Cerium and rhodium substitution in the antiferromagnetic superconductor cerium rhodium indium(5)

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Ce AND Rh SITE SUBSTITUTION IN THE ANTIFERROMAGNETIC
SUPERCONDUCTOR CeRhIn₅

by

Brian Edward Light

Bachelor of Science
University of Nevada, Las Vegas
1999

A thesis submitted in partial fulfillment
of the requirements for the

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ABSTRACT

Ce and Rh site substitution in the antiferromagnetic superconductor CeRhIn₅

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Among heavy fermion materials, a relationship between unconventional superconductivity and magnetism has been observed. The dependence of the magnetic transitions of CeₓLa₁₋ₓRhₓIr₁₋ᵧIn₅ with respect to temperature and magnetic field applied along the tetragonal c and a crystal axes is examined using heat capacity measurements. The magnetic phase diagram of CeRhₓIr₁₋ᵧIn₅ for the applied transitions in the a direction shows the magnetic structure of CeRhIn₅ is unchanged by Ir doping. Doping of Ir in small concentrations is shown to increase the antiferromagnetic transition temperature Tₐ. La doping or higher concentration of Ir is shown to lower Tₐ. This shows the RKKY interaction, which leads to long range order in CeRhIn₅, is of the same magnitude as the Kondo effect. The high temperature tail of the nuclear Schottky peak is seen in all of the data and can be accurately separated to identify the Ce f-electron contribution to the magnetic heat capacity.
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CHAPTER 1

INTRODUCTION

In the simple free electron model, electrons do not interact with each other. This is known as a free electron gas. In real metals, conduction electrons interact with each other and other electrons yet still behave as an electron gas. The same model can be used except the conduction electrons have a different effective mass due to electron-electron interactions. This modified electron gas is called a Fermi liquid. Heavy fermions are metals in which this effective mass is unusually high, up to several thousand times the normal electron mass. This implies that the electron-electron interactions are very strong.

These interactions can be between the conduction electrons and a local magnetic moment that leads to a superconducting ground state. In a superconductor, electrons that typically repel each other attract and form superconducting pairs known as Cooper pairs. This pairing is mediated by some interaction. Superconductivity that is not mediated by phonons is known as unconventional superconductivity. Among heavy fermion materials, magnetically mediated unconventional superconductivity has been observed.

One of these compounds, CeRhIn$_3$ is the subject of this thesis. The dependence of the magnetic transitions with respect to temperature, magnetic field applied along different crystal directions, and doping using heat capacity measurements allows the determination of the magnetic interaction strength in these complicated materials. Precisely determining the changes of magnetic properties with different variables (temperature,
magnetic field, chemical doping, and pressure) gives insight into favorable conditions for unconventional superconductivity.

Previous heat capacity studies have revealed a quasi-2D electronic and magnetic structure in CeRhIn₅ [4][12]. In this thesis, heat capacity measurements on CeRhIn₅, CeₓLa₁₋ₓRhIn₅, and CeRhₓIr₁₋𝑦In₅ with applied fields along the c and a axis are taken and examined. The heat capacity is measured using a standard thermal relaxation method. Field induced transitions in the a direction can be seen in numerous samples. The phase diagram for CeRhₓIr₁₋𝑦In₅ for these transitions is mapped out and compared to the phase diagram of CeRhIn₅ as part of this study.
CHAPTER 2

THEORETICAL BACKGROUND

Structure of CeRhIn₅

In several Ce based compounds, heavy fermion superconductivity appears as the zero-temperature magnetic-nonmagnetic boundary is approached by applied pressure or chemical doping. Experiments and calculations show that superconductivity is unconventional and is mediated by spin fluctuations that are present near this boundary [8]. Calculations also show that the superconducting transition is optimized for two dimensional (2D) magnetic correlations [9]. In many cases, superconductivity develops out of a state in which the electrical resistivity increases quasi linearly with temperature instead of the expected $T^2$ dependence predicted of a Landau-Fermi liquid [5]. As outlined below, CeRhIn₅ is studied in detail because it is a quasi-2D heavy fermion superconductor.

CeRhIn₅ forms a primitive tetragonal HoCoGa₅ structure type according to X-ray diffraction measurements. This structure can be thought of as alternating layers of CeIn₃ and RhIn₂ stacked along the $c$ axis and is illustrated in Figure 1. This quasi-2D structure is thought to be responsible for the higher than typical magnetically mediated superconducting transition temperature of $T_c = 2.1$ K under pressures on the order of 16 kbar. The transition is a weakly first order transition from the antiferromagnetically ordered state that is present below 3.8 K at ambient pressure. This is unlike the smooth
evolution of $T_N$ to zero reported for other Ce-based heavy fermion antiferromagnetic superconductors.

