# Introducing disorder into quantum critical systems

**Cory Bailey, Morehouse College**

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***Advisor: Dr. Greg Stewart***
***Post Doc: Dr. Jungsoo Kim***

## Abstract:

The overall concept of the project is to introduce disorder into quantum critical systems through a grinding technique. The REU student prepared four samples: $U_{0.8}Y_{0.2}Pd_3$, $UCu_4Pd$, $U_{0.07}Th_{0.93}Cu_2Si_2$, and $UPt_4Au$. Using an arc-melting technique, the weighed out elements were arc-melted into a bead that was melted five times. Part of the bead was kept to measure the magnetic susceptibility, and the rest of the bead was ground into a finer powder (less than 25 microns) and a coarser powder (up to 63 microns-in the REU student's case 45 microns). A pellet was made from the less than 25 micron powder (finer). X-ray slides were made out of both the finer and coarser powders.

## INTRODUCTION

From a classical mechanical point of view, physicists have looked at a metal by observing all of the degrees of freedom of the electrons. However, the electronic specific heat of metals was observed to be much less than the expected value of $3\ k_B T$. This discovery led them to see that electrons operating in a metal could not be treated as a Fermi-gas like the proton. Thus, the Fermi-gas theory was proven to be inaccurate.

A Fermi-gas is defined as a collection of non-interacting fermions. In contrast, physicists believed that there was an interaction between electrons, which led to Landau's Fermi-liquid model that accounts for a gas of interacting particles. Landau's model predicts certain temperature dependences at sufficiently low temperatures (often $\leq 1$ K) for physically observable quantities. Seaman presented data at the 1991 American Physical Society meeting that strongly disagreed with the Fermi-liquid model of Landau. From that disagreement, physicists have developed the theory of a non-Fermi liquid, a

deviation from Landau's model. A 'non-Fermi liquid' is a system that exhibits properties disagreeing with those predicted by Landau for a Fermi-liquid. For example, Landau predicted that the specific heat C, divided by the temperature T, approaches a constant (called the Sommerfeld coefficient $\gamma$) as T tends to 0. The low- temperature magnetic susceptibility $\chi$ also remains temperature-independent, while the electrical resistivity $\rho$ goes as $\rho(0) + AT^2$. When Seaman *et al.* discovered that the particle-particle interactions in the $Y_{1-x}U_xPd_3$ were too strong and long-ranged to obey the model predicted by the Fermi-liquid theory (the interactions remained temperature dependent as $T \rightarrow 0$, causing a violation of the simple Fermi-liquid temperature dependences), it led to a new field in condensed matter physics: the non-Fermi liquid. A lot of theoretical explanations followed after (and even-as predictions-before!) this discovery, but no single theory has been successful at describing the behavior of a non-Fermi liquid system [4].

Four main theories played a significant role for experimentalists in explaining non-Fermi liquid behavior. First, metals close to a quantum critical point: during a phase transition at a zero absolute temperature, the quasiparticles diffuse forcibly and fail to act in a way that the Fermi-liquid theory (compare the excited states of the quantum liquid with those of a free Fermi-gas) would predict. Second, metals in one-dimension- the Luttinger liquid: in one-dimensional metals, electrons are unstable and decay into two separate particles that carry the electron's spin and charge respectively. Third, two-channel Kondo models: when two independent electrons can scatter from a magnetic impurity it leaves behind "half an electron." Finally, disordered Kondo models; the scattering from disordered magnetic impurities is too strong to allow the Fermi-quasiparticles to form [1-4]. Although scientists use many ways to identify a system as a

non-Fermi liquid, we are only working with models based on disorder. Through grinding, the idea is that we are going to introduce disorder, which means slightly disturbing (just a little). We use grinding to introduce disorder in the lattice holding the f-ion uranium in a case where the f-electrons are very strongly correlated to one another. It is hoped that a slight disorder in the lattice will have a large effect in the low temperature magnetic susceptibility $\chi$, which is a measure of the correlation strength of the f-electrons. Our systems are disordered because when ground, one sees a slight change in the phase transitions a well as the values of $\chi$.

Our project involved introducing disorder into systems that have interesting electronic properties such as: large magnetic susceptibilities and large specific heats with non Fermi-liquid temperature dependences. These systems are called quantum critical systems. Through experimentation, we found that disorder plays a much larger role in the behavior of the large magnetic susceptibilities and large specific heats than one would expect. Most experiments introduce disorder through doping (replacing an element with another element within a system: this process changes the properties of a sample); however, we introduce disorder through a very simple mechanical grinding process on samples produced by arc-melting. We characterize the disorder by measuring the high-angle x-ray diffraction peaks. Varying degrees of disorder are introduced by making finer (less than 25 micron) powder or coarser (up to 63 micron) powder.

