Observation of de Haas-van Alphen oscillations across the phase diagram of CeRh$_{1-x}$Co$_x$In$_5$

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Abstract. The evolution of the Fermi surface of CeRh$_{1-x}$Co$_x$In$_5$ was studied as a function of Co concentration $x$ via measurements of the de Haas-van Alphen effect. We observe several quantum oscillation frequencies in single crystal samples prepared with values of $x$ spanning the alloy phase diagram. By tracking the evolution of these frequencies as the applied magnetic field is rotated, we establish that the Fermi surface undergoes an abrupt change in shape when $x \simeq 0.4$. This is well away from the quantum critical concentration $x \simeq 0.8$ where antiferromagnetic order gives way to paramagnetism.

1. Introduction

In rare earth compounds the character of the 4$f$ electrons is often dictated by the competition between long range Ruderman-Kittel-Kasuya-Yosida interactions and Kondo screening [1]. Varying the magnetic exchange constant $J$ can tune such a system between ground states in which the 4$f$ electrons behave very differently – an antiferromagnetic (AFM) state where they are localized and magnetically coupled via the conduction electrons, and a paramagnetic ground state where the 4$f$ moments are subsumed into the conduction electron sea. The character of the 4$f$ electrons has direct consequences for the electronic structure, and hence the size and shape of the Fermi surface [2].

The CeTIn$_5$ (T: Co, Rh, Ir) family of compounds offers an ideal system in which to explore the role of the 4$f$ electrons, as the electronic ground state may be varied in a controlled manner, achieved by applying hydrostatic pressure or alloying between different concentrations of T. Applying pressure to antiferromagnetically ordered CeRhIn$_5$ for instance suppress $T_N$ from 3.8 K at ambient pressure toward a quantum critical point (QCP) at a critical pressure $P_c = 2.35$ GPa [3].

The evolution of the Fermi surface in CeRhIn$_5$ through this pressure range was recently investigated by studying the de Haas-van Alphen (dHvA) effect [4], which revealed a dramatic reconstruction at $P_c$, accompanied by the divergence of the cyclotron mass $m^*$ on several sheets. This abrupt change was interpreted as the hallmark of a transition from localized to delocalized...
Figure 1. Fourier spectrum of quantum oscillations observed in CeRh\(_{0.63}\)Co\(_{0.37}\)In\(_5\) with \(B \parallel [001]\), obtained from data collected at 15 mK (upper inset). The temperature dependence of the dHvA amplitude for the 6.2 kT orbit is fitted (solid line) in the lower inset to extract the effective mass \(m^*\).

\(4f\) electron states, an occurrence not easily understood within the traditional spin density wave scenario of an antiferromagnetic quantum critical point \([2]\).

Here we present a dHvA study of the electronic structure of CeRh\(_{1-x}\)Co\(_x\)In\(_5\) as a function of Co concentration \(x\) across the QCP separating the AFM and nonmagnetic phases. Near the critical concentration \(x_c\), where \(T_N \rightarrow 0\), we find no evidence of reconstruction of the observable quasi-2D Fermi surface sheet nor any substantial enhancement of the cyclotron mass. Instead, we find a change in Fermi surface character to occur away from the critical concentration, coincident with a sharp suppression of the superconducting transition temperature.

2. Experimental

Previous dHvA studies of CeRhIn\(_5\) (\(x = 0\)) \([5, 4, 6]\) and CeCoIn\(_5\) (\(x = 1\)) \([7]\) have established the Fermi surface of both compounds to have a common geometry consisting of: (1) a heavy quasi-2D cylindrical sheet giving rise to orbits labeled \(\alpha_i\), (2) a heavy and more complex quasi-2D sheet with orbits \(\beta_i\), and (3) several small, light pockets with corresponding low-frequency orbits \([7]\). Band structure calculations \([8]\) compare well with these observations if the Ce \(4f\) electrons are treated as localized in CeRhIn\(_5\) and itinerant in CeCoIn\(_5\), a picture further supported by comparisons to the non-\(f\)-electron analogue LaRhIn\(_5\) \([5, 9]\).

Our dHvA studies were performed using high-quality, flux-grown single crystals of CeRh\(_{1-x}\)Co\(_x\)In\(_5\) \([10]\). Studies of the \(x\)-evolution of lattice parameters, resistivity \(\rho(T)\), the electronic specific heat \(C/T\) and \(a\)- and \(c\)-axis DC magnetic susceptibility \(\chi(T)\) match the nominal concentrations of these samples with those of previous work \([10]\). Samples with \(x < 0.4\) show signatures only of \(T_N\), while samples with \(0.4 < x < 0.7\) show a coexistence of superconductivity and antiferromagnetism. At higher concentrations, with \(x > 0.7\), only \(T_c\) is observed. A full description of the characterisation procedure and the results of transport and thermodynamic studies is given elsewhere \([11, 10]\).