$\text{CeRhIn}_5$ can be compared to $\text{CeIn}_3$ which is an antiferromagnet at ambient pressure ordering at 10 K, and a superconducting transition at 25 kbar at 100 Mk. It has previously been hypothesized that this is a demonstration of an increase in superconducting temperatures (from 100Mk to 2.1K) due to the reduced dimensionality (from 3D $\text{CeIn}_3$ to quasi-2D $\text{CeRhIn}_5$). The structure of $\text{Ce}_2\text{RhIn}_8$ which consist of alternating two layers of $\text{CeIn}_3$ and one of $\text{RhIn}_2$ is also shown in Figure 1 on the right. While not studied here, $\text{Ce}_2\text{RhIn}_8$ has been studied in detail because the dimensionality of the magnetic interactions is between $\text{CeIn}_3$ and $\text{CeRhIn}_5$.

The magnetically ordered state of $\text{CeRhIn}_5$ is a spiral spin structure with the $\text{CeIn}_3$ layers ordering antiferromagnetically. The direction of the ordered moments in the plane spirals along the c axis rotating 107 degrees per layer of $\text{CeIn}_3$. The spiral structure can be seen in Figure 2. Note that the period is incommensurate with the lattice. The spin structure was first determined by neutron scattering [2]. This magnetic structure is clearly quasi-2D.

**Modeling Heat Capacity**

The system studied is similar to both a two level Schottky system and a spin $1/2$ Ising model. However, there are some large differences. Both of these models will be discussed in order to understand the system studied here. The total entropy of these two models comes to $R\ln 2$ (5.76 J/mol-K) as with any two state system. Here $R$ is the universal gas constant.
Atomic Ce has a moment $j = 5/2$ ($s = 1/2, l = 3$). Because of crystal fields, the moment in CeRhIn$_5$ at low temperatures is $j = 1/2$ ($s = 1/2, l = 0$) which leads to a two state system. However as the temperature increases, some of the higher excitations become available. This implies that for the low temperature transitions, $R \ln 2$ of entropy should be observed. However, it may start to rise above this as temperatures increases.

We know from previous work that, below the transition, there is long-range magnetic order in the form of a spin density wave. This is clearly different than a Schottky system where order is not considered. This is also clearly different from the Ising model, which considers long-range interactions where spins can only be in either an up state or a down state. Even with these differences, both systems are valuable simple models that may help in the understanding of the magnetic transitions observed in CeRhIn$_5$.

First, to examine the Schottky model, consider a system of one particle with two possible states with energy $\varepsilon_0 = 0$ and $\varepsilon_1 > 0$ in thermal contact with a reservoir at temperature $T$. We want to find the energy and heat capacity of the system as a function of temperature. The partition function of the particle is

$$Z = e^{-\frac{\varepsilon_0}{T}} + e^{-\frac{\varepsilon_1}{T}} = 1 + e^{-\frac{\varepsilon_1}{T}}$$

(1)

The average energy is

$$U \equiv \langle \varepsilon \rangle = \frac{e^{-\frac{\varepsilon_0}{T}} \varepsilon_0 + e^{-\frac{\varepsilon_1}{T}} \varepsilon_1}{e^{-\frac{\varepsilon_0}{T}} + e^{-\frac{\varepsilon_1}{T}}} = \varepsilon_1 \frac{e^{-\frac{\varepsilon_1}{T}}}{1 + e^{-\frac{\varepsilon_1}{T}}}$$

(2)

The heat capacity $C_V$ of a system at constant volume is defined as

$$C_V \equiv \left( \frac{\partial U}{\partial T} \right)_V$$

(3)

This gives us
\[ C_V = \frac{\partial}{\partial \tau} \left( \frac{\epsilon_1}{e^{\tau} + 1} \right) = \left( \frac{\epsilon_1}{\tau} \right)^2 \frac{e^{\frac{\epsilon_1}{T}}}{\left( e^{\frac{\epsilon_1}{T}} + 1 \right)^2} \]  

(4)

Using conventional units

\[ C_V = k_B \left( \frac{\epsilon_1}{k_B T} \right)^2 \frac{e^{\frac{k_B T}{A\epsilon}}}{\left( 1 + e^{\frac{k_B T}{A\epsilon}} \right)^2} \]  

(5)

As will be seen, utilizing this result to fit the specific heat data gives poor results except in the highly doped cases. Under the higher doping conditions, the good fit can be explained because of long range order is suppressed by random magnetic vacancies in the lattice.

It should be noted here that what we measure is actually the heat capacity at constant pressure, \( C_p \), not the heat capacity at constant volume, \( C_V \). However, for a solid, the change in volume due to temperature or pressure changes is small, and the difference is disregarded here.

The other simplistic model that is similar to the case studied here is the Ising model. This considers a periodic array of spins, each of which can be in one of two states, up or down. It is postulated that there is an energy of interaction between each pair of nearest neighbors due to the quantum mechanical exchange effect, which depends on whether the spins are like or unalike. No direct magnetic dipole interaction is considered.

Let \( \Delta \epsilon = \epsilon_{\uparrow \uparrow} - \epsilon_{\downarrow \downarrow} \). Given this, ferromagnetism is favored if \( \Delta \epsilon > 0 \), and antiferromagnetism if \( \Delta \epsilon < 0 \). So the entropy \( S \) and heat capacity \( C_V \) are functions of \( k_B T/\Delta \epsilon \). When \( k_B T \ll \Delta \epsilon \), all the spins are aligned in their more favorable state and we
have complete long range order. When \( k_B T >> \Delta \epsilon \), all of the spins are completely disordered by thermal fluctuations.