## EXPERIMENTAL METHOD

### Sample preparation

In this research involving ‘non-Fermi liquid’ behavior, four systems were made: $U_{0.2}Y_{0.8}Pd_3$, $U_{0.07}Th_{0.93}Cu_2Si_2$, $UCu_4Pd$, and $UPt_4Au$. We began by weighing out high-purity elements to the desired stoichiometric proportions to a precision of 0.15 milligrams. After the samples were weighed out, they were placed into an Edmund Bühler arc-melter where they were melted to form a single bead. The samples were melted in an ultra-high-purity argon atmosphere using a zirconium getter. Each sample was made successfully by keeping the mass loss, due to melting, below 0.2%. After melting the samples, the samples were ground using a mortar and a pestle.

## Grinding

The bead was placed in the mortar and gently tapped with the pestle. Once the bead was broken into several pieces we kept a chunk of the bead to measure $\chi$ and the remainder was ground into finer and coarser powders. The grinding process, depending on the sample, usually required 15 minutes. Once the sample was ground and rinsed with acetone carefully to ensure no powder was removed, the powder was placed in sieves (45 μm, 38μm, and 25μm). The 38μm sieve was nested onto the 45 μm sieve and the 25 μm sieve was placed on large weighing paper, which was placed on top of 6" weighing papers. We gently tapped the side of the sieves to ensure that the powder went through the sieves to its respective size.

## Pellet preparation

After gently tapping the sides of the sieves for about two or three minutes, we used large weighing papers to keep track of the different sizes of powders. Immediately after

removing the powder from the sieves, we used about 50 mg of the less than 25 μm powder to make a pellet and about 65 mg of that same powder to prepare an x-ray slide. To make a pellet we used a Carver Laboratory Press, which allowed us to press 50 mg of the less than 25 μm powder into a pellet. Also, with the 45 μm powder an x-ray slide was made

## X-ray diffraction

To x-ray these slides, we used a Phillips XRD 3270 x-ray diffractometer (XRD) at the University of Florida's Major Analytical Instrumentation Center (MAIC). The physics behind the XRD is that high-energy photons with wavelength 1.54056 Å irradiate the sample and some are diffracted by the crystalline structure of the sample. A counter records the number of diffracted photons as a function of their direction. 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 diffraction of the photons, we can identify the structure of the sample. The nice aspect of measuring the x-ray diffraction of the sample is that the high-angle line widths are a direct measure of the lattice disorder. The typical x-ray is performed for 2θ from 20° to 120°. However, in some cases we focused on a few peaks in the high-angle range. When focusing on these peaks, x-rays were performed typically for 2θ from 75° to 100°. These high-angle resolutions allowed us to see peak-broadening in the 25 μm powder. From observations, the lines of the finer powder are supposed to broaden.

## Magnetic susceptibility

After the samples were x-rayed, the magnetic susceptibility, $\chi$ was measured using the Superconducting Quantum Interference Device (SQUID). Magnetic susceptibility is defined as the magnetization of a material in response to an applied magnetic field at a fixed temperature and pressure, or

$$M = \chi_v H, \quad (2)$$

where M is the magnetization of the material (the magnetic dipole moment per unit volume), measured in amperes per meter, and H is the applied field, also measured in amperes per meter. We measured magnetic susceptibility as a function of temperature with an applied external magnetic field. The measurements were performed in a field of 1000 gauss over a temperature range of 2-40 K. Measured values reported in memu per mole.

# RESULTS

## Disordered systems

Two of the systems studied, $U_{0.2}Y_{0.8}Pd_3$ and $U_{0.07}Th_{0.93}Cu_2Si_{2,}$ were disordered. "In further investigation of the evolution of the localized 5f behavior in the U-dilute side of the phase diagram, Seaman *et al.* (1991) discovered that $UPd_3$ occurs in the D024, hexagonal $Ni_3Ti$ structure and is one and only a few unknown U systems in which neutron scattering shows clearly the crystalline electric field levels (Shamir et al. 1978). The photoemission work of Kang et al. (1989) showed a decrease in the energy separation between the localized 5f level and $\varepsilon_f$, the Fermi energy, as U is replaced with Y, which the authors identified as a way to “tune” $\varepsilon_f$ via the change in the conduction-

electron density affected by the substitution of $Y^{3+}$ for $U^{4+}$."[4] "Further, the origin of heavy fermions in the Uranium -- (U-) based intermetallics is controversial. The greater the extent of 5f orbitals has led to debate over itinerant versus localized treatments. In the localized limit it is unclear whether to take the U ions as nominally tetravalent ($5f^2$) or trivalent ($5f^3$), and it has been questioned whether crystal-field excitations are at all relevant. Given the evidence for localized $5f^2$ states in $UPd_3$ work was commenced to dilute this system by substituting U to Y to form $Y_{1-x}U_xPd_3$ ." [2]

