Electronic Structure of CeRhIn₅: dHvA and Energy Band Calculations.

Donavan Hall¹, E. Palm¹, T. Murphy¹, S. Tozer¹, Eliza Miller-Ricci¹, Lydia Peabody¹, Charis Quay Huei Li², U. Alver², R. G. Goodrich², J. L. Sarrao³, P.G. Pagliuso³, J. M. Wills⁴, and Z. Fisk¹
¹ National High Magnetic Field Laboratory, Florida State University, Tallahassee, FL 32310.
²Department of Physics and Astronomy, Louisiana State University, Baton Rouge, LA 70803.
³ Los Alamos National Laboratory, MST-10, Los Alamos, NM 87545,
⁴ Los Alamos National Laboratory, T-1, Los Alamos, NM 87545,
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The de Haas - van Alphen effect and energy band calculations are used to study angular dependent extremal areas and effective masses of the Fermi surface of the highly correlated antiferromagnetic material CeRhIn₅. The agreement between experiment and theory is reasonable for the areas measured with the field applied along the (100) axis of the tetragonal structure, but disagree in size for the areas observed for the field applied along the (001) axis where the antiferromagnetic spin alignment is occurring. Detailed comparisons between experiment and theory are given.

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I. INTRODUCTION

The compounds CeMIn₅ (M=Co, Ir, Rh) are a newly reported family of heavy fermion superconductors. These materials crystallize in the tetragonal HoCoGa₅ structure and are built of alternating stacks of CeIn₃ and Mn₂. CeCoIn₅ and CeIrIn₅ have superconducting transition temperatures of 2.3 K and 0.4 K, respectively, whereas CeRhIn₅ orders antiferromagnetically at 3.8 K at ambient pressure. Applied pressures of order 16 kbar can induce an apparently first-order transition from the magnetically ordered state to a superconducting one with Ƭᵣ= 2.1 K. The particular attraction of these materials is that not only do they represent a substantial increase in the number of known heavy fermion superconductors but also they appear to be quasi-2D variants of CeIn₃, an ambient-pressure antiferromagnet in which superconductivity can be induced at 25 kbar and 100 mK. If one can demonstrate that the reduced dimensionality is responsible for the factor of ten increase in superconducting Ƭᵣ as well as the anomalous evolution from antiferromagnet to superconductor with pressure, their impact will far exceed that of just another family of heavy fermion superconductors. However, much work remains to establish this hypothesis.

Because it displays both ‘high-temperature’ superconductivity as well as unconventional magnetic behavior, CeRhIn₅ seems an ideal candidate for electronic and magnetic spectroscopic probes to establish the degree of reduced dimensionality and its impact on fluctuation spectra. Some progress has been made in establishing the magnetic structure of CeRhIn₅. Nuclear quadrupole resonance and neutron diffraction measurements have shown that the magnetically ordered state of CeRhIn₅ is a spiral spin structure in which the spins in a given CeIn₃ plane are ordered antiferromagnetically, and the direction of the ordered moments in the plane spirals along the c-axis, rotating 107 degrees per layer. Although complete inelastic neutron scattering measurements of the dynamical susceptibility remain in progress, already NQR has established that the temperature evolution of the sublattice magnetization below Ƭ_N is much more rapid than would be expected from mean field theory and is a sign of reduced dimensionality.

To date, studies of the electronic structure of CeRhIn₅ have been far less complete. Cornelius et al. have reported limited de Haas-van Alphen (dHvA) measurements along the principal crystallographic axes using pulsed fields. These measurements reveal an anomalous temperature-dependence of the dHvA amplitude as well as large directional anisotropies in the effective masses of carriers that have been observed.

Here, we report a more comprehensive dHvA study throughout the entire Brillouin zone of CeRhIn₅ and compare our results directly with local-density-approximation band structure calculations. Recently, a similar study of CeIrIn₅ has been reported. Taken together, our results and those of Reference ⁹ allow direct evaluation of the changes in electronic properties of CeMIn₅ as a function of transition metal ion M.

In what follows, we report the details of our dHvA measurements and band structure calculations, discuss the extent to which they are self-consistent, and compare and contrast the electronic structures of CeRhIn₅ and CeIrIn₅.

