Non-Fermi Liquid Behavior in UCoAl

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Abstract

The compound UCoAl was prepared and then characterized by x-ray diffraction. The magnetization versus field curve indicates that a transition between a paramagnetic ground state and a ferromagnetic state occurs. Magnetic susceptibility and resistivity measurements were obtained and curve-fitting was performed. The electrical resistivity was fit to the curve $\rho = \rho_o + AT^\alpha$ for $\alpha = 1.619$, while the susceptibility was fit to several equations. The temperature dependent susceptibility and $T^{1.619}$ dependence of the resistivity indicate that UCoAl may be a non-Fermi liquid.
Introduction

In the mid to late 1950's, Lev Landau published a series of influential works detailing the metallic behavior of compounds that was based on the single electron picture. In his theory, Landau assumed that the compound could be viewed as a gas of Fermi particles (i.e., an electron gas) in which the particle-particle interactions are weak and short ranged, both in time and space, and can thus be regarded as relatively insignificant [1]. The particles described by Fermi-liquid theory are not the original electrons per se, but rather are electron-like “quasiparticles” that are formed from the interacting electron gas [2]. In spite of the known strength of electron-electron interactions, Fermi-liquid theory works surprisingly well for a wide variety of metals, including UPt3, a compound in which the electron interactions are known to be important.

Fermi-liquid theory makes various predictions regarding the temperature dependencies of several physical properties in the low temperature regime [3]. First, the electrical resistivity of the sample is predicted to follow $\rho = \rho_o + AT^2$. Also, the magnetic susceptibility $\chi$ is expected to be temperature independent at these low temperatures (i.e., $\chi = \text{constant}$). Finally, as the temperature approaches 0 K, the specific heat ($C$) divided by the temperature also becomes equal to a constant (known as the Sommerfield coefficient $\gamma$). Thus, for a compound obeying Fermi-liquid theory, plots of $\chi$ and $C/T$ versus temperature should have a region in the low temperature zone where the slope of the line becomes zero.

Then in 1991, Seaman et al. found that the Y$_{1-x}$U$_x$Pd$_3$ system showed a strong deviation from what is predicted by Landau’s Fermi-liquid theory [3,4]. They discovered that the electron-electron interactions were too strong and long ranged to allow a
description by Landau’s theory. With the discovery of this system a new field of physics
was born: non-Fermi liquids. Over the next few years, and leading up to the present day,
a multitude of systems have been found that display non-Fermi liquid behavior. These
systems possess physical properties \((C/T\) and \(\chi\)) having unique temperature dependencies
that can extend for over a decade in the low temperature regime [3]. Different results
occur regarding the electrical resistivity as well. It has been found that the resistivity does
not follow \(T^2\) as in the Fermi liquid theory, but rather it follows \(T^{\alpha}\) with \(\alpha \neq 2\) [3].
Unlike Landau’s Fermi liquid theory, there is not a unifying theory for non-Fermi liquids
that can be used to describe all the systems with this behavior.

In the present experiment, the intermetallic uranium compound UCoAl was
examined in its polycrystalline form for non-Fermi liquid behavior. Previous
experiments have indicated that UCoAl may be at (or close to) a quantum critical point,
making the compound an excellent candidate for possible non-Fermi liquid behavior
[3,5]. If UCoAl turns out to be a non-Fermi liquid, it will be one of a handful of systems
that displays this behavior in its natural state without the aid of doping, field, or pressure.

To determine if UCoAl is a non-Fermi liquid we measured the electrical
resistivity from 0.3 K up to 10.5 K. This process was performed using a standard four-
wire dc setup and utilized liquid helium 3 in order to reach 0.3 K. Also determined was
the magnetic susceptibility as a function of temperature, from 1.8 to 40 K. A
Superconducting Quantum Interference Device (SQUID) magnetometer was employed to
perform this measurement. The data obtained were then fit to various equations that are
known to describe non-Fermi liquid behavior.
Procedure

Sample Preparation

Before experimentation could begin a pure sample of UCoAl had to be prepared. First, we calculated the correct stoichiometric amounts (1:1:1) of each metal via some fairly straightforward mathematics. After determining the proper masses of U, Co, and Al, we cut each metal into pieces of the correct mass. Because uranium is a radioactive element, all the necessary safety guidelines were followed.