For $Th_{0.85}U_{0.15}Cu_2Si_2$, the magnetic susceptibility $\chi$ reveals ordering at about 12 K. Such behavior is reminiscent of heavy-fermion systems with very small ordered moments, where neutron-scattering experiments reveal an ordered moment, but no specific anomaly is seen at the transition temperature. To estimate the magnitude of the ordered moment the remnant magnetization can be used: with 500 emu/ U mol for $Th_{0.85}U_{0.15}Cu_2Si_2$ we get an ordered moment of about 0.1 $\mu_\beta$ per U atom. The extrapolation of the high-field data yields roughly twice this value. Therefore small values of only 0.1-0.2 $\mu_B$ give a rough estimate for the ordered moment of this compound. The data suggest that replacing uranium in $UCu_2Si_2$ by thorium suppresses the magnitude of the ordered moment [5]. When uranium replaces thorium, the system becomes disordered.

We further see the disorder in the systems by looking at the magnetic susceptibility, $\chi$, measurements shown in Figures 1 and 2. From observing the Figures 1 or 2, one can see an increase in the measurement of $\chi$ for the pellet compared to the chunk. However, since the system is already disordered that increase is only about 10%. From our studies with these systems, we know that grinding disturbs the system further

introducing disorder; however, we also know that when a system is already disordered it is difficult to grind the system to try to introduce more disorder. When trying to introduce disorder on an already disordered system, we see a small increase of the magnetic susceptibility as shown in figures 1 and 2.

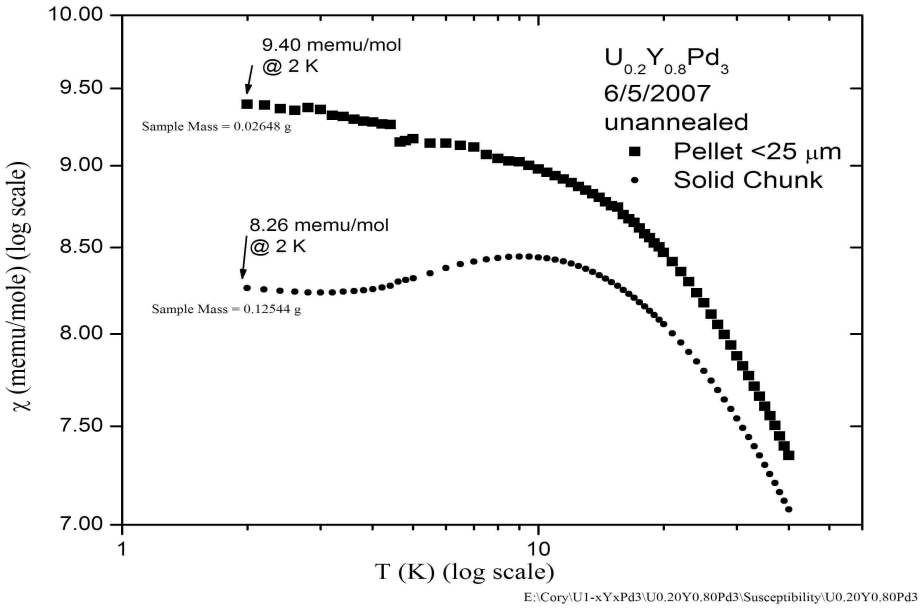


Figure 1: Magnetic susceptibility vs. temperature for $U_{0.2}Y_{0.8}Pd_3$.

