

## Doping experiments on CeRhIn<sub>5</sub>

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### Abstract

The Ce<sub>1-x</sub>La<sub>x</sub>RhIn<sub>5</sub> family of compounds are prepared and examined with different La concentrations for the purpose of suppressing antiferromagnetism. The values of x prepared are x = 0, 0.15, and 0.5. The samples were grown in single crystal form using an In flux technique, then characterized by x-ray diffraction, magnetic susceptibility, and specific heat. Magnetic susceptibility data reveal that as x increases, the antiferromagnetism of heavy-fermion CeRhIn<sub>5</sub> is suppressed.

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## 1. Introduction

In past years much research has been conducted on compounds that can be characterized as heavy-fermions. Heavy-fermions are defined as systems that have very large electron effective masses,  $m_e^*$ . The effective mass of an electron can be characterized by how the electron moves through its energy band. If an electron has a low effective mass, the electron tends to move easily through its band. On the other hand, if an electron has a large effective mass, it tends to remain more localized in the system.

The effective mass of an electron can be determined from both magnetic susceptibility and specific heat measurements. The values of these measurements are proportional to the electron effective mass as the temperature approaches absolute zero,

$$m_e^* \propto \gamma \propto \chi \quad (1)$$

where  $\chi$  is magnetic susceptibility, and  $\gamma$  ( $\equiv C/T$  as  $T \rightarrow 0$ ) is the electron contribution to the specific heat (also called the Sommerfeld coefficient). Thus,  $\chi$  and  $\gamma$  are tens to thousands of times larger in heavy-fermions than in simple metals.

The recent discovery of heavy-fermion behavior in  $\text{CeMIn}_5$ , where  $M = \text{Co}, \text{Ir},$  and  $\text{Rh}$ , has stimulated research on these compounds.  $\text{CeCoIn}_5$  and  $\text{CeIrIn}_5$  have been characterized as low-temperature superconductors with large, easily definable Sommerfeld coefficient [1-2]. The Sommerfeld coefficient for  $\text{CeRhIn}_5$ , on the other hand, is far from easily definable.  $\text{CeRhIn}_5$  is an antiferromagnet showing magnetic

order below its Néel temperature,  $T_N$ , of 3.8 K [3]. Because of this order, only an approximate value of  $\gamma = 400 \text{ mJ/mol K}^2$  has been determined [4].

In the past, cerium compounds being doped with lanthanum have brought important insights to heavy-fermion systems. Alver *et al.* [5] have studied electron characteristics in  $\text{CeRhIn}_5$  by doping La onto the Ce site. They performed de Haas-van Alphen (dHvA) measurements and found as the La concentration increases, the electron effective mass monotonically decreases [5]. Our purpose is to further characterize the electronic properties of  $\text{Ce}_{1-x}\text{La}_x\text{RhIn}_5$  through magnetic susceptibility measurements.

## 2. Experimental

### *Samples*

In order to perform the experiments, high purity samples must be prepared. Following prior research [1-5], we grew samples in single crystal form. First, small stoichiometric amounts of high purity elements were cut in order to achieve a total sample mass of approximately one-half of a gram. The masses were measured within an accuracy of 0.01 mg. Our samples contain both cerium and lanthanum. These elements oxidize in the atmosphere within about 20 min. The cerium and lanthanum elements were sealed in a Pyrex tube under a vacuum to prevent oxidation. In order to grow crystals of  $\text{Ce}_{1-x}\text{La}_x\text{RhIn}_5$  out of excess In ('In metal flux technique') cerium was diluted to 3% of the indium moles [6].

After all the elements had been prepared, they were placed in a high temperature furnace inside of a BeO crucible with lid. We heated each sample from room temperature to  $1200^\circ \text{C}$  at a rate of  $75^\circ/\text{hr}$ . The samples maintained the temperature of  $1200^\circ \text{C}$  for two hours in order to provide good mixing. Then, the sample's temperature

was brought down to 300° C at a rate of 10°/hr, during which time the dissolved elements become supersaturated in the In flux and precipitate out as flat platelet crystals. Below 300° C, the crucible was then cooled to room temperature at 75°/hr. The La concentrations of our samples were  $x = 0, 0.15, \text{ and } 0.5$ .

### *X-ray Diffraction*

When crystals form, the lattice structure must be identified. Unwanted second phase compounds may have formed due to impurities, thermodynamics, or calculation errors. An easy way to test the phase make up of a sample is by using x-ray diffraction data to identify any second phase material. With the use of x-ray diffraction (XRD) at the University of Florida's Major Analytical Instrumentation Center (MAIC), we were able to test the phase purity of our samples.