II. MEASUREMENTS

All of the measurements reported here were made at the National High Magnetic Field Laboratory, Tallahassee, FL using cantilever magnetometry at temperatures between 20 and 500 mK in applied fields ranging from 5 to 18 T. Complete field rotations in the [100] and [001] planes of the tetragonal structure are reported. The sample was grown from an In flux and etched in a 25% HCl in H₂O solution down to a small plate that was mounted on
the cantilever with Apiezon N vacuum grease or GE Varnish. This technique is most sensitive to low frequency oscillations not observed typically with the magnetic field modulation technique.

The dHvA effect, a measurement of the oscillatory part of the magnetization, is a method for determining Fermi surface properties. The oscillatory magnetization $M$ is given by the Lifshitz-Kosevitch (LK) equation (see Ref. 10 for the mathematical details):

$$M = -2.602 \times 10^{-6} \left( \frac{2\pi}{HA^\prime} \right)^{1/2} \times \frac{GFT \exp(-\alpha p x/H)}{p^{3/2} \sinh(-\alpha p T/H)} \sin \left( \frac{2\pi p F}{H} \right) - \frac{1}{2} \pm \frac{\pi}{4},$$  \hspace{0.5cm} (1)

where $\alpha = 1.47(m/\mu_0) \times 10^5$ G/K, $A^\prime$ is the second derivative of the area of the Fermi surface (FS) cross-section that is perpendicular to the applied field, $G$ is the spin reduction factor, $p$ is the harmonic number, and $x$ is the Dingle temperature. The frequencies of the dHvA oscillations are proportional to extremal areas of the FS, and Fermi-liquid theory works well for heavy-fermion materials as has been shown in previous studies.

Since the magnetization of the sample was measured with a torque cantilever, it is necessary to understand how torque is related to the LK equation. dHvA oscillations in the torque arise from anisotropy in the Fermi surface, such that

$$\tau = -\frac{1}{F} \frac{dF}{d\theta} M HV$$  \hspace{0.5cm} (2)

where $F$ is the dHvA frequency, $\theta$ is the angle of the applied field, $M$ is the LK expression above, and $V$ is the volume of the sample. This means that a roughly spherical FS will have a smaller torque signal than a highly elliptical FS.

Two versions of a Fourier transform algorithm were used in the analysis: preliminary analysis was done with a fast Fourier transform (FFT) and in some cases a discrete Fourier transform (DFT) was used to increase the frequency resolution. Fourier spectra of the oscillations obtained for the field parallel to [100] and [001] are shown in Figure 1 and Figure 2. As can be seen there are several fundamental frequencies and several combinations (especially with the $F_7$; e.g. $F_7 - F_6 = 1302$ T). Similar combination frequencies were found in all field directions; their origins will be discussed below.

The fundamental frequencies are plotted as a function of angle in Figure 3. As can be seen the measured frequencies span the range from about 100 T over wide angular ranges to 5 kT for the field applied near [001]. The one defect of the cantilever technique was an angular range of about 40 degrees, producing a small signal amplitude. The measured dHvA oscillations were small for [001] and [100] orientations, and the signal amplitude was maximized at approximately a forty-five degree rotation from each of these principal axes. One might propose that field modulation measurements would be more appropriate in this case, but given the low (less than 1 kT) frequencies measured here, the required modulation fields are technically impractical in the high field ranges used.

### III. DISCUSSION

The total energy and band structure of CeRhIn$_5$ were calculated with a full-potential electronic structure method that uses linear muffin-tin orbitals as bases. Calculations were performed to obtain theoretical structural parameters, one-electron bands and spectral densities, and the structure of the Fermi surface. All calculations used the GGA to treat exchange and correlation and included the spin-orbit interaction in the variational basis. In the range of parameters searched, no magnetic instabilities were found. The calculations reported here are for paramagnetic CeRhIn$_5$ in the simple tetragonal structure (P4/mmm).

