Magnetic field-induced Fermi surface reconstruction and quantum criticality in CeRhIn$_5$

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ABSTRACT
We present detailed results of the field evolution of the de Haas–van Alphen (dHvA) effect in CeRhIn$_5$. A magnetic field-induced reconstruction of the Fermi surface is clearly shown to occur inside the antiferromagnetic state, in an applied field of around $B^* \approx 30$ T, which is evidenced by the appearance of several new dHvA branches. The angular dependence of the dHvA frequencies reveals that the Fermi surfaces of CeRhIn$_5$ at $B > B^*$ and CeCoIn$_5$ are similar. The results suggest that the Ce-4f electrons in CeRhIn$_5$ become itinerant at $B > B^*$ due to the Kondo effect, prior to the field-induced quantum critical point (QCP) at $B_{C0} \approx 50$ T. The electronic states at the field-induced QCP are therefore different from that of the pressure-induced QCP where a dramatic Fermi surface reconstruction occurs exactly at the critical pressure, indicating that multiple types of QCP may exist in CeRhIn$_5$.

1. Introduction
Heavy fermions are prototype examples of strongly correlated electron systems, in which the localised $f$-electrons strongly hybridise with itinerant electrons via the Kondo interaction, leading to a significant enhancement of the effective mass of the quasiparticles. In such systems, the energy scales of various competing interactions are small, so that their ground states may be readily tuned by non-thermal parameters, such as pressure, magnetic field and doping. Quantum critical behaviour has been observed in a number of heavy-fermion compounds upon applying different tuning parameters. One outstanding issue is whether one can find a way to universally characterise quantum critical points (QCPs), just as in the case of classical phase transitions.
In recent years, several theoretical models have been proposed to understand the physics at the QCP in heavy-fermion systems [1,2], among which two different scenarios have been widely discussed. The conventional spin-density wave (SDW)-type QCP [3–5] resembles that of classical phase transitions, where the unusual behaviour in the vicinity of the QCP arises from fluctuations of the order parameter, that is spin fluctuations. Such a QCP has been found in several Ce-based heavy-fermion superconductors [6,7] such as CeCu$_2$Si$_2$ [8] and CeIn$_{2.75}$Sn$_{0.25}$ [9]. However, in the case of an unconventional QCP, in addition to order parameter fluctuations, there is the critical destruction of the Kondo effect at the QCP [10–12], as observed in YbRh$_2$Si$_2$ [13] and CeCu$_{6-x}$Au$_x$ [14]. An important method for distinguishing between the two types of QCP is to examine the nature of the Fermi surface (FS). For an unconventional QCP, the Kondo screening only develops at the QCP, so the $f$-electrons are well localised in the antiferromagnetic (AF) state but delocalised and contribute to the FS in the paramagnetic (PM) state, leading to a sudden increase in the size of the FS upon crossing the QCP. However, for the SDW-type QCP, Kondo screening already occurs within the magnetic state and hence no sharp FS reconstruction is expected at the QCP.

The so-called ‘115’ family [CeMIn$_5$ ($M = $ Co, Rh, Ir)] has provided a valuable opportunity to study quantum criticality, unconventional superconductivity, magnetism and their interplay. An important but open issue in Ce-based heavy fermions is the role of the $4f$-electrons and their degree of itinerancy. The Ce $4f$-electrons are localised in CeRhIn$_5$ at ambient pressure [15], while they are itinerant in CeCoIn$_5$ and CeIrIn$_5$ [16,17]. When a moderate pressure is applied, AF order is suppressed and a QCP accompanied by superconductivity is observed in CeRhIn$_5$ [18,19]. Across this pressure-induced QCP, a sharp change of the FS has been detected via the de Haas–van Alphen (dHvA) effect [20]. Here, the smooth disappearance of AF order and the abrupt jump of the FS are consistent with a Kondo destroying QCP.