This model however has no analytic solution in a three dimensional lattice. It has been calculated for simple three-dimensional systems numerically. The resulting heat capacity has a similar shape to what we see with little or no doping in CeRhIn$_5$. However, the Ising model ultimately does not match CeRhIn$_5$ and is only useful to point out that the calculation for a two state system with long range order produces a heat capacity similar to what is seen here. So with no analytic model to fit the data and no computation of the system yet available, we are left with using an empirical fit to try to understand the system. Below the transition temperature \( T_N \), the heat capacity \( C_p \) can be modeled approximately by

\[
\frac{C_p}{T} = \gamma_0 + \beta_M T^2 + \beta_M' T^2 \exp\left(\frac{-\Delta E}{k_B T}\right)
\]

(6)

where \( \gamma_0 \) is the zero-temperature electronic term, \( \beta_M T^2 \), is the standard antiferromagnetic magnon term, and the last term, \( \beta_M' \exp\left(\frac{-\Delta E}{k_B T}\right) \), is an activated antiferromagnetic magnon term. This activated antiferromagnetic magnon term has been used before in other Ce and U compounds. It comes from an antiferromagnetic spin density wave with a gap in the excitation spectrum due to anisotropy. This is consistent with previous Fermi surface measurements that show a gap in the electronic structure [3].

After subtracting off \( C_p/T \) of LaRhIn$_5$ from CeRhIn$_5$, the magnetic heat capacity \( C_m \) can be observed. This makes the assumption that the specific heat of the lattice is
identical between the two compounds. This is reasonable since La and Ce have identical electronic structures except for the f-electron in Ce. The lattice constants for Ce$_x$La$_{1-x}$RhIn$_5$ change less than 0.6% from $x = 0$ to $x = 1$ [11]. This means that the Debye temperature, $\theta_D \propto \frac{1}{V^{1/3}}$, where $V$ is the volume, changes very little. Since the f-electron is tightly bound to the nucleus, the effect on the lattice is minimal.

Adding on the nuclear term gives us

$$\frac{C_n}{T} = \frac{A}{T^3} + \gamma_0 + \beta_M T^2 + \beta_M' T^2 \exp\left(\frac{-\Delta E}{k_B T}\right)$$

(7)

The nuclear term comes from Zeeman splitting due to nuclei with nonzero spin. This occurs at the In sites of CeRhIn$_5$ and is known as the “nuclear Schottky specific heat.” At a few Tesla, this peak has its maximum in the millikelvin region. The high temperature tail of this peak, or that of any Schottky anomaly, rapidly approaches an $A/T^2$ law. The main problem with this empirical fit is that the terms can vary dependent upon where the chosen stopping point of the data lies. For the approximation to be valid, one has to be far enough away from the peak. Previous work has typically chosen approximately 80% of $T_N$ to be far enough away [4]. For consistency, this study will chose 80% of $T_N$ as well.

Spin Interactions

To understand the magnetic properties of these materials, the most obvious start would be that of the interactions between the spins. The simplest type of interaction would be the magnetic exchange and is usually written in the form of a spin Hamiltonian as $\mathcal{H}_J = -J_i S_i S_j$, which couples the spins at lattice site $i$ and $j$. Direct exchange involves
an overlap of electronic wave functions from the two sites and the Coulomb electrostatic repulsion. Pauli exclusion keeps the electrons with parallel spin away from each other thereby reducing the Coulomb repulsion. The energy difference between parallel and anti-parallel configurations is the exchange energy. In the simplest case such as the hydrogen molecule, an anti-parallel configuration is favored. However, since the wave functions of $f$ electrons between nearest neighbors only slightly overlap, the $J_{ij}$ obtained from the overlap integral is far too small to provide a complete explanation for the coupling in these materials [10].

In metallic alloys that contain $f$-electrons the conduction electrons lead to stronger and longer-range indirect-exchange interactions. This is the now famous Ruderman, Kittel, Kasuya, Yosida (RKKY) interaction with Hamiltonian $\mathcal{H}_{ij} = -J_S \mathbf{S}_i \cdot \mathbf{S}_j$. Placing a local moment (spin $S_i$) into a sea of conduction electrons with itinerant spin $S_I$ causes a damped oscillation in the susceptibility of the conduction electrons, and thereby a coupling between spins ($S_i$ and $S_j$) according to

$$J(r) = 6\pi Z J^2 N(E_F) \left[ \frac{\sin(2k_F r)}{(2k_F r)^2} - \frac{\cos(2k_F r)}{(2k_F r)^3} \right]$$

(8)

where $Z$ is the number of conduction electrons per atom, $J$ is the s-d exchange constant, $N(E_F)$ is the density of states at the Fermi level, $k_F$ is the Fermi momentum, and $r$ is the distance between the two local moments. From this we get that the energy can be characterized as

$$T_{RKKY} \propto J^2 N(E_F)$$

(9)

where below $T_{RKKY}$ long range order exists.
From Equation 8 it is clear that at large distances \( J(r) \propto \frac{1}{r^5} \), which is sufficiently long range so that it can effectively reach a number of nearest neighbor sites. Adding a second local moment will create another RKKY polarization and the two conduction-electron-mediated polarizations will overlap in such a way as to establish a parallel or anti-parallel alignment of the two spins.