We then placed the elemental pieces into an arc melter (Edmund Buhler model) and began the melting process. The arc melter that was used in our experiment provided an inert argon (oxygen free) environment in which we could heat the elements to their respective melting points. To ensure homogeneity, we melted the sample three times, flipping the sample after each melt. The total sample mass obtained was within 0.5 % of the theoretically calculated mass, resulting in 1.13606 g.

X-ray Diffraction

Although the UCoAl bead was now prepared, its crystal structure needed to be verified. More specifically, no second phase should be present. We used the technique of x-ray diffraction to determine phase consistency and singularity.

First, a small portion of the prepared UCoAl sample was broken off and then ground into a fine powder using a mortar and pestle. Next, the sample was glued onto a microscope slide (in a 1” x 1” square) using a glue consisting of iso-amyl acetate and
collodion. The sample was now ready for x-ray analysis at the Major Analytical Instrumentation Center (MAIC).

X-ray diffraction is governed by Bragg’s law of reflection which states,

\[ n\lambda = 2d \sin\theta \quad n = 0,1,2,... \]  \hspace{1cm} (1)

in which \( d \) is the distance between atoms, \( \lambda \) is the x-ray’s wavelength, and \( \theta \) is the angle of reflection [2]. During the diffraction process a detector records the angles at which each x-ray is reflected. While some of the incident rays pass directly through, others strike atoms in the lattice and are reflected at an angle \( \theta \). By using the intensity at each angle as a guide, one can later determine the crystal structure as well as the lattice parameters.

Utilizing *Pearson’s Handbook of Crystallographic Data* [6], along with studies such as Mushnikov et al. [7], we determined that UCoAl crystallized in a hexagonal ZrNiAl - type structure. Inputting the theoretical data from our references into the Powdercell software package, a predicted (theory) diffraction plot was generated.

*Magnetic Susceptibility Determination*

To measure the magnetic susceptibility (\( \chi \)), we used a Superconducting Quantum Interference Device (SQUID). We used the SQUID to measure the magnetization as a function of the applied magnetic field (\( H \)) at a constant temperature (2 K), followed by the susceptibility as a function of temperature for a constant magnetic field. The magnetic susceptibility measurements were performed in fields of 0.1 and 1.5 T, and over a temperature range of 1.8-40 K. During the process we also checked for “spin glass” behavior by performing field-cooled and zero field-cooled susceptibility measurements.
Because of anisotropy concerns, the copper hearth upon which we melted the sample was used as a reference for orientation in these measurements. For each data set, the sample was oriented so that the magnetic field was perpendicular to the plane of the copper hearth. Then, using the Origin graphing package, the appropriate plots were created.

**Electrical Resistivity**

A second important quantity in the classification of a compound as a non-Fermi liquid is the electrical resistivity. Before any measurements were to begin, a resistivity bar of proper size had to be cut. First, a small piece of UCoAl was obtained and glued to a dressing stick of suitable size. Next, a low-speed saw equipped with an ultra-thin, diamond-tipped blade was used to cut the sample into thin slices. Since the electrical resistivity ($\rho$) was to be derived from the measured resistance value ($R$), it was important to make sure that the resistance values would be large enough to work with. The equation for resistance,

$$R = \rho L/A$$  \hspace{1cm} (2)

shows that to obtain a large value of $R$, the sample bar should have a large length ($L$) and a small cross-sectional area ($A$). Thus, the goal in cutting our resistivity bar was to maximize the length while minimizing the cross-sectional area.

After an acceptable resistivity bar was obtained, four platinum leads were attached using a spot welder. Next, the $\rho$-bar was fixed in place on a piece of compensated silicon, held down by electrically insulating GE 1701 varnish. Finally, the platinum wires were attached to the silicon platform using highly conductive silver epoxy. The sample was now ready to be mounted onto the appropriate probe.
The silicon platform, containing the UCoAl resistivity bar, was secured to the probe using insulating grease. Then, each of the wires was soldered to the appropriate location on the probe. After a check of the connections to make sure everything was functioning properly, the probe was ready for use.

The resistivity measurements in our experiment ranged from 0.3-10.5 K. Liquid helium-4 was used as the main coolant, while helium-3 was used to allow us to reach 0.3 K. Using a standard four-wire setup, we gathered the values for resistance as a function of temperature. By using Eq. (2), we were able to determine the resistivity values from the measured resistances. Finally, a plot of resistivity versus temperature was created, and the curve was fit to a corresponding equation.