To study the quantum criticality of CeRhIn$_5$ induced via other tuning parameters, we have carried out systematic measurements using high magnetic fields. Based on detailed measurements of the specific heat, dHvA effect and Hall effect, our previous work mapped the $B$-$T$ phase diagram of CeRhIn$_5$ [21]. With increasing magnetic field, $T_N$ is gradually suppressed and converges to zero temperature at an SDW-type QCP at around 50 T. Additionally, there is a FS reconstruction inside the AF state, with evidence showing that the FS changes from small to large [21]. Here, we provide further evidence for this scenario from detailed dHvA measurements with magnetic fields applied along different crystallographic directions, in particular with the field rotated in the $ac$-plane. The dHvA frequencies can be directly compared with LaRhIn$_5$ and CeCoIn$_5$, in the case of localised and itinerant $4f$-electrons, respectively. In addition, we provide a schematic $B$-$p$-$T$ phase diagram, based on the current results.
2. Experimental details

Single crystals of CeRhIn$_5$ were grown using an influx method [15], with a residual resistance ratio as high as 1000, indicating a very high sample quality. The orientation of the crystals were determined using Laue x-ray diffraction. Measurements of the dHvA effect of CeRhIn$_5$ were performed down to 330 mK using a 75 T pulsed magnet and a 45 T DC magnet at the National High Magnetic Field Laboratory, USA. The dHvA effect is the most sensitive probe of the FS properties of metals, where the oscillation frequency is directly proportional to extremal cross sectional areas of the FS, while the amplitude of the oscillations yields important information concerning the electronic interactions [22]. In a pulsed magnetic field, we performed the dHvA measurements using a susceptometer coil, while in the DC magnet a capacitive torque magnetometer was used. To measure the angular dependence of the dHvA effect, the capacitive torque magnetometer was mounted on a rotating probe. Due to the large magnetisation of the sample in high magnetic fields, the sample plate of the capacitor can be bent significantly away from the $a$-axis, making measurements near to $B \parallel a$ challenging. However, the real angle between the sample and magnetic field can be estimated using the absolute value of the capacitance, which gives rise to an error less than 5$^\circ$.

3. Results

3.1. Pulsed magnetic field, $B \parallel a$

In Figure 1, we present results of the magnetic susceptibility of CeRhIn$_5$ at various temperatures which demonstrate clear signals of dHvA oscillations. Besides the oscillations, a tiny jump around 2.5 T denoted by $B_M$ is detected at low temperatures, which corresponds to the first-order metamagnetic transition reported previously [23–26]. In addition, a step in the signal is observed around $B_c \simeq 50$ T, which is consistent with the presence of an AF to PM phase transition. From combining with the results of the AC-specific heat [21], and magnetisation [26], we are able to map the boundary between these two phases. The dHvA measurements in a pulsed field allow us to investigate the evolution of the FS with magnetic field in a much larger field range.

To reveal the evolution of the FS with increasing magnetic field, we applied a fast Fourier transform (FFT) to the dHvA signals over a range of $[B_i - \Delta B/2, B_i + \Delta B/2]$, where $B_i$ represents the midpoint of the analysed data range and $\Delta B = 10$ T. The FFT spectra for different temperatures and various values of $B_i$ are shown in Figure 2(a)–(d), with the value of $B_i$ marked on each curve. At low field, several sharp peaks located in the frequency range of 200 T–1000 T are well-defined. These frequencies are consistent with the previous results of CeRhIn$_5$ and are also similar to those of LaRhIn$_5$ for $B \parallel a$ [16,23,27]. Since no 4$f$-electrons exist in LaRhIn$_5$, this observation supports that the 4$f$-electrons in CeRhIn$_5$ are localised and do not contribute to the FS at low fields. It is noted
that the vibration of the magnet and other background noise generates several frequencies with broad peaks and high values in the FFT spectra, which shift to higher frequencies with increasing magnetic field, as seen in Figure 2. At very low temperatures, several high frequencies \((f_5 - f_8)\) emerge at around 40 T and their amplitudes grow with increasing magnetic field. In a DC magnetic field, the appearance of new frequencies occurs around \(B^* \sim 30\) T \([21]\), while in a pulsed magnetic field, the high noise level can damp the dHvA oscillations and therefore the new frequencies are seen here above \(B^*\). The new dHvA frequencies vary from about 8000 T to 15,000 T, well beyond those observed at low fields, indicating a sudden reconstruction of the FS with large cross sectional areas. Evidence for the delocalisation of the \(4f\)-electrons in \(\text{CeRhIn}_5\) at \(B > B^*\) comes from a comparison with the band structure calculations of \(\text{CeCoIn}_5\) \([16,28]\), where the new frequencies \(f_5 - f_8\) should correspond to the \(\alpha\) (band 15) and \(\beta\) branches (band 14). Additionally, upon increasing the temperature the amplitudes of the high frequency oscillations are suppressed much faster than those with lower frequencies. At \(T = 0.8\) K, the high dHvA frequencies still appear near 55 T (above \(B_c\)), but cannot be resolved at \(T = 1.0\) K. According to the Lifshitz–Kosevich (LK) formula \([22]\), such a fast suppression of the high-frequency dHvA oscillations indicates that these bands have higher effective masses. The enhancement of the effective mass can also be understood as resulting from the presence of Kondo screening inside the AF phase.