The Kondo effect describes the condition when non-interacting or very dilute magnetic moments are located in a non-magnetic medium. This condition is actually difficult to realize due to the RKKY interaction that creates a situation where there is always at least some weak interaction even between distant moments. So if the RKKY interaction is small, then the only remaining interaction is between the local moment and the conduction-electron spin \( s \). It can be described by the \( s-d \) or \( s-f \) exchange Hamiltonian, \( H_y = -J S \cdot s \). At high temperatures the impurities behave like free paramagnetic moments. Below a characteristic temperature known as the Kondo temperature \( T_K \), the isolated impurity becomes non-magnetic due to its interaction with the conduction electrons. For \( T < T_K \) there is a gradual loss of local moment as the conduction electrons begin to form a surrounding cloud of oppositely polarized spin. This formation is not a phase transition, but a slow transformation on a logarithmic temperature scale. The energy of this is characterized by

\[
T_K \propto \exp \left( -\frac{1}{J_K N(E_F)} \right)
\]  

(10)

Now imagine the situation in CeRhIn\(_5\) where the Kondo effect and RKKY interaction are of approximately the same magnitude. It is unclear what may happen if the magnetic coupling constant is changed slightly. For the RKKY interaction, an increase in the
magnetic coupling constant increases the energy and therefore characteristic temperature of transition. However the Kondo effect would tend to broaden and decrease the transition with an increase in the magnetic coupling constant. These competing interactions are shown in Figure 3. We are therefore left with the need to experimentally test this by changing the magnetic coupling constant.

Effects of Doping

By doping La and Ir into CeRhIn$_5$, the magnetic interactions are altered. Doping Ir will increase the magnetic coupling constant, $J$, while La will decrease it. In the case of Ir, the magnetic coupling constant $J$ increases since the wave function of Ir extends farther than that of Rh. The conduction band mediates the interactions between two Ce f-electrons as discussed in the RKKY interaction and Kondo effect. Therefore, if there is a stronger overlap of wave functions with Ir, then the Ce f-electrons will have a stronger coupling. Replacing Ce atoms with La atoms not only removes a magnetic moment thereby introducing disorder, it also decreases the magnetic coupling constant. For the same reasons with Ir, doping La will therefore decrease the coupling strength between the Ce f-electrons.

From previous work it is known that Ir doping will eventually lower $T_N$ as seen in Figure 4 which implies the Kondo effect is dominant. This study determines the effect of very small amounts of Ir doping. One of the primary purposes for this study is to determine the effects on the magnetic structure by changing the magnetic coupling constant.
The disorder caused by doping La into CeRhIn$_5$ should also have some noticeable effects. Any transitions will be broader as the local ordering temperature for the Ce f-electrons will vary. Also, the onset of long-range order has to completely disappear if sufficient La substitution occurs.

Other work has shown that doping of La does not change the magnetic structure of CeRhIn$_5$ [1]. This work will determine if doping of Ir in small amounts changes the magnetic structure of CeRhIn$_5$ by creating a phase diagram of magnetic field versus temperature for CeRh$_{0.9}$Ir$_{0.1}$In$_5$. This is critical to determine so that conclusions made about the effects of changing the magnetic coupling are valid. It must be determined whether Ir changes the magnetic structure or only changes the magnetic coupling and corresponding energies.
CHAPTER 3

EXPERIMENTAL TECHNIQUE

Single crystals of CeRhIn$_5$, Ce$_2$RhIn$_8$, Ce$_x$La$_{1-x}$RhIn$_5$, and CeRh$_2$Ir$_{1-y}$In$_3$ were grown in an indium flux as described elsewhere [6]. These crystals where obtained in collaboration with Los Alamos National Laboratories.

Heat capacities were measured using a Quantum Design Physical Properties Measurement System (PPMS). The system uses a standard relaxation technique. A sample is placed upon a small alumina stage with a heater and thermometer attached suspended on gold-palladium alloy wires. Good thermal contact between the sample and stage is maintained by the use of thermal grease. A heat pulse is generated in the heater and the temperature response is measured. This response is fitted to a theoretical model which yields the heat capacity. A complete description of this model can be found elsewhere [7]. A mounting with the heat capacity measurement stage is placed on a He-3 probe described below.

Figures 5 – 7 show diagrams of the experimental arrangement. These figures were taken from the PPMS manual. Temperature is controlled in two main steps. The temperature inside the sample space is controlled by several heaters, an impedance tube from the helium bath and a vacuum pump.

