Anisotropic Elastic Properties of CeRhIn$_5$

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The structure of the quasi two dimensional heavy fermion antiferromagnet CeRhIn$_5$ has been investigated as a function of pressure up to 13 GPa using a diamond anvil cell under both hydrostatic and quasi-hydrostatic conditions at room ($T = 295$ K) and low ($T = 10$ K) temperatures. Complementary resonant ultrasound measurements were performed to obtain the complete elastic moduli. The bulk modulus ($B \approx 78$ GPa) and uniaxial compressibilities ($\kappa_a = 3.96 \times 10^{-3}$ GPa$^{-1}$ and $\kappa_c = 4.22 \times 10^{-3}$ GPa$^{-1}$) found from pressure-dependent x-ray diffraction are in good agreement with the ultrasound measurements. Unlike doping on the R$\delta$ site where $T_c$ increases linearly with the ratio of the tetragonal lattice parameters $c/a$, no such correlation is observed under pressure; instead, a double peaked structure with a local minimum around 4-5 GPa is observed at both room and low temperatures.

I. INTRODUCTION

Ce based heavy fermion (HF) antiferromagnetic (AF) compounds have been the subject of intensive investigations due to their unconventional magnetic and superconducting properties. In these compounds the electronic correlations, the magnetic ordering temperature and the crystal field effects are sensitive to pressure, and pressure induced superconductivity has been observed in a variety of compounds such as CePd$_2$Si$_2$, CeCu$_2$Ge$_2$, CeRh$_2$Si$_2$ and CeIn$_3$. The appearance of superconductivity in these systems and the deviation from Fermi liquid behavior as a function of pressure are still challenging problems to be studied. Recently, HF systems with the formula Ce$M$In$_5$ ($M =$ Co and Ir) have been reported to become superconductors at ambient pressure$^{16}$, while CeRhIn$_5$ is an antiferromagnet at ambient pressure ($T_N = 3.8$ K and $\gamma \approx 400$ mJ/mol K$^2$). These compounds crystallizes in the HoCoGa$_5$ structure with alternating stacks of CeIn$_3$ and $M$In$_2$ along the c axis. Thermodynamic, NQR, and neutron scattering experiments all show that the electronic and magnetic properties of CeRhIn$_5$ are anisotropic in nature. The AF ordering in CeRhIn$_5$ is suppressed with applied pressure and superconductivity is observed at 1.6 GPa with $T_c = 2.1$ K. Like CeIn$_3$ the bulk nature of the SC state in CeRhIn$_5$ has been unambiguously established under pressure. The AF state is suppressed at a pressure of around 1.2 GPa and coexists over a limited pressure range with the superconducting (SC) state$^{6,12,15}$. The value of $T_c$ in magnetically mediated superconductors is believed to be dependent on dimensionality in addition to the characteristic spin fluctuation temperature. Theoretical models and experimental results suggest that SC state in CeRhIn$_5$ may be due to the quasi-two dimensional (2D) structure and anisotropic AF fluctuations which are responsible for the enhancement of $T_c$ relative to CeIn$_3$. A strong correlation between the ambient pressure $c/a$ ratio and $T_c$ in the Ce$M$In$_5$ compounds is indicative of the enhancement of the superconducting properties by lowering dimensionality (increasing $c/a$ increases $T_c$)$^{14}$. In order to explain the evolution of superconductivity induced by pressure and the suppression of AF ordering, it is important to probe the effect of pressure on structure for this compound and look for possible correlations between structural and thermodynamic properties.

Here we report on high pressure x-ray diffraction measurements performed on CeRhIn$_5$ up to 13 GPa at high ($T = 295$ K) and low ($T = 10$ K) temperatures under both hydrostatic and quasi-hydrostatic conditions. As the measured linear compressibilities are similar for both the $a$ and $c$ directions, the results for all pressure measurements, both hydrostatic and quasihydrostatic, are similar. The elastic properties obtained from the high pressure measurements are compared to the full set of elastic constants obtained from resonant ultrasound (RUS) measurements, and excellent agreement is found in the measured bulk modulus ($B \approx 78$ GPa) from both techniques. We find no direct correlation between $c/a$ and $T_c$ as a function of pressure. Rather, a double peaked structure with a local minimum around 4-5 GPa is observed for $c/a$ at both room and low temperatures.