### 3.2. DC magnetic field, \(B \parallel c\)

In Figure 3, we present the dHvA signals of \(\text{CeRhIn}_5\) measured in a 45 T DC field for \(B \parallel c\) at three temperatures. At high temperature \((T = 4.0\) K\) quantum oscillations with two dominant frequencies can be clearly resolved from the data and the FFT spectra of the signal (for \(30\) T < \(B < 45\) T\) shows that

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**Figure 1.** (colour online) Magnetic susceptibility (with arbitrary units) as a function of magnetic field at various temperatures for \(B \parallel a\). Data are equally shifted for comparison. \(B_M\) and \(B_c\) mark the critical fields for the metamagnetic transition and the AF transition, respectively.
Figure 2. (colour online) FFT spectra of the dHvA oscillations for $\text{CeRhIn}_5$ at various magnetic fields with $B \parallel a$. The values of $B_i$ and the measured temperatures are given in (a)–(d). Here a field interval of $\Delta B = 10 \text{T}$ is used for each FFT spectrum. Curves of different $B_i$ are equally shifted along the vertical direction.
Figure 3. (colour online) dHvA signals of CeRhIn$_5$ for $B \parallel c$ at temperatures of (a) 4.0 K, (b) 3.0 K and (c) 0.33 K. The corresponding FFT spectra for $30 \, \text{T} < B < 45 \, \text{T}$ are shown in (d)–(f). The dHvA oscillations are obtained by subtracting the background shift from the magnetic field dependence of the capacitance of the cantilever.
Figure 4. (colour online) A comparison of the FFT spectra for $B \parallel c$, in the case of (a) $B < B^*$ ($11 \, \text{T} < B < 30 \, \text{T}$) and $B > B^*$ ($30 \, \text{T} < B < 45 \, \text{T}$) for CeRhIn$_5$; (b) CeRhIn$_5$ ($B < B^*$) and LaRhIn$_5$ from Ref. [20]; (c) CeRhIn$_5$ ($B > B^*$), and CeCoIn$_5$ [29]. For $B > B^*$, the new frequencies are shown in red. The curves are distinguished by different colours and the upper curve is shifted along the vertical axis for clarity.
these frequencies correspond to 80 T and 3620 T. Both values are comparable to the cross sectional areas of the $\sigma$ and $\alpha_{2,3}$ branches, as reported in previous experiments and theoretical calculations [16,27]. Upon cooling the sample to lower temperatures, the dominant frequency ($\alpha_3$) shows no clear shift across $T_N$. However, several new dHvA oscillation frequencies could be observed above 30 T as shown by the FFT spectrum displayed in Figure 3(f), which is similar to the case where $B \parallel a$. These new frequencies are not observed either in the calculations assuming localised $f$-electrons or dHvA measurements in small magnetic fields. The evolution of the dHvA frequencies below and above $B^*$ at 0.33 K is plotted in Figure 4(a), in which we could resolve several dHvA frequencies with sharp peaks. Frequencies marked as $\alpha'_2$ (5000 T), $\alpha'_1$ (5700 T) and $\beta'_1$ (11,900 T) appear only above $B^*$, which are not detectable below 30 T [16,27]. From the comparison in Figure 4(a), we found the origin of the new frequencies likely results from an expansion of the FS above $B^*$.