The impedance tube when open allows He-4 from the liquid helium reservoir into the cooling annulus. By warming the impedance tube above 4.18 K, the boiling point of He-
4 at atmospheric pressure, a gas bubble is formed inside the tube thereby closing it. To reach temperatures between 400 K and 4.18 K, all that is required is allowing He-4 into the cooling annulus or activating the heaters in the sample chamber. Below 4.18 K, a vacuum pump lowers the pressure inside the cooling annulus. This decreases the vapor pressure of He-4 and therefore the boiling point temperature. This method is known as evaporative cooling. The limiting factor with this method of cooling is the effectiveness of the pumps. As the pressure gets lower, the pumps efficiency declines. This places an ultimate limit depending on the pumps as to what temperature can be reached with a particular fluid. This leads to the next stage of cooling.

Inside the sample space a He-3 probe is situated where the temperature can be lowered further. He-3 has a boiling point of 3.20 K. The He-4 cooling process is used to condense He-3. This allows for the same process as with He-4 to achieve lower temperatures. The primary difference is that the He-3 system is much smaller and is closed to collect all the He-3 after it is pumped. This is due to the prohibitive cost of He-3. Using He-3, a temperature of about 0.35 K can be reached.

Crystal orientation could be determined visually since relatively large single crystals were used. Only in the case of Ce$_{0.5}$La$_{0.5}$RhIn$_5$ was this inadequate. An error of 5% in the orientation angle is estimated using this method. The error is the most significant source of error in this measurement since the two different crystal orientations have been shown to behave quite differently under magnetic field.

AC susceptibility and DC magnetization measurements were made on Ce$_{0.5}$La$_{0.5}$RhIn$_5$ to search for magnetic “glassiness” using the PPMS as well. This data did not result in any conclusive findings and has been omitted from this work.
Resistivity measurements under pressure were attempted using a clamp cell. These measurements failed, as the clamp cell did not seal reliably. A preliminary calibration for a manganin wire pressure gage was accomplished after many trials. There were other problems with this arrangement as well. These include an indium surface layer affecting resistance measurements as well as temperature control and accuracy. Eventually this measurement was abandoned due partly to other groups completing and publishing it separately.
CHAPTER 4

RESULTS

The magnetic heat capacities of CeRhIn$_5$, Ce$_x$La$_{1-x}$RhIn$_5$, and CeRhyIr$_{1-y}$In$_5$ are shown in Figures 8 through 18 for values $x = 0.97$, $0.9$, $0.8$, $0.5$ and $y = 0.95$, $0.9$ and fields of $H = 0$, $50$kOe, and $90$kOe. The magnetic heat capacity was determined by subtracting the heat capacity of LaRhIn$_5$ with the assumption that the lattice contributions from LaRhIn$_5$ and CeRhIn$_5$ are very close.

In the case of Ce$_{0.5}$La$_{0.5}$RhIn$_5$, the crystal orientation could not be determined visually. Rotating the crystal and measuring the heat capacity showed little difference. The broadening of the peaks as more La is doped into the crystal makes it more difficult to differentiate between crystal directions. For $x = 0.5$, the data taken is shown on both the a and c direction plots.

Due to the interestingly large entropy that remains in Ce$_{0.5}$La$_{0.5}$RhIn$_5$ at the lowest temperatures measured here, a more complete picture of the heat capacity with respect to magnetic field was taken. This can be seen in Figure 19. As can be seen, when the field is applied, the tail at the lowest temperatures moves higher in temperature to the peak seen. It then appears to converge with the first peak which is apparently unaffected by field. At $90$kOe, the peak appears to be in the form a standard Schottky peak although broader and probably two overlapping peaks.
Figure 20 shows the phase diagram made for CeRh$_{0.9}$Ir$_{0.1}$In$_5$ with respect to magnetic field and temperature. This was constructed to compare it to a previous phase diagram of CeRhIn$_5$ which can be found in the upper right corner of Figure 20. Clearly, even with the shift in $T_N$, the magnetic structure of CeRh$_{0.9}$Ir$_{0.1}$In$_5$ is unchanged from CeRhIn$_5$. The magnetic structure in regions I and II is a spin density wave that is incommensurate with the lattice where region II has a larger magnetic moment on each Ce atom. Region III corresponds to a spin-density wave that is commensurate with the lattice [15].

A sampling of the data used to construct this is shown in Figure 21. Three peaks can clearly been seen at around 20kOe. Two of the peaks apparently converge into one at higher fields.

AC susceptibility and DC magnetization measurements were also attempted on Ce$_{0.5}$La$_{0.5}$RhIn$_5$ to further understand its electronic properties. Unfortunately, there were several experimental difficulties. The problem with the AC technique used here was with eddy currents. Also as mentioned the crystals were grown in an Indium flux. This can leave some metallic indium impurities. Although not sufficient to affect a bulk measurement such as heat capacity, any surface layer would drastically affect susceptibility. The superconducting transition of indium was seen, which confirmed that these impurities were present.
CHAPTER 5

ANALYSIS

Previous zero magnetic field beat capacity studies on CeRhIn₅ show that the anisotropic crystal structure leads to a quasi-2D electronic and magnetic structure. Further measurements of CeRhIn₅ and Ce₂RhIn₈ with applied fields along the c axis show that both systems behave like typical heavy fermion compounds as both \( T_N \) and \( \gamma_0 \) decrease as field is increased. Completely different behavior is seen when the field is directed along the a axis. Here \( T_N \) is found to increase and numerous field induced transitions, both of first and second order are observed. These transitions correspond to magnetic field-induced changes in the magnetic structure.