Quantum oscillation measurements were carried out to temperatures as low as 10 mK using a second harmonic field modulation technique \([12]\). We observed oscillations in the field range 14-
18 T, and a typical data set is shown in the top inset of Fig. 1. This data is Fourier-transformed to obtain the dHvA frequency $F$ (Fig. 1, main panel), a direct measure of the cross-sectional Fermi surface area perpendicular to the applied field. The cyclotron effective mass $m^*$ is extracted from the temperature dependence of the dHvA amplitude by fitting the data with the standard Lifshitz-Kosevich formula [12], as shown in the bottom inset of Fig. 1. The error bars in our effective mass fits are determined by the noise floor of the dHvA spectrum at each temperature.

We rule out the possibility of significant phase separation in our samples in a number of ways. Previous studies [13, 10] on crystals from the same growth batches as ours show a continuous evolution of lattice parameters as a function of Co concentration. Both a- and c-axis constants increase linearly as expected by Vegard’s law, with the unit cell volume decreasing with $x$ as expected by the replacement of Rh with the smaller ion Co. This is direct evidence for a continuous, homogeneous substitution of Co for Rh in this series.

The observation of dHvA signals from all samples studied also suggests that inhomogeneities arising from chemical substitution are negligible. A calculation of the cyclotron length for the 6 kT orbit observed in Fig. 1 at a field of 14 T gives a length of 0.15 $\mu$m, requiring grains arising from any phase separation to be at least this size, a possibility ruled out by x-ray analysis. Although it is unusual to see quantum oscillations in samples far from stoichiometric conditions, we note that Co and Rh are isovalent, and the principle role of these atoms in the CeRh$_3$-Co$_2$In$_5$ system is as a passive bystander, controlling the separation between the CeIn$_5$ planes. Comprehensive quantum oscillation studies in CeTIn$_5$ materials where La substitutes for Ce may be taken as further evidence that controlled alloying resulting in high quality, homogeneous, single crystals is achievable in this system [14].

3. Analysis

3.1. Comparison to to CeRhIn$_5$ and CeCoIn$_5$

In order to track the evolution of the Fermi surface as a function of $x$, we compare our oscillation spectra observed at six different $x$ concentrations to those of the end-member compounds CeCoIn$_5$ and CeRhIn$_5$. In particular, we focus on the rotational dependence of the observed dHvA frequencies $F(\theta)$, obtained by varying the angle $\theta$ between the c-axis and the applied field direction $\vec{B}$. Since $F(\theta)$ is uniquely determined by the geometry of the Fermi surface, a comparison of CeRh$_{1-x}$Co$_x$In$_5$ to previously obtained data on CeRhIn$_5$ [5] and CeCoIn$_5$ [7] allows for an unambiguous determination of exactly where the Fermi surface changes from the geometry where the 4$f$ electrons are localized (‘Rh-like’) to that where the 4$f$ electrons contribute to the Fermi volume (‘Co-like’).

Fig. 2 shows $F(\theta)$ data for selected samples spanning the alloying phase diagram. The angular dependence of the observed dHvA frequencies of the $x = 0.37$ and $x = 0.40$ samples, represented circles, exhibit excellent agreement with the CeRhIn$_5$ reference data taken from the literature [5]. This is particularly true of the $F(0) = 6.2$ kT frequency which follows the $\beta_2$ orbit of CeRhIn$_5$ (and also that of LaRhIn$_5$). In contrast, the samples with $x \geq 0.5$ match only the CeCoIn$_5$ reference data, as shown by the squares in the lower two panels. In these samples $F(\theta)$ follows the exact angular dependence of the $\alpha_3$ orbit of CeCoIn$_5$ [7], starting at $F(0) = 4.5$ kT and increasing as $\theta$ is increased. In each case $F(\theta)$ follows either the CeRhIn$_5$ or CeCoIn$_5$ reference data, and there is no ambiguity in matching the data sets. $F(\theta)$ for further concentrations are available elsewhere [11] and agree with the general trend presented in Fig. 2.

Hence, it appears that the band structure of CeRh$_{1-x}$Co$_x$In$_5$ exhibits a change from ‘Rh-like’ to ‘Co-like’ just above $x = 0.40$, deep in the AFM phase and significantly below the critical concentration $x_c$. This is surprising, since the $x = 0.50$ sample possesses an AFM ground state (i.e., it lies below $x_c$), yet the characteristics of its $\alpha$ Fermi surface sheet are exactly the same as $x = 0.85$ and pure CeCoIn$_5$.
3.2. Comparison to CeRhIn$_5$ under pressure

This observation is empirically different from the situation in the analogous pressure-tuned system CeRhIn$_5$ which shows a jump in the frequencies of the $\alpha$ and $\beta$ orbits at a pressure of $P_c = 2.35$ GPa [5], exactly where $T_N \to 0$ [15]. In other words, unlike CeRhIn$_5$ our study demonstrates that a change in Fermi surface character occurs deep in the AFM region of the phase diagram, well before $T_N$ is suppressed to zero, as shown in panel a) of Fig. 3. This is intriguing, since studies have established strong parallels between alloy- and pressure-tuning of these systems [10], despite very different levels of disorder. Our samples typically had $\rho_0 \simeq 2\mu\Omega\text{cm}$, while pure CeRhIn$_5$ can be grown with $\rho_0 \simeq 40n\Omega\text{cm}$ [16].