In order to perform XRD, the samples must be prepared on a diffraction slide. We begin by collecting a small portion of our sample approximately 100 mg in size. These pieces are then ground into a fine powder and glued to a microscope slide in a small 1 x 1 cm square. The glue used during this process is a mixture of one part amyl acetate and seven parts collodium. This particular mixture is used because it does not affect the experiment.

The underlying physics of XRD is rather simple. High-energy photons, x-rays, bombard the sample and are diffracted by the crystal structure of the target sample. At certain incident angles, large intensities of x-rays are diffracted from the sample. Using Bragg's law, the sample structure can be identified. Bragg's law states,

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

where  $\lambda$  is the photon wavelength,  $d$  is the distance between atoms in the lattice, and  $\theta$  is the angle of reflection of the photons. After we have taken XRD data, we use this to determine the lattice parameters of our sample. The lattice parameters for a crystal structure are proportional to the atom spacing  $d$  and the Miller indices, denoted by  $(h k l)$ . The particular proportionality of the parameters of a lattice depends on the specific lattice structure. For example, in the tetragonal structure under consideration, the lattice parameters,  $a$  and  $c$ , are

$$\frac{1}{d^2} = \frac{(h^2 + k^2)}{a^2} + \frac{l^2}{c^2}, \quad (3)$$

where  $d$  is the distance between atoms.

### *Magnetic Susceptibility*

A large number of metals and metallic compounds exhibit magnetic properties. They range from paramagnetism to ferromagnetism. The different types of magnetism depend on the electrons in a system. The spins and coupling of electrons determine what kind of magnetism will occur in a substance. For example, if a system's electrons couple together with parallel spins, the substance is said to be ferromagnetic. On the other hand, if the electrons couple anti-parallel to one another, then the compound is said to be antiferromagnetic. CeRhIn<sub>5</sub> is inherently an antiferromagnet.

We measured the magnetic susceptibility of our samples using a Superconducting

Quantum Interference Device (SQUID). Our susceptibility measurements ranged from 1.8 - 300 K under an applied magnetic field of 1000 G. Measurements were taken with the magnetic field parallel and perpendicular to the basal plane of our samples, because the susceptibility of CeRhIn<sub>5</sub> has been shown to be anisotropic [4].

### *Specific Heat*

For simple metals, like gold or copper, the specific heat follows the form

$$C(T) = \gamma T + \beta T^3 \quad (3)$$

where the  $\gamma$  term is the electron contribution, and the  $\beta$  term is the lattice contribution to the specific heat. As can be determined from this formula, the lattice contribution to the specific heat dominates at high values of  $T$ , and the electron contribution dominates at low values of  $T$ . In our experiment, we measured the specific heat from 0.3 - 300 K using the thermal relaxation time constant method [7].

### **3. Results**

X-ray diffraction data of Ce<sub>1-x</sub>La<sub>x</sub>RhIn<sub>5</sub> are displayed in Fig. 1. Some unexplained discrepancies appear between predicted and observed intensity values; however, based on consistent peak positioning, we believe Ce<sub>1-x</sub>La<sub>x</sub>RhIn<sub>5</sub> forms in the expected tetragonal crystal structure.

As shown in Fig. 1, as  $x$  increases, intensity peaks are shifted to the left at higher angles. This is due to the atomic radius of our selected elements: cerium is smaller than

lanthanum. Because of this size difference, the lattice parameters increase, evident as the (2 0 6) peak angle shifts left with increasing lanthanum. From Bragg's law it can be concluded that as the angle decreases, the value for  $d$  increases, hence lattice parameters increase.

In Figs. 2a-b, plots of  $\chi$  vs.  $T$  for each of our samples are displayed.

Antiferromagnetism is evident by the peak at 3.8 K parallel to the basal plane in  $\text{CeRhIn}_5$ . Looking at Fig. 2a, we see for  $x = 0.15$ ,  $\chi$  decreases for low  $T$ . Shifting to Fig. 2b, we see  $\chi$  rise at low  $T$  with increasing La concentration. For  $x = 0.15$ ,  $T_N$  is shifted to a lower temperature, while for  $x = 0.5$ , we see no magnetic order down to 1.8 K. Measurements at the lowest temperatures show a dramatic increase in  $\chi$  for  $x = 0.5$ .

Specific heat analysis is incomplete at this time - data will be presented in an upcoming publication.

#### 4. Conclusions

X-ray diffraction and magnetic susceptibility measurements were used to characterize  $\text{Ce}_{1-x}\text{La}_x\text{RhIn}_5$ . We were successful in suppressing antiferromagnetism by doping  $\text{CeRhIn}_5$  with lanthanum. As  $\gamma$  and  $\chi$  are proportional to  $m_e^*$  and  $dHvA$  measurements support decreasing  $m_e^*$  with increasing La concentration, the behavior of  $\text{Ce}_{0.85}\text{La}_{0.15}\text{RhIn}_5$  follows expectations. The increases in  $\gamma$  and  $\chi$  at low temperatures in  $\text{Ce}_{0.5}\text{La}_{0.5}\text{RhIn}_5$  are unexplained at this point. Further work is planned to explain this phenomenon.