II. EXPERIMENT

CeRhIn$_5$ single crystals were grown by a self flux technique$^{9}$. The single crystals were crushed into powder and x-ray diffraction measurements show the single phase nature of the compound. In agreement with previous results$^{16}$, the crystals were found to have tetragonal symmetry with cell parameters $a = 4.6531(1)$ Å, $c = 7.538(9)$ Å.
The high pressure x-ray diffraction (XRD) experiments were performed using a rotating anode x-ray generator (Rigaku) for Runs 1-4 (λ =0.7093 Å) and synchrotron x-rays at HPCAT (λ =0.4218 Å), Sector 16 at the Advanced Photon Source for Run 5 and the low temperature measurement. The sample was loaded with NaCl or MgO powder as a pressure calibrant and either a 4:1 Methanol ethanol mixture (hydrostatic) or NaCl (quasi-hydrostatic) as the pressure transmitting medium in a Re gasket with a 180μm diameter hole. High pressure was achieved using a Merrill-Basset diamond anvil cell with 600 μm culet diameters. The XRD patterns are collected using an imaging plate (300 × 300 mm² ) camera with 100 × 100 μm² pixel dimensions. XRD patterns were collected up to 13 GPa at room (T = 295 K) and low (down to T = 10 K) temperatures. The low temperature measurements were made in a continuous flow cryostat. The images were integrated using FIT2D software. The structural refinement of the patterns was carried out using the Rietveld method on employing the FULLPROF and REITICA (LHPM) software packages. The RUS technique is described in detail elsewhere.

By measuring the resonant frequencies of a well aligned single crystal of CeRhIn₅, we can determine the full set of room temperature elastic constants. This will give the adiabatic bulk modulus Bₐ rather than the isothermal bulk modulus B₀ found in the pressure measurements.

### III. RESULTS AND DISCUSSION

In Fig. 1, we show the XRD patterns for CeRhIn₅ obtained at two different quasi-hydrostatic pressures with NaCl used as the pressure transmitting media. The raw data (crosses), Rietveld fit to the data (solid line through the data points), fit reflections (vertical lines) and the difference between the fit and experiment (solid line near bottom) are all shown. Fig. 2 shows the diffraction data at five different pressures. Diffraction peaks from the Re gasket (labeled g), NaCl (labeled *) and CeRhIn₅ (no label) are all observed in Fig. 2. The known equation of state for NaCl²¹ or the standard ruby fluorescence technique was used to determine the pressure. The refinement of the CeRhIn₅ XRD patterns was performed on the basis of the P4/mmm space group (No. 123). The HoCoGa₃ structure in which CeRhIn₅ crystallizes contains layers of cubo-octahedra of the structural type of AuCu₃ and layers of PtHg₂ structure type. The unit cell consists of Ce atoms situated at the corners and In atoms at two inequivalent sites. In1 is surrounded by Ce and located at the top and bottom faces while In2 is stacked between Ce-In and Rh layers. The hybrid structure is related to both CeIn₃ and Ce₃RhIn₅. When comparing the crystallographic data and bulk modulus of CeIn₃ it is evident that the Ce atom in CeRhIn₅ experiences a chemical pressure of 1.4 GPa at ambient conditions⁶.⁹

The results of the Rietveld refinement at different pressures have been listed in Table 1.

![Diagram](image-url)

**FIG. 1:** Rietveld refinements for the high pressure x-ray diffraction patterns of CeRhIn₅ at 0.26 GPa and 7.54 GPa. The NaCl pressure marker and various reflections from CeRhIn₅ are labeled.

| Pressure  | Rietveld parameters |
|-----------|---------------------|
| 0.26 GPa  | 1.47 GPa 3.97 GPa 5.18 GPa 7.54 GPa |
| a(Å)      | 4.6263(3) 4.5718(3) 4.5712(3) 4.5298(3) |
| c(Å)      | 7.505(1) 7.409(1) 7.396(1) 7.337(1) |
| In2 (z)   | 0.3036(3) 0.3049(4) 0.3089(3) 0.3058(3) |
| Bₐ(Å²)    | 0.3(1) 0.5(2) 0.3(1) 0.5(1) |
| Bₐ(Å²)    | 0.9(1) 1.3(3) 1.0(2) 1.7(1) |
| Bₐ(Å²)    | 1.7(2) 3.8(4) 1.4(3) 1.4(2) |
| Bₐ(Å²)    | 1.4(1) 1.3(1) 1.2(1) 0.8(7) |
| R_p (%)   | 2.1 2.6 2.5 2.1 |
| R_wp (%)  | 3.0 3.8 3.5 3.0 |

**TABLE I:** Room temperature structural parameters, isotropic thermal parameters B, and R factors of CeRhIn₅ at different pressures. The crystal structure is tetragonal and space group symmetry is P4/mmm (No.123) with Z = 1. The atomic sites are Ce at 1a [ 0, 0, 0 ]. Rh at 1b [ 0, 0,0.5 ]. In1 at 1c [ 0.5, 0.5, 0 ] and In2 at 4i [ 0, 0.5, z ].