Structural parameters were obtained by simultaneous variation of volume, $c/a$, and the single internal structural parameter to minimize the total energy. Substantially converged results were obtained with a set of 45 irreducible (486 total) Brillouin zone points. The calculated volume, $c/a$ ratio, and internal parameter are given in Table I. Conventional electronic structure theory (with itinerant Ce 4f electrons) applied to a simple paramagnetic cell seems to give a satisfactory description of the structural properties of CeRhIn$_5$. Also given in Table I are the bulk modulus calculated at both the experimental and theoretical volumes and the Density of States (DOS) at the Fermi energy ($E_F$) calculated at the experimental volume.

The band structure, calculated at the experimental volume, along high symmetry lines in the Brillouin zone is shown in Figure 4. Bands of predominantly Ce 4f character dominate the band structure at and just above $E_F$. The set of bands along $X \rightarrow \Gamma \rightarrow Z \rightarrow R$ are indicative of the nesting found in the Fermi surface discussed below. The electron DOS, shown in Figure 5, shows the predominance of Ce 4f character at the Fermi energy. The DOS at $E_F$, while fairly large, does not indicate a dramatic structural instability.

Fermi surfaces were calculated using a fine mesh (4800 irreducible points, corresponding to a $48 \times 48 \times 32$ grid) of the Brillouin zone) with a potential converged with 270 irreducible Brillouin zone points. Three doubly degenerate bands, which we label band 90, band 92, and band 94, cross the Fermi energy. The Fermi surface sheets formed by these three bands are shown in Figures 6, 7, 8. In the figures, $\Gamma$, the center of the conventional Brillouin zone, is at the origin, i.e. at the corners of the cubes. The lowest energy sheet, band 90, shown in Figure 6, consists of hole surfaces centered around $\Gamma$ and $X$, giving relatively low (~600 T) dHvA frequencies. The next
sheet, shown in Figure 7, has both hole and electron surfaces stretched along the tetragonal axis. The last sheet, shown in Figure 8, is predominantly an electron surface, although there is a large area ([001]) hole surface on that sheet. This sheet consists of cylinders surrounding [001] axis and a lattice-structure surface. The dHvA frequencies corresponding to extremal areas are given in Tables I and II.

We measured the temperature dependence of the dHvA amplitudes in the primary crystallographic directions. We did not observe the anomalous temperature-dependence of the dHvA amplitudes noted in Ref. 8, where for B || [001] the amplitudes of the oscillations increase with increasing temperatures between 0.4 and 1.2 K then decrease. Our measurements, carried out between 25 and 500 mK, show decreasing amplitudes with increasing temperature in all cases.

The measurements reported here agree substantially with Cornelius et al.14 after a 5% correction to their data, all but one (4708 T) of the frequencies reported in Ref. 8 also are observed here. One major difference is the measurements along [001] where we see many more frequencies. Our measured frequencies in this direction run low as compared to calculations, but we share two frequencies measured by Cornelius et al.: our F7 and F8 (see Table IV). Also, the masses we measure for these orbits are in excellent agreement (see Table V). Cornelius et al. report a high mass orbit at 4686 T which we do not see. Another major difference is our measured masses for [100] are higher when compared with those of Cornelius et al. (see Table V); a fact consistent with our non-observance of the anomalous temperature dependence.

IV. COMPARISON OF EXPERIMENT WITH THEORY

We start the comparison between the calculated frequencies and the measured frequencies with those calculated and observed for the field along the [100] direction. In this case there are many more measured frequencies than the calculated FS gives. We attribute this situation to strong magnetic interactions, between electrons on different parts of the FS, or to torque interactions due to the use of the cantilever. Either of these effects can give rise to a series of frequencies that are combinations of two fundamental frequencies. The measured fundamental frequencies in Table I are indicated as F1 through F7, and some of the combinations that are differences between the fundamentals or harmonics of a fundamental and another fundamental frequency are shown in Figure 3. It should be noted that the same fundamental frequencies are subtracted from both the fundamental 2176 T and the first two harmonics of 2176 T (F7) to give the observed combination frequencies (see the inset in Figure 3). In one case, 1744 T is the first harmonic of the fundamental 874 T (F6) frequency.

Five frequencies ranging from 9 to 26 T obtained in the band calculations are too low to be observed experimentally. That is, in the field range where oscillations are observed (from about 7 to 18 T) there are too few oscillations for these frequencies to be measurable. In addition there are four electron orbits grouped into two pairs, 245 T and 215 T plus 175 T and 110 T, for which we observe only two frequencies, 212 T and 106 T. Again, the Fourier transform may not have sufficient resolution due to an insufficient number of oscillations to resolve all of these frequencies.