The first question that can be answered is whether the total magnetic entropy of the compounds studied approaches our theoretical value of \( R \ln 2 \) (5.76 J/mol-K). The magnetic entropy of the each sample and field was calculated by integrating \( C_m/T \) with respect to \( T \). These integrations are shown together in Figure 22. They are not shown separately due to the lack of difference among most of the samples. The only sample that is clearly different is Ce₀.₅La₀.₅RhIn₅. These are the red lines that fall below \( R \ln 2 \). This is either due to a large zero temperature entropy or that a substantial part of the peak is at a lower temperature than measured. It is inconclusive which this is without measuring at a lower temperature. However, since the entropy approaches approximately \( R \ln 2 \) by 20 K for all of the samples, this leads to the conclusion that using some two state or similar
model is valid. The continued trend above RhIn2 is either, as mentioned before, the onset of higher excitations being activated or a difference in the lattice between LaRhIn5 and Ce\textsubscript{x}La\textsubscript{1-x}Rh\textsubscript{y}Ir\textsubscript{1-y}In\textsubscript{5}.

In all of the data collected, except for Ce\textsubscript{0.5}La\textsubscript{0.5}RhIn\textsubscript{5}, the nuclear term can clearly be seen to be separate from the other peaks. The nuclear term can be seen as an upturn in the heat capacity at the lowest temperatures and is most notable at a magnetic field of 90 kOe in Figures 10, 12, and 18. This allows the nuclear term to be fitted using the \( A/T^2 \) law and subtracted from Ce\textsubscript{0.5}La\textsubscript{0.5}RhIn\textsubscript{5}. This subtraction is valid since the internal fields the nuclear spins of indium see should be independent of the difference in the electronic structure of CeRhIn\textsubscript{5} and LaRhIn\textsubscript{5}. Figure 19 shows Ce\textsubscript{0.5}La\textsubscript{0.5}RhIn\textsubscript{5} with the nuclear term subtracted. The fit of the nuclear terms can be seen in the 'a' column of Table 1. Together they were fit to

\[
a = c^2 (H_0 + H)^2anumber{11}

where \( H_0 \) is the internal magnetic field, \( H \) is the applied field, and \( c \) is a constant. The fit can be seen in Figure 23 where \( c = 0.0723 \pm 0.0035 \frac{J}{mol \cdot kOe} \) and \( H_0 = 22.2 \pm 4.5 \text{kOe} \).

This makes the approximation that \( B = H \) inside material. In order to show the fit is valid for different doping, the constants are plotted as a function of composition in Figures 24 and 25.

The data is fitted using the empirical fit mentioned previously

\[
\frac{C_m}{T} = \frac{A}{T^3} + \gamma_0 + \beta_M T^2 + \beta_M' T^2 \exp \left( -\frac{\Delta E}{k_B T} \right)\]

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The cut off point chosen here was approximately 80% of $T_N$. These fits can be seen in Table 1. The error shown is twice the standard deviation or 5% which ever is greater. Note that the energy term has a large error that should normally be well known due to the precision and width of the peak using a Schottky or Ising model. This implies some definite limitations of this empirical fit. Ultimately, this method is unsatisfying when trying to distinguish the fitted terms for the different material consistencies. Only the nuclear Schottky term fits very well. No real trend can be determined for $\beta_m$ or $\beta_m'$. The stated error is probably too low since residual data from the fits show that the error is mostly systematic in nature. The La 20% data did not even numerically converge, so no fit is shown. Notice that the La 10% data errors are very large implying that the fit is simply inappropriate. $T_N$ has relatively low error since this is determined by the peak positions not the fit. A more rigorous fit to the entire curve would be ideal. Similar systems have been modeled using Mote Carlo simulations, however that is outside the scope of this study.

Even with these limitations, several insights can be gained from the fitted data. $T_N$ clearly decreases with increased doping of La while $T_N$ increases for doping of Ir. This is contrary to what has been found previously for more heavily doped CeRh$_{y}$Ir$_{1-y}$In$_5$. This implies that CeRhIn$_5$ is near the equilibrium point for the Kondo effect and RKKY interaction. Slight increases in the magnetic coupling will increase the ordering temperature, however as magnetic coupling becomes too large, the ordering temperature will eventually fall.

