Effects of High Magnetic Field on Phases of Ce

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Abstract

Cerium is a very interesting element. It has three solid state phases, including two, the α and magnetic γ phases, that are isostructural, both having fcc lattice structures. The third, β phase, has a dhcp lattice structure. The $\alpha \rightleftarrows \gamma$ phase change is assumed to be purely electronically driven because it is isostructural. Cerium's single localized $4f$ electron is believed to play a crucial role in this phase transition. By coupling the $4f$ electron with a high magnetic field and observing the effects on the sample's optical conductivity, it is hoped that the phase change and the role of the $4f$ electron, explained in several competing theories, can be better understood. Additionally, understanding the phase changes in cerium and determining the effect of a high magnetic field on its electronic structure could help in understanding the transitions between the six different phases of plutonium.

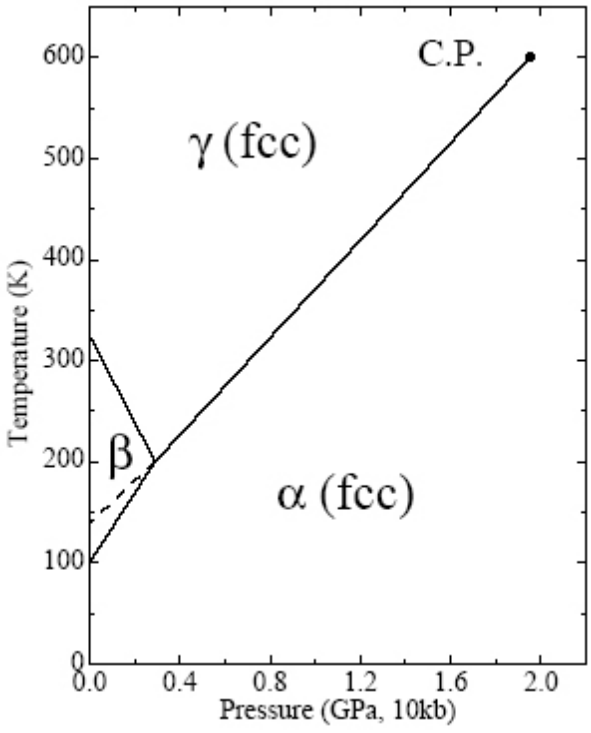

Figure 1: Phase diagram for Ce taken from [1].

1. INTRODUCTION

Cerium is one of the most fascinating elements. Its elemental phases include an antiferromagnetic, a paramagnetic, and a superconducting phase. This is widely ascribed to its single localized $4f$ electron close to the Fermi level, which we hope to couple with a magnetic field. It is the most reactive of all rare earth metals save europium, oxidizing quickly in moist air.

Figure 1 shows the phase diagram for cerium. It has several solid-state phases, exhibiting an isostructural phase transition between the face-centered cubic (fcc) α and magnetic γ states at 0.7GPa and 298K. The α phase has a volume 14-17% smaller than the γ phase. These phases become indistinguishable at a critical point at $\sim$1.9GPa and 600K. The third phase, the β phase, occurrs at low pressures ($<\sim$.3GPa) between the α and γ phases and has a double hexagonal close-packed (dhcp) structure. The electronic structure of this phase bears some similarity to that of the γ phase.

Several optical spectroscopy experiments have been performed on cerium, showing that the α phase is more conductive than the γ phase, while both experience similar trends of diminishing conductivity with increasing frequency [1]. Experimental and theoretical spectra