A further departure from the case of pressure tuned CeRhIn$_5$ can be seen by comparing the behaviour of the effective mass $m^*$ across the phase diagram. A sharp divergence in $m^*$ was observed at $P_c$ via dHvA measurements in CeRhIn$_5$ for both the $\alpha$ and $\beta$ sheets [4], with masses of the $\beta$ orbits enhanced up to ten times over their $P=0$ values. Thermodynamic studies of CeRh$_{1-x}$Co$_x$In$_5$ suggest a similar behaviour might be expected. The electronic specific heat coefficient was extracted from heat capacity measurements through the superconducting and AF transitions, and the estimated value of $\gamma$ shows a marked enhancement near $x_c$ [10], which reproduced in the panel c) of Fig. 3.

Our dHvA measurements however rule out the $\alpha$ sheet as a major contributor to the enhanced specific heat. The mass of the $\alpha_3$ orbit was tracked through the region near $x_c$, and remained constant within error as shown in panel b) of Fig. 3. This is highly suggestive of a sheet- or band-dependent divergence in the density of states, possibly limited to the $\beta$ Fermi surface,

Figure 2. (Colour) Rotational dependence of the dHvA frequencies of CeRh$_{1-x}$Co$_x$In$_5$ samples for selected values of $x$, with $\vec{B}$ rotated from [001] ($\theta = 0$) towards the [100] direction. Panels a) and b) show data for the $x = 0.37$ and 0.40 samples respectively, compared to reference data for the end member of the series CeRhIn$_5$ ($x = 0$) [5]. Panels c) and d) show the concentrations $x=0.50$ and $x=0.85$ compared to data for CeCoIn$_5$ ($x = 1$) [7]. For each value of $x$ $F(\theta)$ matches one or the other sets of reference data, but not both.
Figure 3. Panel (a) reproduces the alloying phase diagram of CeRh$_{1-x}$Co$_x$In$_5$[10], showing where our study establish a transformation in the character of the Fermi surface. Panel (b) shows the effective mass of the $\alpha_3$ orbit versus Co concentration $x$, determined by fits to the temperature dependence of the dHvA amplitude. Panel (c) shows the electronic coefficient of the specific heat across the same range of $x$, adapted from Jeffries et al. [10]. Panel (d) shows an analysis of the cyclotron effective mass for the $\alpha_3$ orbit as a function of magnetic field for the $x=0.85$ sample. The line is a fit to the data, and shows no obvious dependence over the range of fields studied.

which is unique to the CeRh$_{1-x}$Co$_x$In$_5$ alloy series. No field dependence of the effective mass was observed, as seen in panel d) of Fig. 3, where the temperature dependence of the oscillation amplitude was analyzed over small windows of field. The data shows the $\alpha_3$ orbit for the $x=0.85$ sample, but the lack of field dependence was confirmed at all concentrations $x$.

It should be remarked that although our sample characterization was performed in zero field, we do not expect the phase diagram of CeRh$_{1-x}$Co$_x$In$_5$ to change dramatically in the presence of fields of 15 T, as AF order has been shown to be very robust in this system. The application of fields on the order of 10 T applied either along the $c$-axis [17] or in the basal plane [18] fails to noticeably shift $T_N$ in CeRhIn$_5$, while the polarizing field in this compound has been established to be on the order of 50 T. A similar insensitivity to field has also been observed by one of us in an alloyed sample with $x=0.37$, where resistivity measurements in 9 T show no change in $T_N$ [19].

4. Conclusion

In summary, quantum oscillation measurements across the phase diagram of CeRh$_{1-x}$Co$_x$In$_5$ have uncovered a qualitative change in the Fermi surface that occurs well below the quantum critical concentration separating antiferromagnetic and paramagnetic ground states. Through this quantum critical point, the lack of any change in the effective mass of observed heavy carriers associated promotes a scenario involving electronic band-dependent quantum criticality.

Acknowledgments

The authors acknowledge F. Ronning, M. Ogata, H. Watanabe, M.A. Tanatar, I. Vekhter and G. Lonzarich for useful discussions. This work is supported by the National Science Foundation (DMR 0335173), Dept. of Energy (DE FG02-04ER56105) EPSRC, the Royal Society, Trinity College and the Yousef Jameel Foundation. J. P. and M. S. acknowledge support from a Royal Society short visit grant.

5. References

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