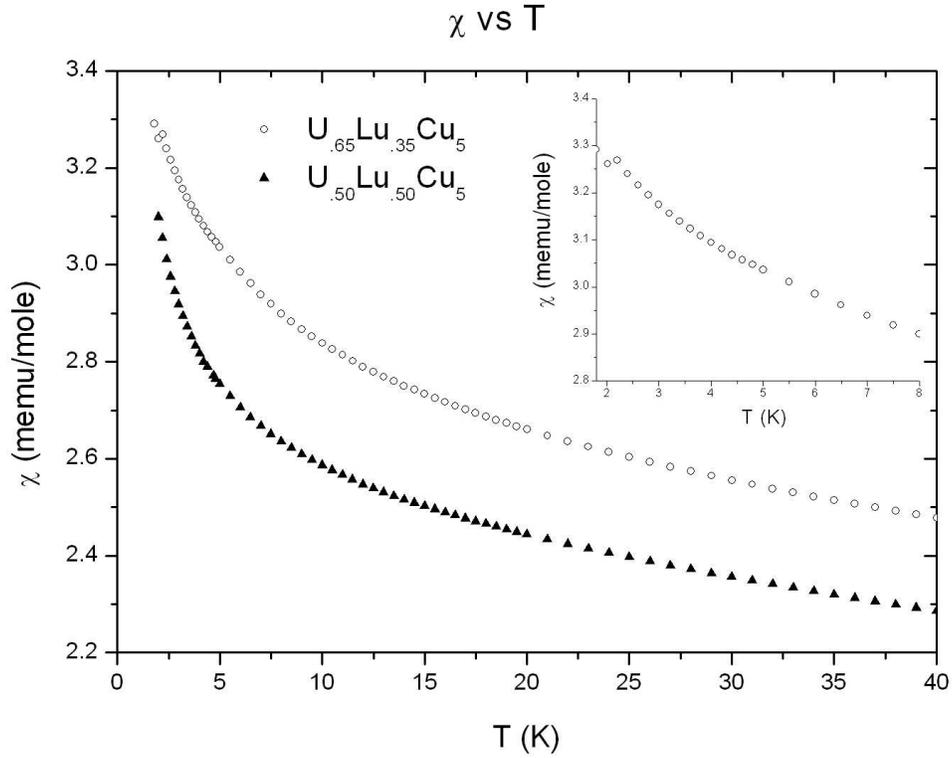


Fig. 3: Magnetic susceptibility vs. temperature for both $U_{0.65}Lu_{0.35}Cu_5$ and $U_{0.5}Lu_{0.5}Cu_5$. The $U_{0.65}Lu_{0.35}Cu_5$ data contain a small kink around 5K, which could be indicative of a small antiferromagnetic transition, while the $U_{0.5}Lu_{0.5}Cu_5$ susceptibility is purely paramagnetic.