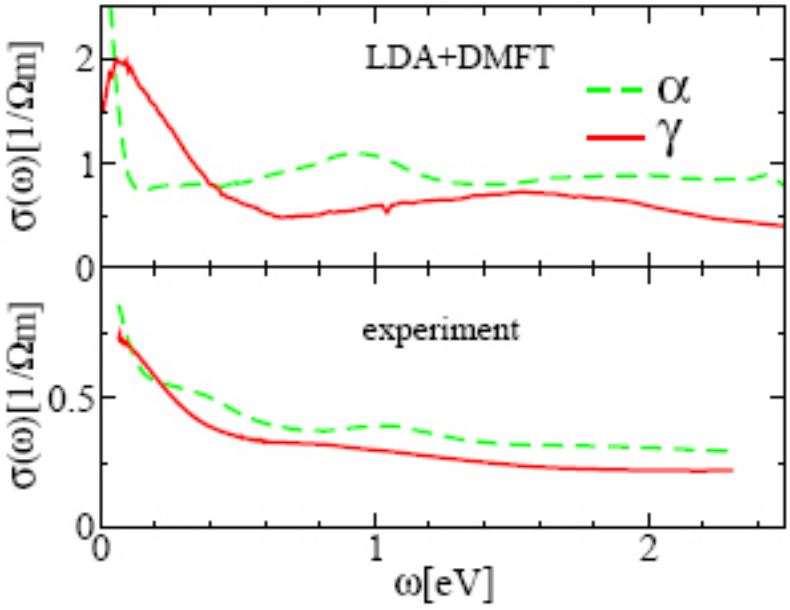

Figure 2: Optical conductivity for α and γ Ce. The top panel shows a theoretical calculation using LDA+DMFT [2] and the bottom shows experimental data [3]

are showin in figure 2.

2. THEORY

Many theories have been proposed to explain the $\alpha \rightleftarrows \gamma$ transition [6, 7, 9, 10]. Since it is isostructural, it is assumed to be purely electronically driven. In most of these theories, cerium's single $4f$ electron plays a crucial role because it extends further from the center of the atom than $4f$ electrons in other lanthanides and has an energy very close to that of the conduction electrons. In addition, the f electron has a very anisotropic orbit which is delocalized in the α phase and localized in the γ phase.

In the α and γ phases, cerium has a fractional valence of 3.67 and 3.06, respectively [5]. The density of states for α and γ cerium are given in figure 3. The band-structure is characterized by a broad d band with a width of $\sim$10eV and a narrow $4f$ band with a width of $\sim$1eV, with the α and γ phases differing mainly in the broadening of the $4f$ band [8]. In the α phase, the f electron is delocalized, while in the γ phase, it is localized.

One theory, the Promotional Model, explains the volume change by asserting that the