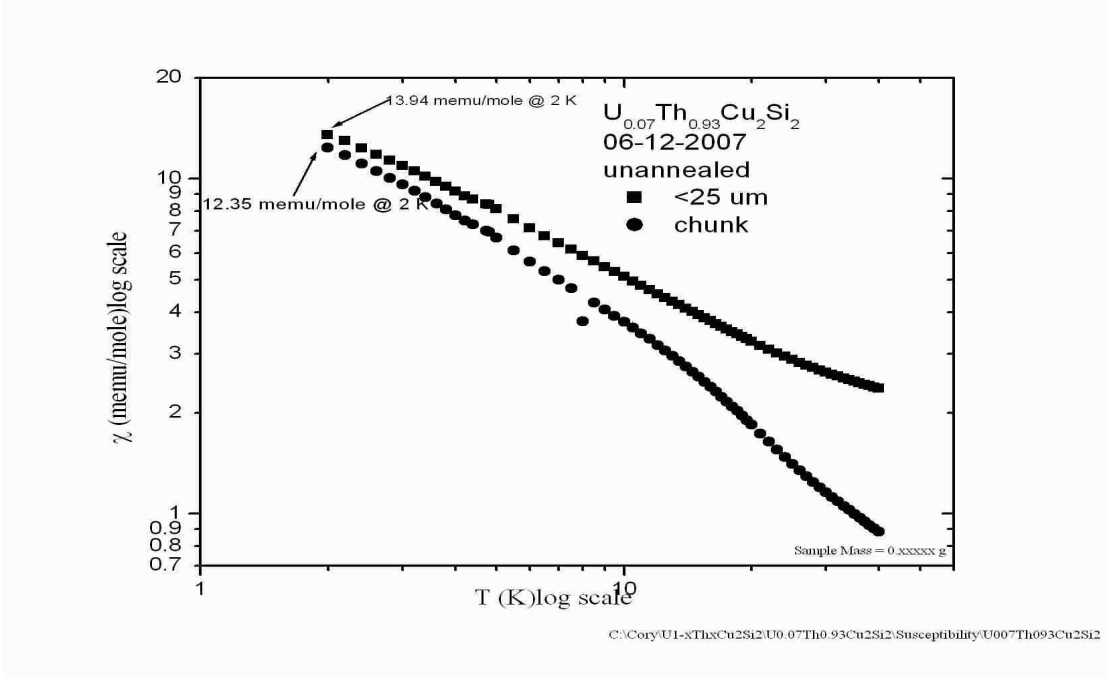


Figure 2: Magnetic susceptibility vs. temperature for $U_{0.07}Th_{0.93}Cu_2Si_2$.

## Atomic radii and disorder

According to the x-ray diffraction spectra as shown in figures 3 and 4, these two disordered systems do not produce a perfectly cubic shape--they are somewhat hexagonal. In our studies, we have come to find out that most ordered systems follow some sort of pattern and that pattern is a cubic pattern. However, in $U_{0.2}Y_{0.8}Pd_3$ and $U_{0.07}Th_{0.93}Cu_2Si_2$ we have a mixture of both cubic and hexagonal patterns, which further makes the systems disordered. These systems are also ordered and disordered in part due to the atomic radii of each element as seen in Table 1. When the atomic radius of the second element is larger than the atomic radii of the first element of the system, the system is disordered. For example, in the systems $U_{0.2}Y_{0.8}Pd_3$ and $U_{0.07}Th_{0.93}Cu_2Si_2$, the atomic radii of both thorium (1.77 Å) and yttrium (1.80 Å) are larger than the atomic radius of uranium (1.54 Å). However, if the atomic radius of the second element is

smaller than the atomic radii of the first element in the system, the system is ordered. For example, in the systems $UCu_4Pd$ and $UPt_4Au$ the atomic radii of Cu (1.28 Å) and Pt (1.38 Å) are smaller than the atomic radius of uranium (1.54 Å).

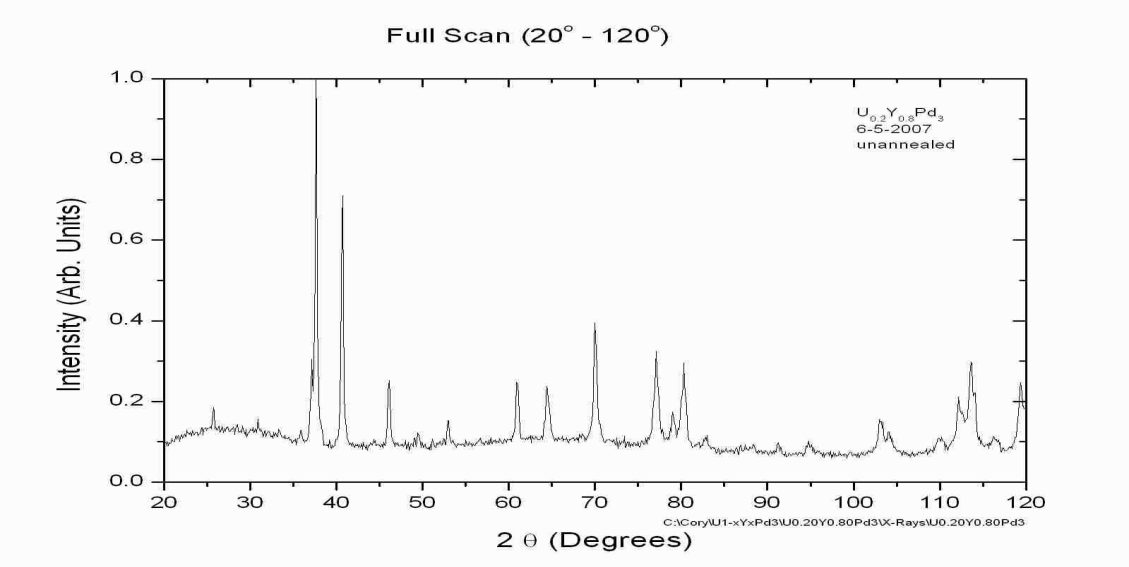


Figure 3: X-ray diffraction spectrum for $U_{0.2}Y_{0.8}Pd_3$.