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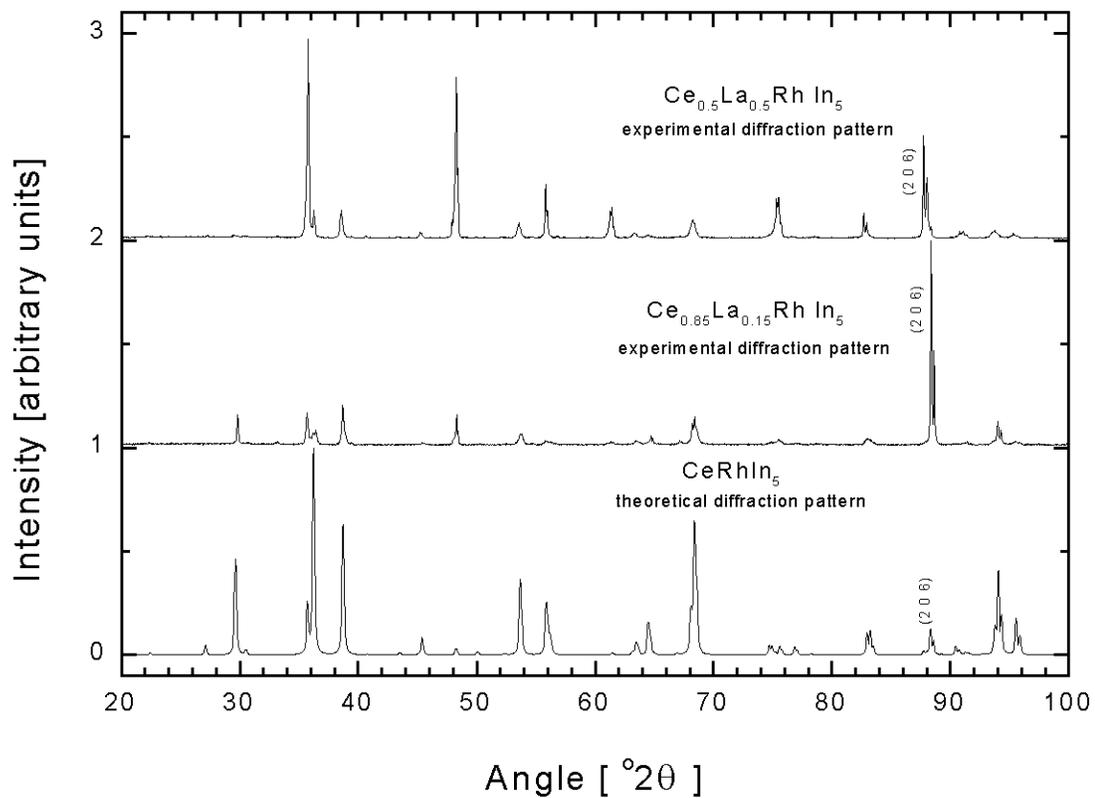


FIGURE 1: X-Ray Diffraction pattern comparison for tetragonal  $\text{Ce}_{1-x}\text{La}_x\text{RhIn}_5$ . Note the (2 0 6) peak shifts to lower angles (lattice parameters increase) as lanthanum concentration increases.