La doping is consistent with this. Since it has now been determined that the RKKY effect is dominant for CeRhIn$_5$, decreasing the magnetic coupling constant should lower
This can be seen in all of the La doped data. There are still some unanswered questions remaining for Ce$_{0.5}$La$_{0.5}$RhIn$_5$. This particular sample was studied in detail because of its interesting behavior. Although one peak looks to be remarkably Schottky in appearance, it seems to be field independent. A second peak is clearly field dependent. This peak increases in temperature as field increases. At around 30 kOe it merges with the field independent peak. The higher temperature peak is so broad it is uncertain whether this is a transition or not. Comparing the 50\% La to the 20\% La in Figures 8 though 12, the data look very similar coming down from higher temperatures. Then, the 20\% La has a clear transition where the 50\% does not. The original reason for the AC susceptibility measurement was to determine if Ce$_{0.5}$La$_{0.5}$RhIn$_5$ formed a “spin glass.” This was hypothesized due to the apparent high zero temperature entropy. It is still uncertain however.

Ce$_{0.5}$La$_{0.5}$RhIn$_5$ is near the point where order begins to take shape but never achieves full long range order. Other work has shown that antiferromagnetism is completely suppressed by 40\% La [11]. However after field is applied it is possible, although unlikely, that magnetic order is restored. If there is magnetic ordering, it is still unclear what it is. The work presented in this thesis will be used in conjunction with further studies to determine the nature of Ce$_{0.5}$La$_{0.5}$RhIn$_5$.  

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BIBLIOGRAPHY


APPENDIX

TABLES AND FIGURES

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Figure 1: The tetragonal structure of CeRhIn$_5$ and Ce$_2$RhIn$_8$ is shown here in comparison with CeIn$_3$.
Figure 2. Magnetic structure of CeRhIn$_5$. Only Ce sites are shown in the structural unit cell. Moments in the A plane exhibit simple AF order. The disk denotes the moment rotating plane. The dashed line traces the incommensurate spiral along the axis. This figure is taken from Wei Bao et al. Phys. Rev. B 62, R14621 (2000).
Figure 3. Shown here are the characteristic temperature curves for the Kondo effect and RKKY interaction for a generic material. Since the Kondo temperature rises exponentially and the RKKY interaction goes as $T^2$, the Kondo effect will eventually become dominant as the magnetic coupling constant $J$ increases.
Figure 4. Take directly from P. G. Pagliuso et al. Phys. Rev. B 64, 100503 (2001). Temperature-composition phase diagram of CeRh$_{1-x}$Ir$_x$In$_5$. $T_{\text{none}}$ indicates the absence of (additional) phase transitions for $T>350$ mK. This is shown here to illustrate the purpose for the Ir doping done in this study. Notice that as Ir composition increases, $T_N$ goes down. The question that part of this study answers is at very small doping what happens to $T_N$. 
Figure 5. PPMS: Physical Property Measurement System. This is the system used to measure heat capacity and magnetization.
**PPMS Probe**