The remaining five measured frequencies are in the same range as the predicted frequencies from the band calculation. Differences in detail occur, but it appears that for the field in this direction the measurements give strong support to the predicted FS.

The agreement between the calculated and measured frequencies is not as good for the field in the [001] direction. There are three calculated high frequencies, 11.268 kT, 12.126 kT, and 12.326 kT, whereas only one frequency in this range is observed, 12.280 kT. This high frequency only is observed in the lowest temperature data, 20 mK, and indicates a large effective mass for this frequency. The fact that only one rather than three frequencies are observed may indicate that the electron surface is much more cylindrical than is obtained from band theory. For the remainder of the frequencies, only the total number and order of the observed frequencies agree with the calculations.

In the band calculation the fact that the sample is antiferromagnetic over all of the measurement field ranges has not been taken into account. In the model that has been taken for the band calculations, the Ce 4f electron is completely itinerant and there are no localized moments to cause the magnetism. Therefore, it is most likely that spin density waves exist in the electronic system and drastically alter the FS. This is the accepted situation in Cr and may be the case here. A much more involved theory is required to predict the FS under these circumstances. Exchange interactions could cause the FS to be split into separate spin up and spin down sheets. In order for this to be determined from dHvA measurements several harmonics of these frequencies would have to be measured. We did not observe the harmonics in these measurements, and need to make measurements to higher fields at mK temperatures to do so.

V. COMPARISON OF CERHIN5 WITH CEIRIN5

The structure of the Fermi surface of CeRhIn5 is essentially the same as that calculated for CeIrIn5 by Haga et al., except that the frequency of the hole orbit perpendicular to [001] arising from (what we label) band 94 near Γ is ∼ 11000 T, rather than ∼ 15000 T reported in Reference 8. Our own calculations for CeIrIn5 find a frequency close to that calculated for CeRhIn5, ∼ 11600
The high frequency reported in Reference 3 was not seen experimentally. The difference in the two calculations may be in our inclusion of the spin orbit interaction or in the fineness of the grid used to calculate the Fermi surface.

The dHvA measurements on CeIrIn₅ find eight branches for rotations in the [100] and [110] planes of the tetragonal structure. Most of these branches are associated with large quasi-2D undulating cylinders that show the expected $1/\cos(\theta)$ dependence of the measured frequencies with $\theta$ being the angle at which the field is applied away from the [100] axis. We see a similar behavior in the high frequency orbits for CeRhIn₅. Band structure calculations predict, in Ref. 4 and in Ref. 17, in addition, that several small pieces of FS should exist in both CeRhIn₅ and CeIrIn₅. These frequencies were not observed in Ref. 5 for CeIrIn₅ but are observed here in CeRhIn₅. These differences likely are a result of the measurement techniques employed and not due to differences in the actual Fermi surfaces of the respective materials. Overall, the result of these angular dependent dHvA measurements on CeRhIn₅ along with the band calculations show that the Fermi surface has both small pockets of 3D character and large undulating cylinders that should give rise to a 2D character in other properties.

From the temperature dependence of the signal amplitudes we have calculated the effective masses of the observed frequencies in cases where signals were observable over the entire temperature range of measurement. There are two cases where no masses are reported here: (1) for $F'_1 - F'_3$ in Table III the masses are sufficiently light that we observe no amplitude change with measurement uncertainty up to 0.5 K, and (2) for $F_7$ in Table I and $F'_5$, $F'_6$, $F'_9$ and $F'_5$ in Table II the signals are observed only at the lowest temperatures. The masses are obtained from fits to the Dingle reduction factor in Eq. 1:

$$R_T = \frac{\pi \lambda}{\sinh(\pi \lambda)},$$

where $\lambda = 2\pi p k T/\beta B$, $\beta = e h/(m/m_0)c$, and $m/m_0 = m^*$ is the effective mass. All of the effective masses we obtain in this manner are greater than 1, and compare favorably to the values found in Ref. 5 from the high temperature dHvA data. We note that none of the measured masses are sufficiently large to account for the measured value of gamma of 400 mJ/mole-K from high temperature specific heat measurements. On the other hand, there is a loss of entropy due to magnetic ordering, and the inferred value of gamma below $T_N$ is 56 mJ/mole-K, much more in line with the masses reported here. The effective masses measured on CeIrIn₅ are larger than we observe in CeRhIn₅ consistent with the larger value of gamma measured in CeIrIn₅ (in which no magnetic order is observed).