Results

Fig. 1 shows the theoretical and experimental x-ray diffraction data for UCoAl. While there appear to be a few inconsistencies regarding the intensity values, the lack of extraneous peaks and the proper peak positioning led us to believe that our sample crystallized in the correct ZrNiAl structure type. Table I shows a comparison of the lattice parameters generated by the UNITCELL program from experimental \( h-k-l \) values, with those from a prior study. The two parameter sets are within the acceptable error of one another, thus serving as a second check of the crystal lattice, and further strengthening the argument that the correct structure was obtained.

Let us now shift our attention to Fig. 2, which shows the magnetization \( (M) \) versus applied magnetic field data \( (H) \) obtained by a SQUID. One can see a sigmoidal shaped
curve, with a metamagnetic transition occurring at around 6500 gauss (0.65 T). We shall
discuss the possible origin of this transition in the next section.

The susceptibility versus temperature plots (Fig. 3), shows that there is no
appreciable difference between the field-cooled and zero field-cooled susceptibilities.
Thus we may temporarily rule out "spin glass" behavior until proven otherwise. While
\( \chi \) was measured at 0.1 and 1.5 T, we shall focus our attention on the 0.1 T results.
However one may note that for \( \chi \) vs. \( T \) in \( H = 0.1 \) T, a maximum in the susceptibility
curve develops at about 17 K. This maximum, described by Sechovsky et al., then
disappears in the measurements performed at 1.5 T [8].

Using an equation that has previously been used to describe some non-Fermi
liquids (see [3]), the low-temperature susceptibility regime was fit to an equation of the
form,

\[
\chi^{-1} = \chi^{-1} + AT^\alpha. \tag{3}
\]

Fig. 4 shows that this equation holds true over almost a decade of temperature (2-10 K)
with \( \chi^{-1} = 14.8 \text{ mol} \cdot \text{emu}^{-1}, A = 0.66 \text{ mol} \cdot \text{emu}^{-1} \cdot \text{K}^{-1.55} \), and \( \alpha = 1.55 \). For the second fit
we defined an effective susceptibility (\( \chi_{\text{eff}} \)) where,

\[
\chi_{\text{eff}} = (\chi^{-1} - \chi^{-1})^{-1}. \tag{4}
\]

\( \chi_{\text{eff}} \) plotted versus temperature on logarithmic scales (Fig. 5), indicates a linear
relationship. The best fit line for this equation yielded,

\[
\log \chi_{\text{eff}} = 0.22 - 1.61 \log T. \tag{5}
\]

Fig. 5 shows that there is strong adherence to this relation from 2.5-9 K. Finally, we fit
our data to the commonly used

\[
\chi = \chi_0 + AT^\alpha. \tag{6}
\]
A suitable fit was obtained for $\chi_0 = 0.069 \text{ emu} \cdot \text{mol}^{-1}$, $A = 0.005 \text{ emu} \cdot \text{mol} \cdot \text{K}^{-1}$, and $\alpha = 1$, thus illustrating that the behavior between 2 and 7 K is linear.

A plot of resistivity as a function of temperature (Fig. 7) demonstrates that the data fit well to the curve,

$$\rho = 22.30 + 0.339T^{1.619} \quad (7)$$

over the entire temperature range of 0.33-10.46 K. However, as illustrated in Fig. 8, the fit for $\rho \sim T^{5/3}$ is only slightly worse. Although closer examination of Figs. 7 and 8 reveals that scattering is evident below about 1.5 K, this result of $\rho \sim T^{5/3}$ verifies previous work, such as that by Havela et al. [5], and motivates an interesting point that will be discussed shortly.

**Discussion and Conclusions**

Based on the x-ray diffraction data (Fig. 1), as well as the lattice parameters, we concluded that the UCoAl sample we made was of the correct crystal structure. As shown in Fig. 2, a metamagnetic transition takes place in a critical field of about 6500 gauss (0.65 T). It has been speculated that at the critical field a first-order transition is induced, causing UCoAl to go from a paramagnetic ground state to a ferromagnetic state [8-11]. Whilst it is not exactly known what causes this transition, some ideas have been proposed. Due to some remarkable parallels with previously studied $3d$ itinerant metamagnets (i.e., YCo$_2$), it has been hypothesized that the transition in UCoAl is a $5f$ itinerant metamagnetic transition [10,12-13]. Thus, in low magnetic fields UCoAl appears to have a non-magnetic ground state; however, in fields a bit higher than the
critical field, a magnetic moment of 0.3 $\mu_B$ is induced on the uranium atoms and the compound enters a ferromagnetic state [7].