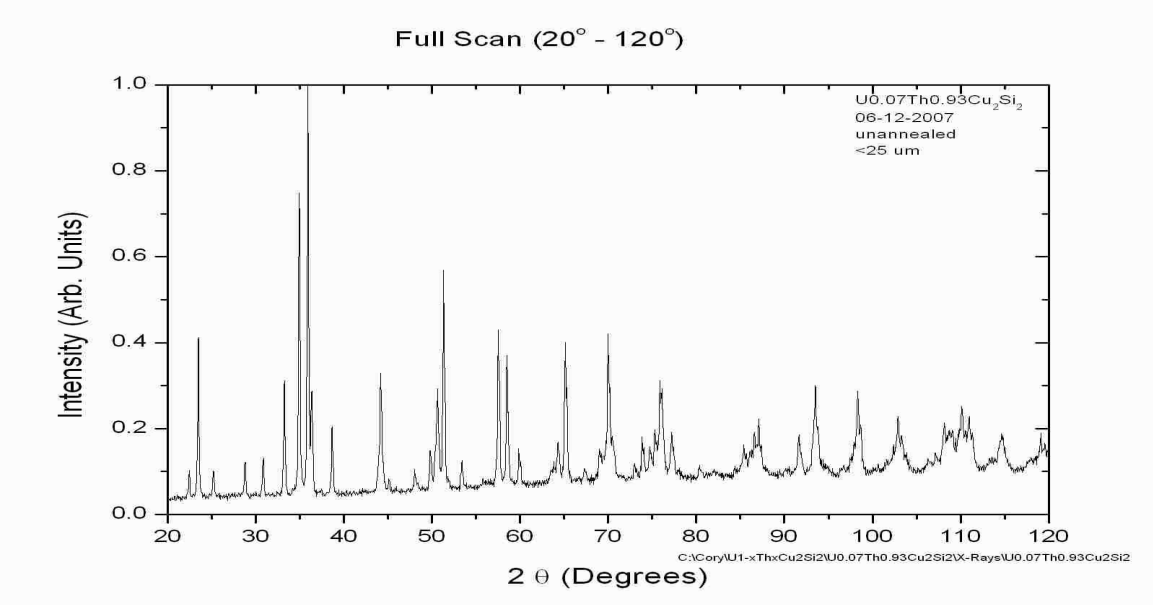


Figure4: X-ray diffraction spectrum $U_{0.07}Th_{0.93}Cu_2Si_2$.

Table 1: Atomic radii

| Element | Atomic Radius (Å) |
|---|---|
| uranium (U) | 1.54 |
| palladium (Pd) | 1.37 |
| thorium (Th) | 1.77 |
| copper (Cu) | 1.28 |
| silicon (Si) | 1.32 |
| platinum (Pt) | 1.38 |
| gold (Au) | 1.44 |
| yttrium (Y) | 1.80 |

## Ordered systems

The non-Fermi liquid system $UCu_{5-x}Pd_x$ was discovered soon after $U_{0.2}Y_{0.8}Pd_3$, and was reported to have C/T ∝- ln T below 10 K down to 0.3 K, the lowest temperature measured, for x= 1.5, where as C/T for x = 1.0 was found to vary as $T^{-0.32}$ between 1 and 10 K. $UCu_{3.5}Pd_{1.5}$ and $UCu_4Pd$ both occur in the cubic, cF24 AuBe5 structure [3]. Further contributing to the disorder of the $UCu_4Pd$ system is not all the of Pd is on the Be I site and all of the Cu is on the Be II site. However, 24 ± 3 % of the Pd occupies Be II sites [4]. This holds true as well for the $UPt_4Au$ system, which also occurs in the cubic,

cF24 AuBe5 structure. Instead of all the Cu being on the Be II site, some (but not all) of the Pt is on the Be II site as well.

When viewing the magnetic susceptibilities of these two systems (see figures 5 and 6), one can see that the magnetic susceptibility increased by 20% on grinding. The increase of 20% in the magnetic susceptibility shows us that the systems are ordered. Theoretically, the magnetic susceptibility was supposed to increase by 20% or higher to show that the system was indeed ordered. This significant increase in magnetic susceptibility shows two things: first, the system was indeed ordered and second, we introduced disorder when grinding the sample, which is exactly what we were trying to do. We further see that these two systems, $UCu_4Pd$ and UPt4Au are ordered from the perfect cubic shape that their x-ray diffraction spectra produce (see figures 7 and 8).