During the refinement, a total of nineteen parameters have been optimized which include the background, scale factors, lattice parameters, profile parameters, temperature factors, zero point shift parameter and atomic coordinate. Initially the refinement has been started for two phases in most cases including the pressure calibrant, and at higher pressures an additional phase for the gasket has been added. At higher pressures, considerable changes in the isotropic temperature factors are observed for In1.
FIG. 2: X-ray diffraction patterns collected at various pressures for CeRhIn$_5$. Peaks labeled (g) correspond to the Re gasket and (*) to the NaCl pressure marker. Peaks without a label are from the CeRhIn$_5$ sample.

In$_2$ and Rh during the refinement.

The $V(P)$ data has been plotted for CeRhIn$_5$ for quasi-hydrostatic (Run 1 and Run 2) and hydrostatic (Runs 3-5) measurements in Fig. 3. Since the maximum volume compression is only of the order of 10%, the $V(P)$ data has been fit using a least squares fitting procedure to the second order Murnaghan equation of state

$$P = \frac{B_0}{B'_0} \left[ \left( \frac{V_0}{V(P)} \right)^{B'_0} - 1 \right].$$

For the room temperature ($T = 295$ K) data in Fig. 3, we find $B_0 = 78.4 \pm 2.0$ GPa and $B'_0 = 5.6 \pm 0.6$. The RhIn$_2$ layers in CeRhIn$_5$ appear to stiffen the structure relative to CeIn$_3$ which has a smaller bulk modulus ($B = 67$ GPa)$^{22}$. The bulk modulus value compares well with the values reported for other HF systems$^{24,25,26,27}$. Fig. 3 also shows the ratio of the lattice constants $c/a$ as a function of pressure. For all of the measurements, there appears to be a double peak structure with a local minimum around 4-5 GPa. Note that the isotropic thermal parameters for the In sites, in particular the In1 site, have their largest values around 4 GPa. The initial values of the linear compressibilities (average values from the hydrostatic measurements for $P < 2$ GPa) are $\kappa_a = (3.96 \pm 0.08) \times 10^{-3}$ GPa$^{-1}$ and $\kappa_c = (4.22 \pm 0.10) \times 10^{-3}$ GPa$^{-1}$. The similarity between the measured values of $\kappa_a$ and $\kappa_c$ are likely the reason that no discernible difference is found for the hydrostatic and quasi-hydrostatic cases. The $P-V$ data shows that the system retains its crystal structure up to the pressure limit (13 GPa) investigated.

We have also investigated the $V(P)$ behavior at low temperature ($T = 10$ K). As the superconducting transition has a maximum around 2 K, it is desirable to obtain structural data in the low temperature regime when try-
there is a significant enhancement of $c/a$ temperature at $P$ relative to the ambient pressure thermal expansion data of $V/V_0$. The solid line through the volume data is a fit to all of the data using $B_0 = 78.2 \pm 5.2$ GPa and $B'_0 = 4.8$. The dashed line shows the ambient pressure $c/a$ value. The solid line through the $c/a$ data is a guide for the eye.

FIG. 4: Normalized volume $V/V_0$ and ratio of tetragonal lattice constants $c/a$ plotted versus pressure for CeRhIn$_5$ at 10 K. The solid line through the volume data is a fit to all of the data using $B_0 = 78.2 \pm 5.2$ GPa and $B'_0 = 4.8$. The dashed line shows the ambient pressure $c/a$ value. The solid line through the $c/a$ data is a guide for the eye.

As mentioned, a strong correlation between the ambient pressure $c/a$ ratio and $T_c$ in the CeMIn$_5$ compounds has been observed (increasing $c/a$ increases $T_c$). To further investigate the variation of $c/a$ with pressure and temperature, we plot the value of $c/a$ as a function of temperature at $P = 6.9$ GPa in Fig. 5. As can be seen, there is a significant enhancement of $c/a$ at 6.9 GPa relative to the ambient pressure thermal expansion data of $B^{th}/B_0 = 1 + \beta \gamma_{th} T$, where $\beta = 4.6 \times 10^{-5}$ K$^{-1}$ is the volume thermal expansion coefficient and $\gamma_{th}$ is the thermal Gruneisen parameter which is typically of the order of unity. At room temperature then, one

FIG. 5: Ratio of tetragonal lattice constants $c/a$ plotted versus temperature for CeRhIn$_5$ at $P = 6.9$ GPa. The solid line is from ambient pressure thermal expansion data (Ref. 28).

Takeuchi et al. Unlike the ambient pressure data, $c/a$ appears to increase as temperature is lowered at 6.9 GPa. Taken as a whole, the current results seem to show no direct correlation between the values of $c/a$ and $T_c$. Though the lattice does contract at ambient pressure as temperature is lowered which would lead to a higher bulk modulus, the expected change is within our experimental uncertainty. The variation of $c/a$ as a function of pressure again shows a double maximum structure at low temperature in a manner similar to the room temperature data.