**Figure 6.** Shown here is the probe that is located inside the PPMS.
Figure 7. The He-3 refrigerator with the calorimeter puck is shown here. Temperatures down to 0.35 K can be reached. The samples are placed on the sample stage and are adhered with thermal grease.
Figure 8. The magnetic heat capacity of CeRhIn₅ and related lanthanum doped compounds is shown here at a zero magnetic field. The magnetic heat capacity is found by subtracting the heat capacity of LaRhIn₅ from the heat capacity of each of the above compounds. Theoretically, the lattice contribution of LaRhIn₅ and CeRhIn₅ should be the same. In other words, the extra f electron in cerium does not effect the lattice heat capacity, only the magnetic component.
Figure 9. The magnetic heat capacity of CeRhIn₅ and related lanthanum doped compounds is shown here at a magnetic field of 50kOe in the a direction of the crystal lattice. All of the measurements taken were from single crystals. Field direction was determined by visually orientating the single crystals in the a, b, and c directions and measuring the heat capacity. Note that the a and b directions are identical due to the symmetry of the lattice and are simply referred to as a. The estimated error for the orientation of the crystal is 5 degrees.
Figure 10. The magnetic heat capacity of CeRhIn$_5$ and related lanthanum doped compounds is shown here at a magnetic field of 90kOe in the a direction of the crystal lattice. All of the measurements taken were from single crystals. Field direction was determined by visually orientating the single crystals in the a, b, and c directions and measuring the heat capacity. Note that the a and b directions are identical due to the symmetry of the lattice and are simply referred to as a. The estimated error for the orientation of the crystal is 5 degrees.
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Figure 12. The magnetic heat capacity of CeRhIn$_5$ and related lanthanum doped compounds is shown here at a magnetic field of 90kOe in the c direction of the crystal lattice. All of the measurements taken were from single crystals. Field direction was determined by visually orientating the single crystals in the a, b, and c directions and measuring the heat capacity. Note that the a and b directions are identical due to the symmetry of the lattice and are simply referred to as a. The estimated error for the orientation of the crystal is 5 degrees.
Figure 13. The magnetic heat capacity of CeRhIn$_5$ and related Iridium doped compounds is shown here at a zero magnetic field. The magnetic heat capacity is found by subtracting the heat capacity of LaRhIn$_5$ from the heat capacity of each of the above compounds. Note that in order to observe a difference between the compounds, a range of 3K to 5K has been selected.
Figure 14. The magnetic heat capacity of CeRhIn₅ and related iridium doped compounds is shown here at a magnetic field of 50kOe in the a direction of the crystal lattice. All of the measurements taken were from single crystals. Field direction was determined by visually orientating the single crystals in the a, b, and c directions and measuring the heat capacity. Note that the a and b directions are identical due to the symmetry of the lattice and are simply referred to as a. The estimated error for the orientation of the crystal is 5 degrees. Again, the temperature range shown is 3K to 5K.
Figure 15. The magnetic heat capacity of CeRhIn$_5$ and related iridium doped compounds is shown here at a magnetic field of 90kOe in the $a$ direction of the crystal lattice. All of the measurements taken were from single crystals. Field direction was determined by visually orientating the single crystals in the $a$, $b$, and $c$ directions and measuring the heat capacity. Note that the $a$ and $b$ directions are identical due to the symmetry of the lattice and are simply referred to as $a$. The estimated error for the orientation of the crystal is 5 degrees.
Figure 16. The magnetic heat capacity of CeRhIn$_5$ and related iridium doped compounds is shown here at a magnetic field of 50kOe in the c direction of the crystal lattice. All of the measurements taken were from single crystals. Field direction was determined by visually orientating the single crystals in the a, b, and c directions and measuring the heat capacity. Note that the a and b directions are identical due to the symmetry of the lattice and are simply referred to as a. The estimated error for the orientation of the crystal is 5 degrees.
Figure 17. The magnetic heat capacity of CeRhIn₅ and related iridium doped compounds is shown here at a magnetic field of 90kOe in the c direction of the crystal lattice. All of the measurements taken were from single crystals. Field direction was determined by visually orientating the single crystals in the a, b, and c directions and measuring the heat capacity. Note that the a and b directions are identical due to the symmetry of the lattice and are simply referred to as a. The estimated error for the orientation of the crystal is 5 degrees.
Figure 18. The magnetic heat capacity of CeRh$_{0.9}$Ir$_{0.1}$In$_5$ is shown here with respect to field in the c direction of the crystal lattice. Clearly $T_N$ moves to lower temperatures as field is applied. This is standard antiferromagnetic behavior. All of the samples that show magnetic ordering behave similarly for fields in the c direction so only one sample is shown.
Figure 19. A more complete study of Ce_{0.5}La_{0.5}RhIn5 was completed in order to understand if the highly doped showed signs of non-fermi liquid behavior. The data shown here is the magnetic heat capacity. The hyperfine term has already been subtracted.
Figure 20. Shown here is a phase diagram for CeRh$_{0.9}$Ir$_{0.1}$In$_5$ with respect to temperature and magnetic field in the a crystal direction. The data points shown are the peak positions of the transitions seen in heat capacity measurements. The two peaks seen in previous figures in the a directions can be seen here at 50kOe and 90kOe. As the field reaches approximately 20kOe, three peaks are easily discernable. Previous work constructed a similar phase diagram for CeRhIn$_5$. A selection of the data used to construct this is shown in Figure 13. In the upper right are the phase diagrams for CeRhIn$_5$ and Ce$_2$RhIn$_6$ from Cornelius et al. Phys. Rev. B 64, 144411 (2001). The magnetic structure in regions I and II is a spin density wave that is incommensurate with the lattice where region II has a larger magnetic moment on each Ce atom. Region III corresponds to a spin-density wave that is commensurate with the lattice.
Figure 21. Shown here is a sample of the data used to create the phase diagram for CeRh$_{0.9}$Ir$_{0.1}$In$_3$ with respect to temperature and magnetic field in the a crystal direction. The peak positions seen here in the heat capacity measurements imply a phase transition. As the field reaches approximately 20kOe, three peaks are easily discernable.
Figure 22. Shown here is the entropy of all of the samples studied. Although they show some variance, all of them approximately reach Rln2 at around 20K. The curves which lie below Rln2 are that of Ce₃La₃RhIn₅. Clearly for Ce₃La₃RhIn₅ there is some missing entropy due to the measurement not going low enough in temperature. This is apparent in figures 1-6 where one can see the data still is not approaching zero for Ce₃La₃RhIn₅ as T goes to 0K.
Figure 23. For all of the data possible to separate the nuclear term from the other contributions to the magnetic heat capacity, the hyperfine constant was collected and fitted to \( \sqrt{A} = c(H_0 + H) \). This fit was used to subtract the nuclear term from Ce\(_{0.5}\)La\(_{0.3}\)RhIn\(_5\).
Figure 24. Shown here is the nuclear Shottky constants for the samples as a function of Ir doping. This is shown in order to make it convincing that the doping composition has little effect on the nuclear Shottky constant.
Figure 25. Shown here is the nuclear Shottky constants for the samples as a function of La doping. This is shown in order to make it convincing that the doping composition has little effect on the nuclear Shottky constant.
Table 1. Shown here is the fit of the magnetic heat capacity to \( C_m / T = \gamma_0 + B_\mu T^2 + B'_\mu T^2 \exp(-E_g/k_B T) \) with varying magnetic fields in different crystal directions of various samples. Duplicate entries are separate measurements on the same conditions to determine reproducibility. The stated error is twice the standard deviation or 5% which ever is larger.
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