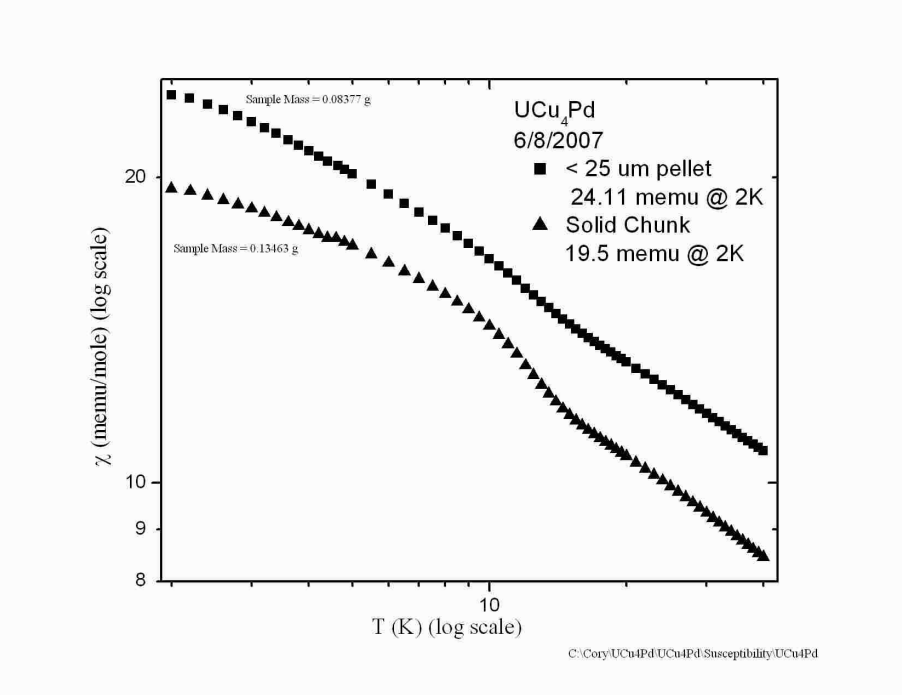


Figure 5: Magnetic susceptibility vs. temperature for $UCu_4Pd$.

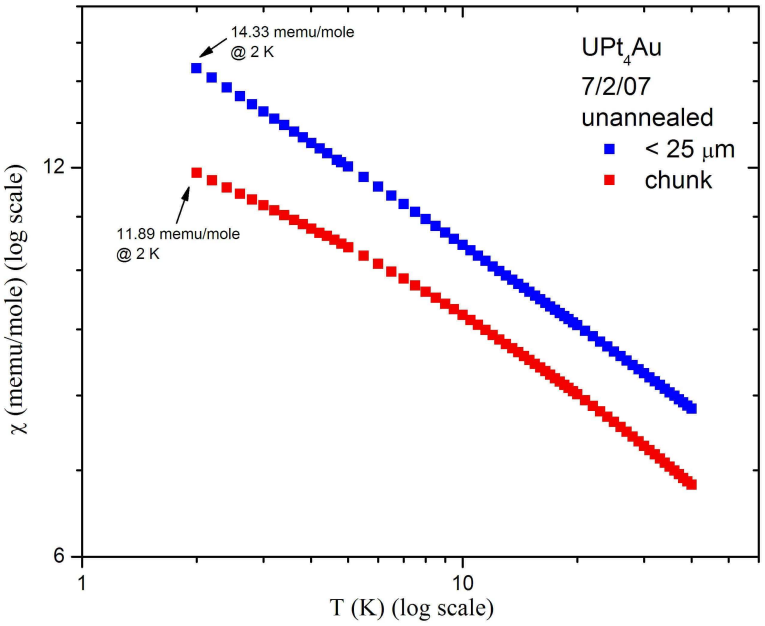


Figure 6: Magnetic susceptibility vs. temperature for $UPt_4Au$.