VI. CONCLUSION

We have determined the electronic structure of CeRhIn₅. This determination includes both the results of dHvA measurements and energy band calculations. The major findings are:

- The Fermi surface consists of structures arising from two hole bands and one electron band. The band calculations assume a completely itinerant $4f$ electron from the Ce atoms and the measurements confirm this assumption. One of the hole surfaces and the electron surface are open along the $c$ - axis and could give rise to some rather two-dimensional character to the electronic properties.

- While the energy band calculations are for paramagnetic CeRhIn₅ and the measurements are on anti-ferromagnetic CeRhIn₅, the agreement between experiment and theory is good for electron areas perpendicular to the $a$-$b$ plane, but deviate in size for orbits in this plane. This disagreement is thought to be due to spin density waves in the c axis direction that give rise to the anti-ferromagnetism in the a - b plane.

- The electronic effective masses that are measured all are greater than the free electron mass and are anisotropic, varying from less than $m_0$ to 6$m_0$ depending on direction.

Overall the Fermi surface of anti-ferromagnetic CeRhIn₅ is similar to that found previously in superconducting CeIrIn₅. Thus, while their ambient pressure ground states are completely different, both of these materials have large electron - electron interactions and rather similar electronic structures.

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| TABLE I. Structural parameters of primitive tetragonal CeRhIn₅. Calculated parameters were obtained using GGA and brillouin zone convergence as discussed in the text. The parameter $x$ is the Wykoff coordinate of displacement of face-centered In atoms along $\tau$. Experimental values from Hegger et al. |
|-----------------|-----------|-----------------|--------|
| Space Group     | P4/mmm   | (Vexp = 163.4 Å³) |
| V/Vexp          | 1.023    | (Vexp = 163.4 Å³) |
| c/a             | 1.626    | c/aexp = 1.620   |
| x               | .302     | xexp .306       |
| B (Vexp)        | 87 GPa   |                 |
| B (Vcal)        | 78 GPa   |                 |
| Dos(EF)         | 10.8 eV⁻¹|                 |
TABLE II. Measured and calculated dHvA frequencies for CeRhIn\(_5\) with \(\vec{B}\) along [100]. Frequencies listed for Cornelius et al. have been corrected by 4.5 percent (private communication).

| calc. F (T) | band | F (T) | Symb. | F (T) |
|-------------|------|-------|-------|-------|
| 2139        | 92 (h)| 2176  | F\(_7\) |       |
| 849         | 92 (e)| 874   | F\(_6\) | 861   |
| 761         | 92 (e)| 722   | F\(_5\) | 714   |
| 551         | 92 (h)| 446   | F\(_4\) | 492   |
| 362         | 90 (h)|       |       |       |
| 342         | 90 (h)|       |       |       |
| 337         | 90 (h)| 324   | F\(_3\) | 295   |
| 283         | 94 (e)|       |       |       |
| 245         | 90 (h)|       |       |       |
| 214         | 94 (e)| 212   | F\(_2\) | 219   |
| 175         | 94 (e)|       |       |       |
| 110         | 94 (e)| 106   | F\(_1\) | 105   |
| 26          | 92 (h)|       |       |       |
| 21          | 92 (h)|       |       |       |
| 10          | 90 (h)|       |       |       |
| 9           | 90 (h)|       |       |       |

TABLE III. Measured and calculated dHvA frequencies for CeRhIn\(_5\) with \(\vec{B}\) along [001].