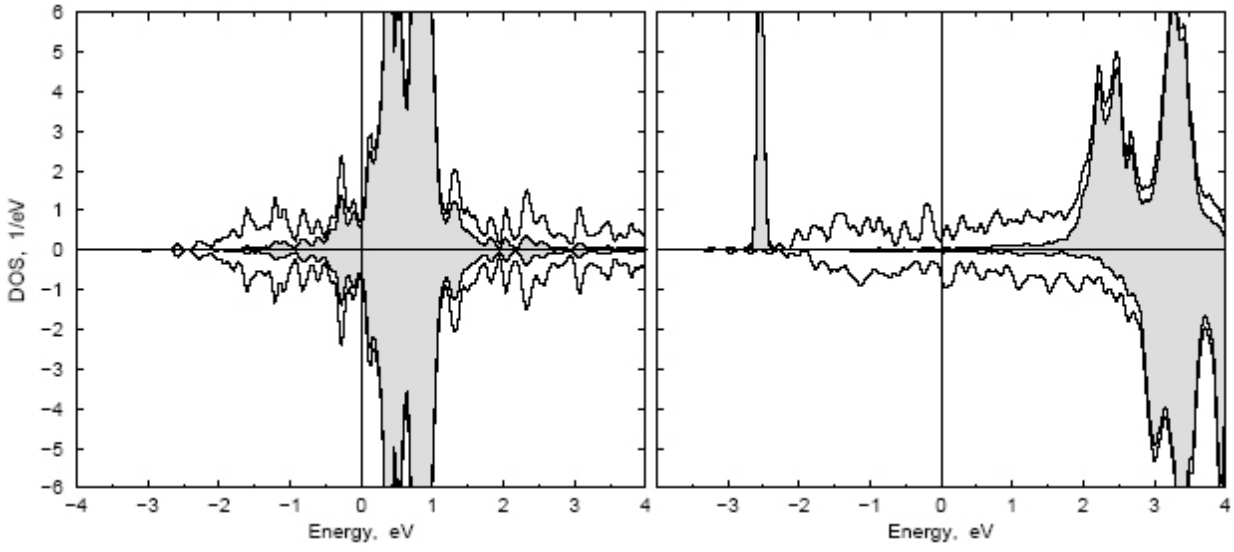

Figure 3: Spin resolved density of states for α Ce on the left and γ Ce on the right. Upper panels show spin up and lower show spin down. Taken from [4].

localized $4f$ level becomes at least partially delocalized and enters the valence band. Calculations performed by A. de Vries [6] show that this leads to a decrease in radius for all of the outer bands. However, recent photo emission experiments show that this is unlikely due to the amount of energy involved. In the γ phase, the $4f$ level lies at $\sim$2eV, not close to the Fermi level. Thus, in order to excite a $4f$ electron to the conduction band, an energy 2 orders of magnitude higher than the elastic work needed to compress γ Ce to α Ce is required.

Another theory, the Mott Transition, proposed by Johansson [7], asserts that an increase in pressure forces several electron bands to merge at the Fermi level. The f electron is itinerant in the α phase but localized in the γ phase, which prevents it from participating in bonding in the γ phase. This is theorized to cause the volume collapse.

A third theory, proposed by Göepert and Meyer, focuses on the two local minima in the $4f$ atomic wave function. Cerium is between similar elements on the periodic table whose $4f$ level is localized in the inner minimum to the right and in the outer minimum to the left. Since cerium is in the middle, it could have a delocalized $4f$ electron which provides less screening of the nucleus and would allow the $6s$ and $5d$ orbitals to collapse in the α phase.

In the Kondo Volume Collapse Model, Allen and Martin show that an increase in pressure can cause the f-orbitals and d-orbitals to hybridize [9, 10]. When this occurs, it is believed

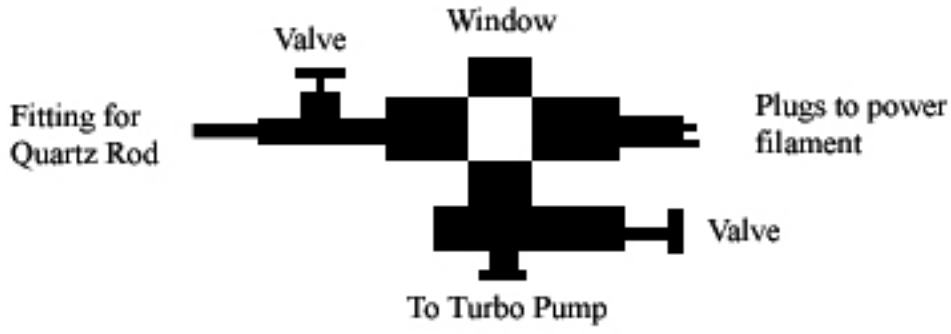

Figure 4: Evaporator

that the f electron spends more time closer to the nucleus.

Placing α Ce in a high magnetic field will induce the Zeeman effect, which splits the atomic energy levels into $2L + 1$ levels. This will produce predictable effects on the sample that are immediately visible through changes in the sample's optical conductivity. It has been shown that application of a magnetic field can hinder a temperature-driven phase change in Ce [11]. Whether the band splitting alone can induce a phase change will provide valuable data for the understanding of phase transitions in cerium.