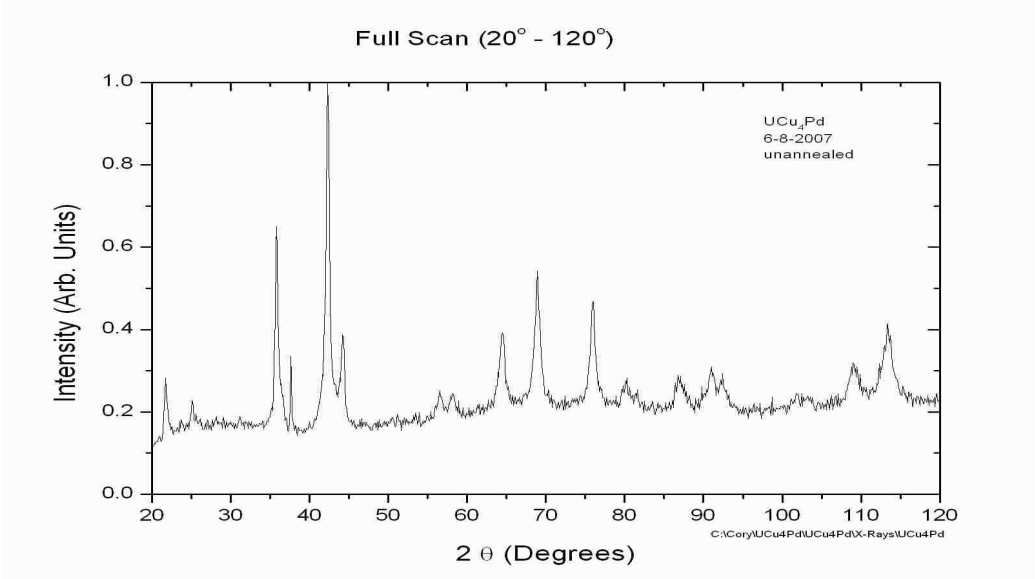


Figure 7: X-ray diffraction spectrum for $UCu_4Pd$.

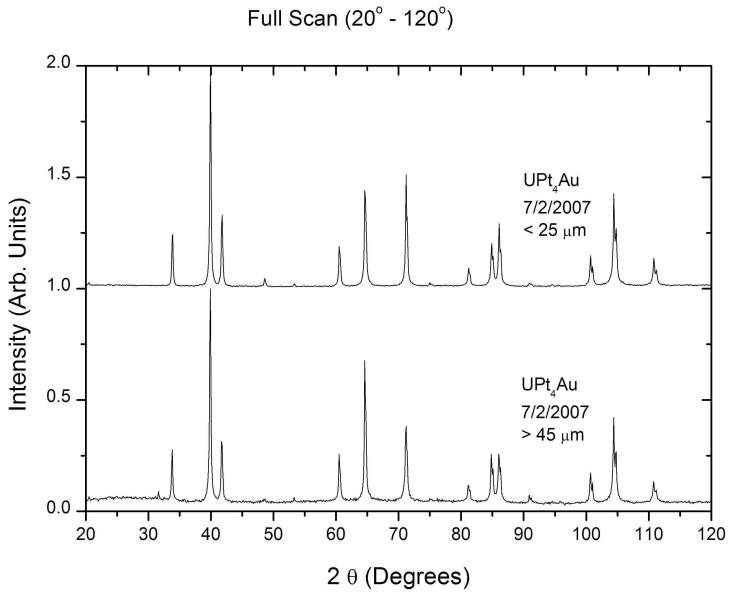


Figure 8: X-ray diffraction spectrum for $UPt_4Au$.

## High-angle scattering

When we introduced disorder through a grinding technique, we further began to look at the x-ray diffraction spectra. In doing so, we noticed that some of the samples had interesting peaks at very high angles. So, we x-rayed the samples at high angles (typically from 75°-100°: in some cases 110°-117°).When we looked at the x-ray diffraction spectra at very high angle ranges, we observed peak broadening. Almost all the samples, except $UPt_4Au$, were measured at high-angle resolutions; however, we did not see peak broadening in all the samples. We mostly saw peak broadening in the ordered systems as seen in figures 9 through 11.