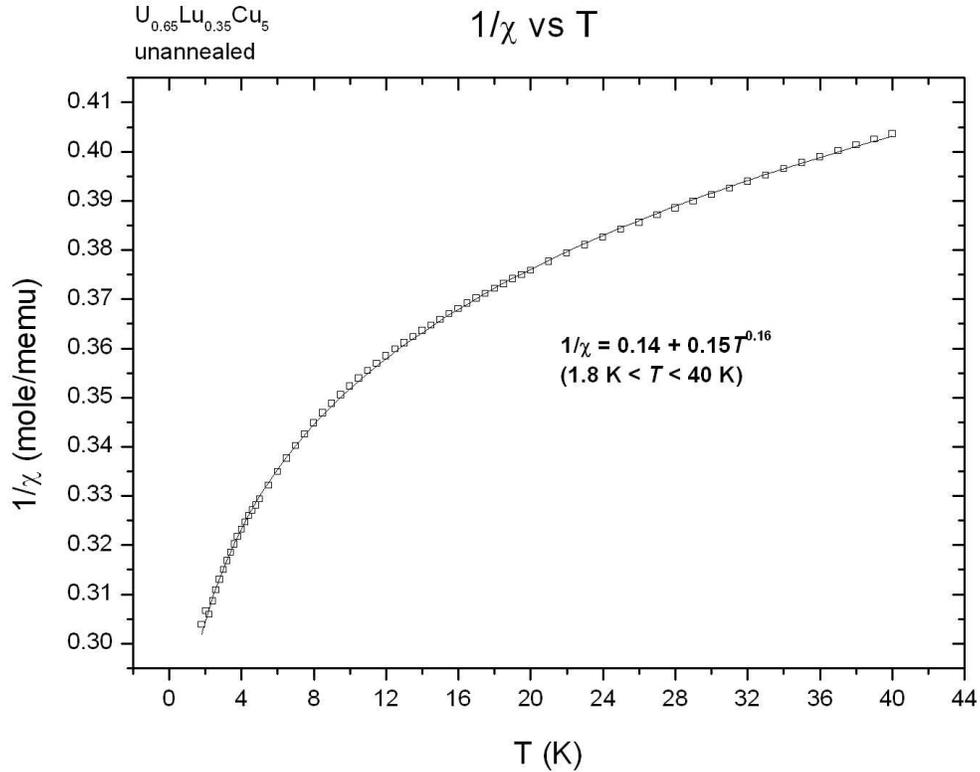


Fig. 4: A curve fit of $\chi^{-1} = \chi_0^{-1} + AT^a$ for $U_{0.65}Lu_{0.35}Cu_5$ over the range 1.8 K to 40 K.

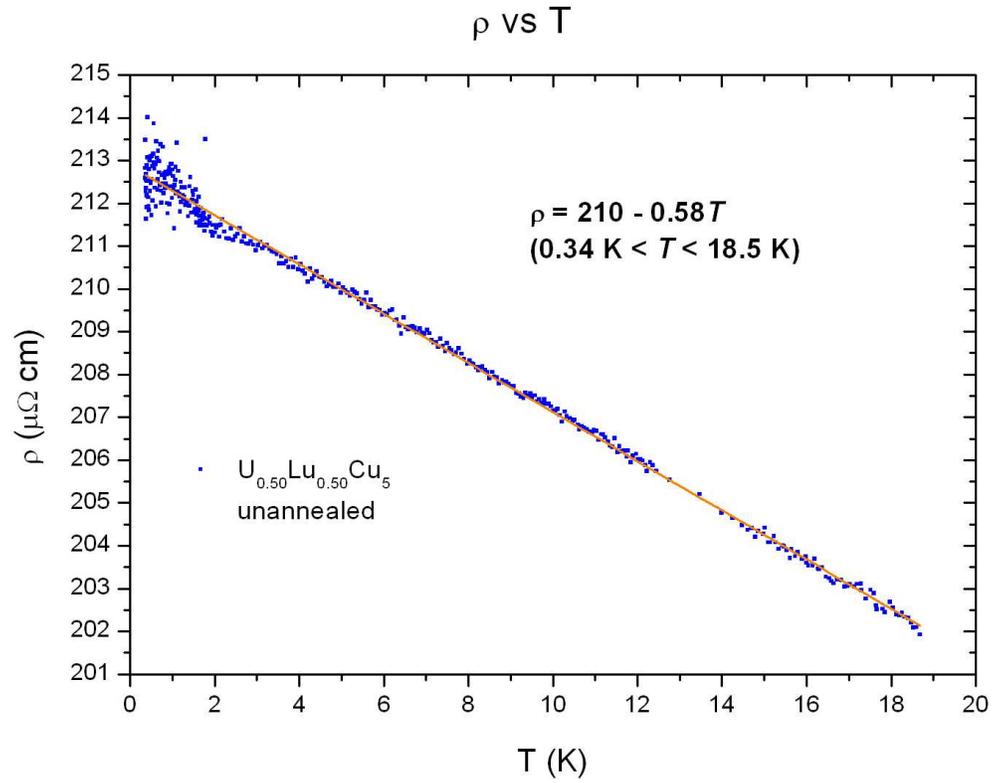


Fig. 5: Fit of $\rho = 212.9 - 0.58T$ to the resistivity of $\text{U}_{0.5}\text{Lu}_{0.5}\text{Cu}_5$ over the range of 0.34 K to 18.5 K.