3. APPARATUS AND PROCEDURE

To perform optical spectroscopy, cerium is evaporated onto the end of a quartz rod. In order to do this, an evaporator was constructed as illustrated in figure 4. Cerium is placed in a basket made of tungsten wire and attached to the electrical feedthrough on the right which has plugs to provide electrical power. A quartz rod is inserted into the fitting on the left. The apparatus is then evacuated and the tungsten filament powered until the cerium evaporates and deposits a thin film on the quartz rod.

In order to illuminate the sample, a white light source and a series of lenses is set up as shown in figure 5. Two aspheric lenses are used to collimate the light from the white light source and placed so that the light rays are essentially parallel in a four inch region where the first plano convex lens will be placed. This lens, with a long focal length (1m), and another plano convex lens, with a very short focal length (.05m), operate like a simple telescope to focus the light on the tip of the quartz rod. After passing through the rod, the light is collected and passed through a spectrometer. A streak camera then records the

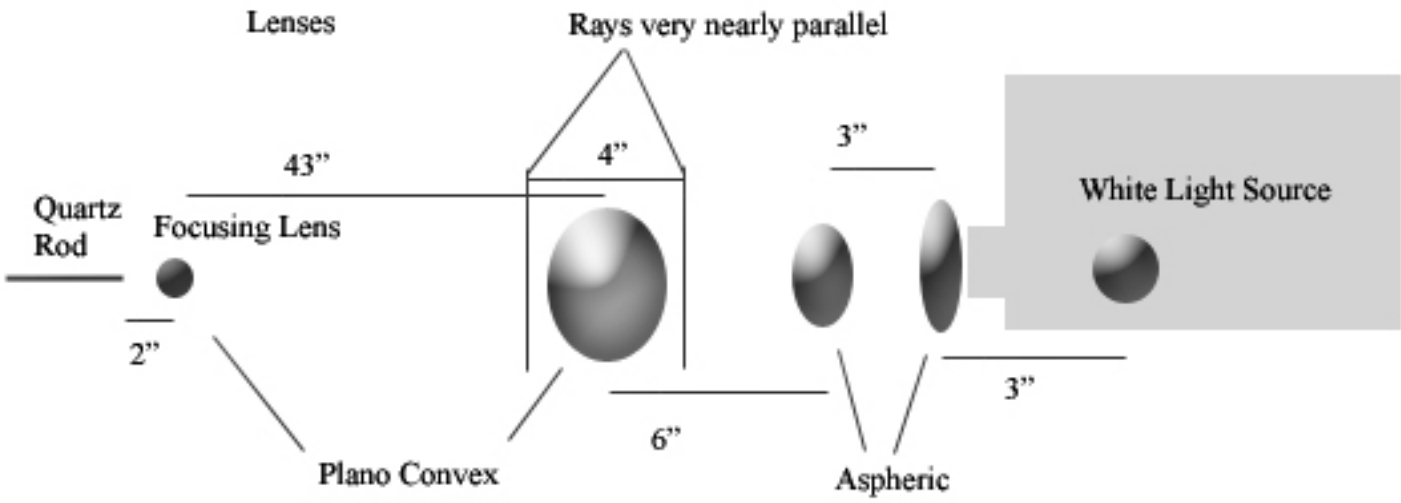

Figure 5: Optics

intensity for the respective energy.

The quartz rod is positioned in such a way that the thin film of cerium is located in the center of the single turn magnet apparatus, shown in figure 6. This will provide magnetic fields in the range of 100-300T in a 6μs (2μs rise, 4μs fall) half-sinusoid pulse, shown in figure 7. It is powered by a capacitor bank of 24μF low-inductance 60kV capacitors, which are fired in parallel to discharge 3.8MA. The single turn itself is made of copper 3mm thick in the shape of a circle 10mm in diameter.

4. CONCLUSION

Currently, the single turn magnet is nearing completion. The evaporator has been built and the optics are set up.

This experiment will indicate whether a magnetic field is sufficient to induce a phase transition in cerium. By tracking the optical conductivity, it will show either a correlation between band splitting and a phase transition or that the transition is independent of band splitting.