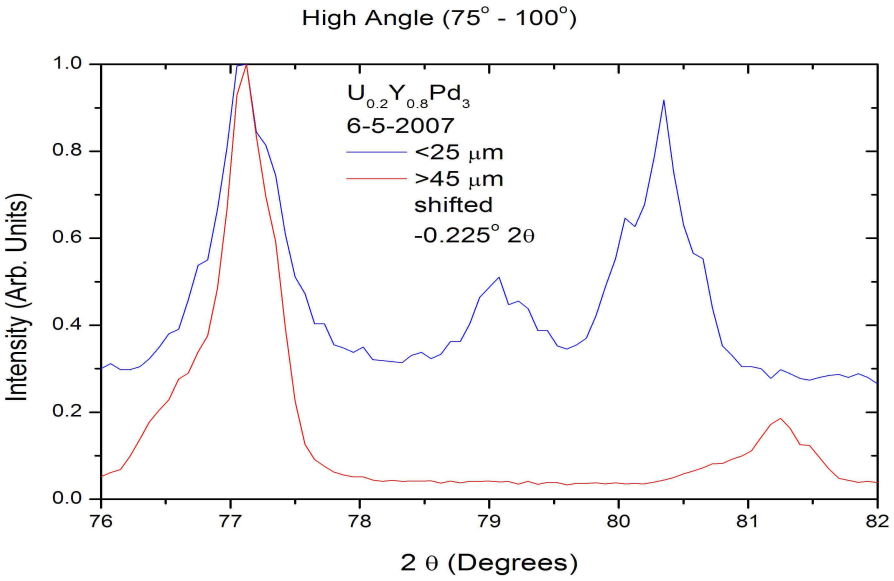


Figure 9: High-angle diffraction spectrum of $U_{0.2}Y_{0.8}Pd_3$

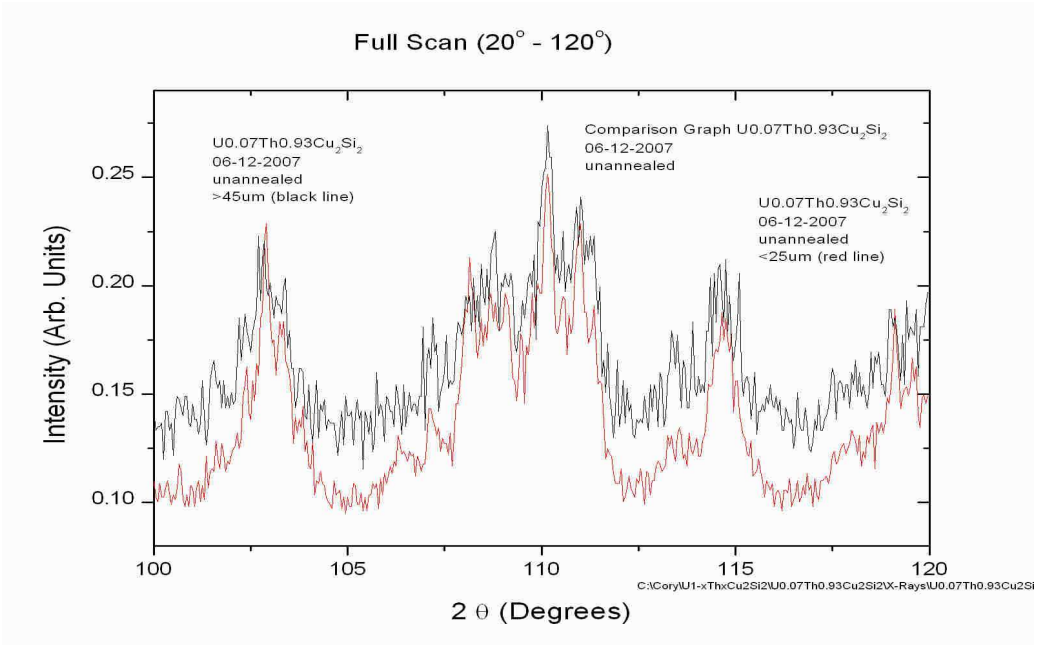


Figure 10: High-angle diffraction spectrum for $U_{0.07}Th_{0.93}Cu_2Si_2$

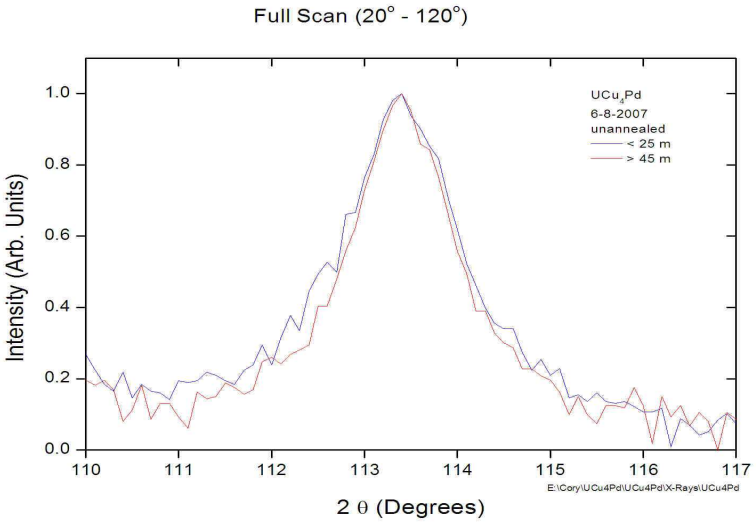


Figure 11: High-angle diffraction spectrum of $UCu_4Pd$

## CONCLUSIONS

During this project, our main objective was to introduce disorder, through grinding, into systems with large magnetic susceptibilities and large specific heats. By introducing disorder into these quantum-critical systems, we hoped to see an increase in the magnetic susceptibilities and specific heats. Throughout our research, we observed a 20% increase in the magnetic susceptibilities of the ordered systems. However, due to time constraints, we were not able to measure the specific heats of these quantum critical systems. In this research project we postulated several theories: first, $U_{0.07}Th_{0.93}Cu_2Si_2$ and $U_{0.2}Y_{0.8}Pd_3$ were disordered systems; second, UCu4Pd and $UPt_4Au$ were ordered systems; third, one can not introduce disorder onto an already disordered system; and four, when introducing disorder through a grinding process on an ordered system, a consistent increase or higher will occur in the magnetic susceptibility. Although we could

not measure the specific heat, our prediction is that there should also be a 20% increase in the specific heat.

## ACKNOWLEDGMENTS

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