| calc. F (T) | band | meas. F (T) | Symb. | F (T) |
|-------------|------|-------------|-------|-------|
| 12326       | 92 (e)| 12280*      | F\(_{10}\) |       |
| 12126       | 92 (e)|            |       |       |
| 11268       | 94 (h)|            |       |       |
| 6878        | 92 (e)| 7200       | F\(_9\) |       |
| 5562        | 94 (e)| 6120       | F\(_8\) | 6256  |
| 4502        | 94 (e)| 3600       | F\(_7\) | 3605  |
| 4331        | 94 (e)| 2740       | F\(_6\) |       |
| 3951        | 94 (e)| 2160       | F\(_5\) |       |
| 686         | 90 (h)| 1412       | F\(_4\) |       |
| 597         | 90 (h)| 562        | F\(_3\) |       |
| 585         | 90 (h)| 216        | F\(_2\) |       |
| 451         | 90 (h)| (115)      | F\(_1\) |       |

TABLE IV. Measured masses for dHvA frequencies for CeRhIn\(_5\) with \(\vec{B}\) along [100]. Our data also are compared to that of Cornelius et al.

| Symb. | F (T) | m* (m\(_e\)) | F (T) | m* (m\(_e\)) | Ref. |
|-------|------|--------------|------|--------------|-----|
| F\(_6\)| 874  | 3.3 ± 0.6    | 861  | 1.31 ± 0.22  |     |
| F\(_5\)| 722  | 2.0 ± 0.8    | 714  | 1.17 ± 0.09  |     |
| F\(_4\)| 446  | 3.9 ± 0.9    | 492  | 0.72 ± 0.14  |     |
| F\(_3\)| 324  | 5.5 ± 1.0    | 295  | 0.93 ± 0.15  |     |
| F\(_2\)| 212  | 2.9 ± 0.4    | 219  | 0.99 ± 0.10  |     |
| F\(_1\)| 106  | 3.3 ± 0.6    | 105  | 1.36 ± 0.23  |     |

TABLE V. Measured dHvA frequencies for CeRhIn\(_5\) with \(\vec{B}\) along [001]. Frequencies marked with asterisks appear only at the lowest temperatures. Calculation of masses for the frequencies marked with daggers was not possible because the amplitudes did not change significantly over the measured temperature range. The frequency in parentheses is weak due to a low number of oscillations in the sweep range.

| Symb. | F (T) | m* (m\(_e\)) | F (T) | m* (m\(_e\)) | Ref. |
|-------|------|--------------|------|--------------|-----|
| F\(_{10}\)| 12280* | – | – | – | – |
| F\(_9\)| 7200* | – | – | – | – |
| F\(_8\)| 6120 | 6.1 ± 0.3 | 6256 | 6.5 ± 0.8 | – |
| F\(_7\)| 3600 | 4.6 ± 1.0 | 3605 | 4.8 ± 0.4 | – |
| F\(_6\)| 2740* | – | – | – | – |
| F\(_5\)| 2160* | – | – | – | – |
| F\(_4\)| 1412 | 4.5 ± 0.3 | – | – | – |
| F\(_3\)| 562† | – | – | – | – |
| F\(_2\)| 216† | – | – | – | – |
| F\(_1\)| (115)† | – | – | – | – |
FIG. 1. DFT of dHvA data taken with the applied field parallel to the [100]. The inset shows some of the high frequencies that are combinations of the fundamentals.

FIG. 2. FFT of dHvA data taken with the applied field parallel to the [001].

FIG. 3. Frequency as a function of angle. Calculated frequencies for [001] and [100] are indicated with bowties.

FIG. 4. The GGA band structure of paramagnetic CeRhIn$_5$ calculated using experimental structural parameters.
FIG. 5. The GGA density of states of paramagnetic CeRhIn$_5$ calculated using experimental structural parameters. Ce 4f and 5d projections are shown in (a), and Rh and In projections in (b).

FIG. 6. The first sheet of the paramagnetic CeRhIn$_5$ Fermi surface. The electron surface is illuminated; these are hole surfaces.

FIG. 7. The second sheet of the paramagnetic CeRhIn$_5$ Fermi surface. The electron surface is illuminated.

FIG. 8. The third sheet of the paramagnetic CeRhIn$_5$ Fermi surface. The electron surface is illuminated.

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* Present addresses: EMR, Middlebury College, Middlebury, VT 05753; LP, Smith College, Northampton, MA 01063; CQHL, Mount Holyoke College, South Hadley, MA 01075

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