Plutonium is similar to cerium in that it exhibits several solid state phases. Nevertheless,

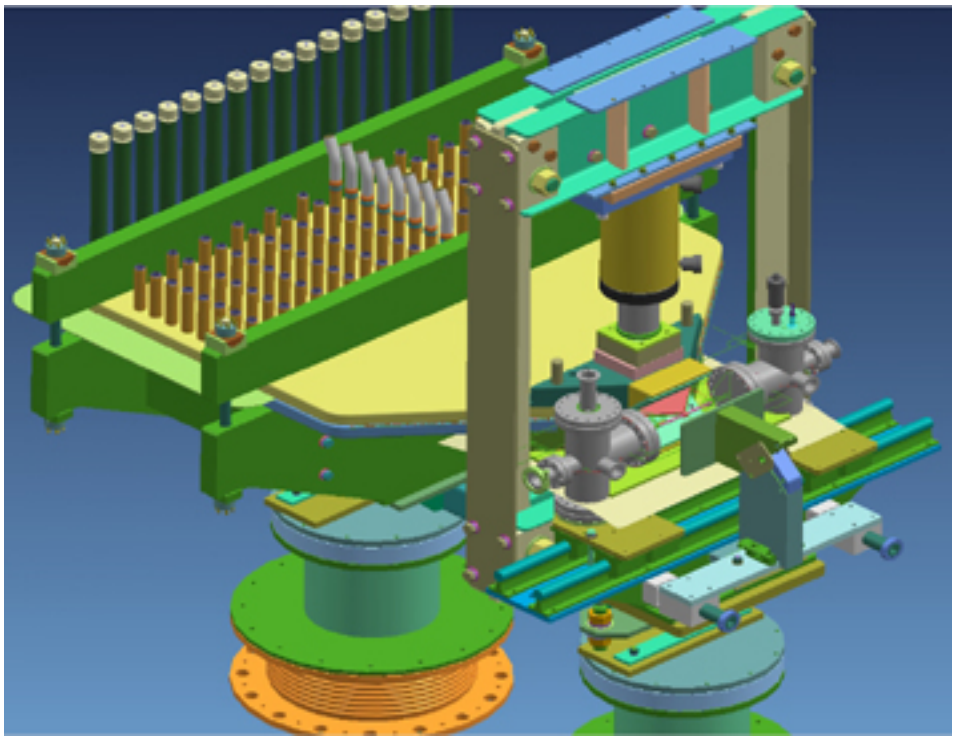

Figure 6: Single turn magnet

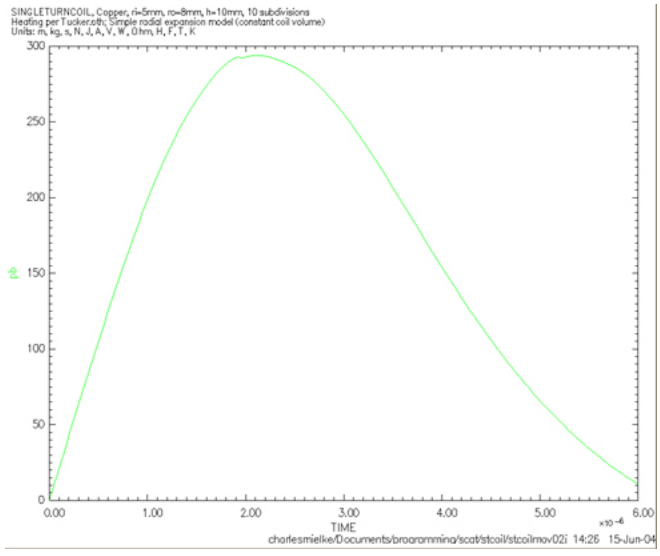

Figure 7: Single turn magnetic field pulse

Pu has six phases and thus the process of its phase changes is more complicated. It is hoped that after understanding the mechanisms of phase changes in Ce, this knowledge can be applied to better understanding Pu.

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