The complete set of elastic constants were measured using the RUS technique and the results are shown in Table II. The values of the adiabatic compressibility $B^S$, tetragonal shear modulus $C^t$, and linear compressibilities ($\kappa_a$, $\kappa_c$) can be calculated from the measured elastic constants and are given by

$$B^S = \frac{C_{33}(C_{11} + C_{12}) - 2C_{13}^2}{2C_{33} + C_{11} + C_{12} - 4C_{13}}$$

and

$$C^t = \frac{1}{6} (2C_{33} + C_{11} + C_{12} - 4C_{13}).$$

The results are displayed in Table II. The value of $B^S$ is slightly larger than the isothermal value $B_0$ obtained from the pressure measurements. This is to be expected as the ratio $B^S/B_0 = 1 + \beta \gamma_{th} T$, where $\beta = 4.6 \times 10^{-5}$ K$^{-1}$ is the volume thermal expansion coefficient and $\gamma_{th}$ is the thermal Gruneisen parameter which is typically of the order of unity. At room temperature then, one
then expects $B^s / B_0 \approx 1.01 - 1.02$ which is in reasonable agreement with our experimental value of $1.05 \pm 0.03$.

In all of the measurements, the $c/a$ ratio is found to have a double peaked structure. As mentioned previously, the hybridization between the Ce 4f electrons and the conduction electrons should mainly depend on the distance between Ce and its nearest neighbors. In fact, a simple model to estimate the hybridization by means of a tight-binding calculation shows that the hybridization should have the relatively strong $d^{-6}$ dependence for hybridization between f and d electrons, where d is the distance between the atoms containing the d and f electrons (in our case, this would be Rh and Ce respectively).

To examine the pressure dependence of $d$, the Ce-In1 and Ce-In2 bond lengths are plotted in Fig. 6 for the hydrostatic measurements. The Ce-In1 bond is less compressible than the Ce-In2 bond. The Ce-In2 data appears to display plateaus between 0-2 and 3-5 GPa. The structural results may be compared with the high pressure resistivity experimental data reported for CeRhIn$_5$. The smooth decrease in the Ce-In1 bond length with pressure shows a gradual decrease with increasing pressure. Our data is not sufficient to make any definite conclusions along these lines. The smooth decrease in the Ce-In1 bond length would lead one to expect the typical inverse parabolic $T_K(P)$ dependence consistent with theoretical calculations and measurements on CeRhIn$_5$ and most heavy fermion superconductors.

IV. CONCLUSIONS

We have studied the elastic properties of the heavy fermion system CeRhIn$_5$ using resonant ultrasound and hydrostatic and quasihydrostatic pressures up to 13 GPa using x-ray diffraction. The bulk modulus ($B = 78$ GPa) and uniaxial compressibilities ($\kappa_a = 3.96 \times 10^{-3}$ GPa$^{-1}$ and $\kappa_c = 4.22 \times 10^{-3}$ GPa$^{-1}$) found from pressure-dependent x-ray diffraction are in good agreement with the ultrasound measurements. Unlike doping experiments which hint at a strong correlation between the $c/a$ ratio and $T_K$, pressure shows no such correlation as a double peaked structure with a local minimum around 4-5 GPa is found at 295 K and 10 K.

| Elastic Constant | Value (GPa) |
|------------------|-------------|
| $C_{11}$         | 146.7       |
| $C_{12}$         | 45.8        |
| $C_{14}$         | 43.4        |
| $C_{13}$         | 141.4       |
| $C_{15}$         | 54.0        |
| $C_{16}$         | 41.8        |

| Moduli          | Value (GPa) |
|-----------------|-------------|
| $B^s$ (RUS)     | 82.5        |
| $C^s$ (RUS)     | 43.2        |
| $B_0$ (P)       | 78.4        |

| Compressibilities | Value (GPa$^{-1}$) |
|-------------------|--------------------|
| $\kappa_a$ (RUS)  | 4.09 $\times 10^{-3}$ |
| $\kappa_c$ (RUS)  | 3.96 $\times 10^{-3}$ |
| $\kappa_a$ (P)    | 3.96 $\times 10^{-3}$ |
| $\kappa_c$ (P)    | 4.22 $\times 10^{-3}$ |

FIG. 6: The measured Ce-In1 and Ce-In2 bond lengths for both hydrostatic measurements on CeRhIn$_5$ as a function of pressure.

| TABLE II: A summary of the CeRhIn$_5$ elastic constants $C_{ij}$ measured using resonant ultrasound and the various moduli and compressibilities measured by resonant ultrasound (RUS) and pressure (P). |
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