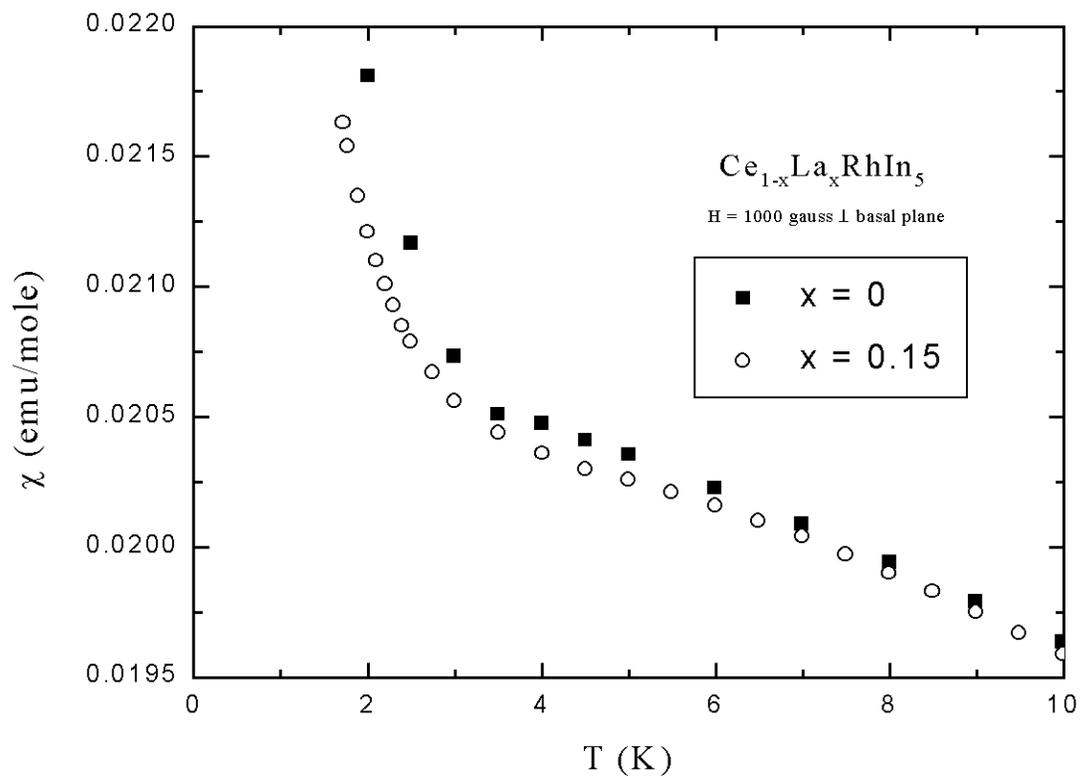


FIGURE 2a: Magnetic susceptibility data for  $\text{Ce}_{1-x}\text{La}_x\text{RhIn}_5$  measured with the applied magnetic field *perpendicular* to the basal plane. Note the small suppression of  $\chi$  with La doping.

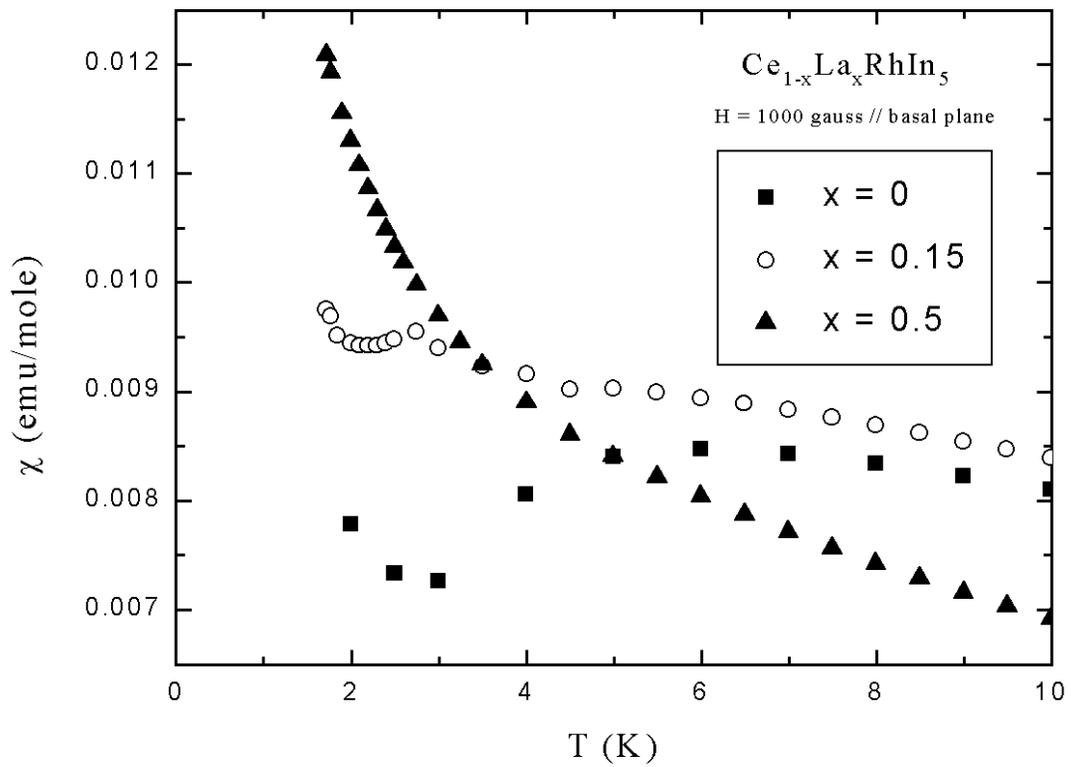


FIGURE 2b: Magnetic susceptibility data for  $\text{Ce}_{1-x}\text{La}_x\text{RhIn}_5$  measured with the applied magnetic field *parallel* to the basal plane. Note the antiferromagnetism transitions in the  $x = 0, 0.15$  samples and the antiferromagnetic suppression in the  $x = 0.5$  sample.