Considering the susceptibility as a function of temperature (Fig. 3), we see that the $H = 1.5$ T curve follows a $1/T$ dependence at higher temperatures, while more complex behavior occurs at lower temperatures. This behavior is characteristic of ferromagnetism and is what we would expect from the measurements obtained in a field of 1.5 T [14]. As illustrated by the $M$ versus $H$ curve (Fig. 2), at this field UCoAl is ferromagnetic. More importantly, one can see that the behavior of $\chi$ in the low temperature domain, for both 0.1 and 1.5 T, is not constant (i.e., not temperature independent), but rather the behavior appears to have some sort of temperature dependence. This is a key characteristic of non-Fermi liquids as previously noted. While a variety of temperature dependencies exist for $\chi$, we chose to look at a few more common fits, the results of which, for 0.1 T, are indicated in the previous section (see [3] for commonly used equations).

Finally, let us examine the low temperature electrical resistivity measurements. As noted prior, non-Fermi liquid resistivity has temperature dependencies that are different from the $\rho \sim T^2$ that is predicted by Landau's Fermi liquid theory. As shown in Fig. 7, one sees that the $\rho$ versus $T$ plot in fact fits well to $\rho \sim T^{1.619}$, not the $\rho \sim T^2$ predicted by Landau's theory. However, the curve also adheres closely to $\rho \sim T^{5/3}$ for about a decade of temperature, as illustrated in Fig. 8. Although the resistivity curve was extended to a lower temperature than previously shown [5], some scattering is still present below 1.5 K. More experimentation must be performed in order to more accurately determine the low-temperature electrical resistivity.
The $T^{5/3}$ temperature dependency is an interesting issue in itself. This proportionality is precisely what is predicted by the self consistent renormalization model for a three-dimensional ferromagnet [15]. In this model an attempt to explain the behavior of itinerant magnetic systems is made by analyzing spin fluctuations in the vicinity of magnetic phase transitions [3]. However, other predictions made by this theory do not hold true. It predicts that $\chi \sim T^{-4/3}$, a form that is nowhere near describing our data for UCoAl. Clearly, more studies must be performed and/or the theory may need refinement.

After analyzing the data, we can conditionally accept UCoAl as being a non-Fermi liquid until other studies prove differently. These findings, along with previous studies, have provided evidence that UCoAl is one of a handful of compounds that exhibit non-Fermi liquid behavior in their natural state, without the aid of doping, pressure, or an applied magnetic field.

One cannot stress enough the importance of lower temperature work. Experimentation down to millikevin (or even lower) temperatures may reveal that Fermi-liquid behavior is restored (i.e., CeNi2Ge2, see [3]) or it may result in new temperature dependencies being found. Other future experimentation may include the effects of doping, pressure, or field on UCoAl's non-Fermi liquid behavior.

We will be taking specific heat data shortly in the future.

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References


Table

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<th>Author</th>
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<tr>
<td>Mushnikov et al. [7]</td>
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<tr>
<td>This work</td>
<td>a = 668.0, c = 396.9</td>
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Figures

Fig. 1: Theoretical and experimental x-ray diffraction data

Fig. 2: Magnetization versus applied magnetic field for UCoAI at $T = 2.0$ K
Fig. 3: Magnetic susceptibility measured as a function of temperature for fields of 0.1 and 1.5 T.

Fig. 4: Low-temperature fit of $\chi^{-1} = \chi_{s}^{-1} + AT^\sigma$.
Fig. 5: Fit of $\chi_{\text{eff}}$ vs. temperature for UCoAl

$\log (\chi_{\text{eff}}) = 0.22 - 1.61 \log (T)$
$(2.5 \text{ K} < T < 9.0 \text{ K})$
$1/\chi_0 = 14.8$

Fig. 6: Low-temperature fit of $\chi = \chi_0 + AT^2$

$\chi = 0.069 - 0.005T$
$2K < T < 7K$
Fig. 7: Best-fit curve for electrical resistivity versus temperature

\[ \rho = 22.3 + 0.34 \ T^{1.19} \]
\[ 0.33K < T < 10.46K \]

Fig. 8: Low-temperature fit of \( \rho \sim T^{5/3} \) as predicted by self-consistent renormalization theory

\[ \rho = 22.4 + 0.30 \ T^{5/3} \]
\[ 0.33K < T < 10.46K \]