The dHvA frequencies measured by us at low fields are similar to the results of LaRhIn$_5$ [20], as shown in Figure 4(b). It is clear that dHvA frequencies of these two cases are highly similar. Since LaRhIn$_5$ contains no $f$-electrons, this suggests that the $f$-electrons in CeRhIn$_5$ can be reasonably treated as localised below $B^*$. In Figure 4(c), we compare the new frequencies obtained in high field with those of CeCoIn$_5$ [29], where we focus on the closed orbits of the dominant $\alpha$ and $\beta$ bands. Other closed orbits such as $\gamma$, $\epsilon$ and $\zeta$ orbits cannot be clearly identified in either CeCoIn$_5$ [29] or CeRhIn$_5$, due to their small sizes and an insufficient experimental resolution. It can be seen that the new frequencies are similar to those of CeCoIn$_5$. It is known that the Ce 4$f$-electrons are itinerant in CeCoIn$_5$ and therefore our results indicate that the appearance of the new dHvA frequencies above $B^*$ is due to the delocalisation of Ce 4$f$-electrons, which occurs well inside the AF state. Figure 4 also reveals that the $\alpha_3$ and $\beta_2$ branches are nearly independent of magnetic field. These two branches centre around the A-point in $k$-space, while other expanded branches ($\alpha'_2$, $\alpha'_1$, and $\beta'_1$) centre around the M-point, suggesting that the hybridisation strength may have a $k_z$ dependence in momentum space.

3.3. DC magnetic field with $B \parallel ac$

In order to study the FS topology of CeRhIn$_5$, we also performed angular dependent dHvA measurements in the $ac$ plane on this sample up to 45 T at 0.33 K. As shown in Figure 5(a), clear quantum oscillations can be observed at different $\theta$, where $\theta$ is the angle between the magnetic field and the $c$-axis. Due to the relatively large magnetisation of CeRhIn$_5$, it is experimentally difficult to measure the angular dependence of the dHvA effect close to $a$-axis. However, quantum oscillations with high resolution were obtained for $0^\circ \leq \theta \leq 60^\circ$. The period and shape of the oscillating signal change continuously with increasing $\theta$. After performing a FFT to the data above $B^*$, we obtained the angular dependence of the new frequencies, which are displayed in Figure 5(b) by solid symbols. To
Figure 5. (colour online) (a) dHvA oscillations at various angles in the ac plane for a magnetic field range of 30–45 T, where $\theta$ is the angle between the magnetic field and c-axis. The data were collected in a 45 T DC magnet and a smooth background signal has been subtracted from the raw data (b) Comparison of the angular dependence of the large dHvA frequencies of CeRhIn$_5$, and CeCoIn$_5$ [28]. The dashed–dotted lines are $\sim 1/\cos \theta$ fits to the experimental data.
Figure 6. (colour online) Schematic $B$-$p$-$T$ phase diagram of CeRhIn$_5$. The different phases are labelled in the figure, where AF, P and SC represent antiferromagnetic, paramagnetic and superconducting phases, respectively, while the subscripts S and L represent small and large FS, respectively. Symbols are experimental data reproduced from this work as well as the literature [18,19,21,33]. All the dashed and dotted lines are guide to the eyes.

Further compare with CeCoIn$_5$, we also plot the typical dHvA frequencies of CeCoIn$_5$ in the same figure (open symbols) [28]. For clarity, we only plot the high frequencies. For both compounds, the data show nearly $\sim 1/\cos \theta$ behaviour (dashed–dotted lines) which is expected for a quasi-cylinder-like FS. Despite small differences which may arise due to a small misalignment at high fields, the similarity of the two data-sets displayed in Figure 5 indicates a similar FS topology between CeRhIn$_5$ at high fields and CeCoIn$_5$.

4. Discussion and concluding remarks

From our previously reported dHvA, specific heat and Hall effect results [21], we have concluded a 3D-SDW QCP in CeRhIn$_5$ at 50 T, with a FS reconstruction around 30 T. Here, we provide further data and analysis of the dHvA results. Based on all the measurements of the dHvA effect and Hall effect, we can reach a consistent picture of a magnetic field induced FS reconstruction. Furthermore, Moll et al. recently reported measurements of the resistivity of CeRhIn$_5$ fabricated into a microstructure using a focused ion beam technique [30], where a similar FS reconstruction is also observed. Various effects may induce new oscillations in dHvA signals at high magnetic fields, such as metamagnetic transitions, Zeeman splitting, magnetic breakdown or the formation of new magnetic order [31]. Firstly, a thermodynamic phase transition or metamagnetic transition at $B^*$ is not likely, since no anomaly in the magnetisation or heat
capacity can be detected around $B^*$. Zeeman splitting is also an unlikely origin for this behaviour in CeRhIn$_5$, due to the new frequencies being much higher than those in low magnetic fields, which can therefore not result from splitting the low frequencies. For a quasi-2D FS such as in CeRhIn$_5$, the magnetic breakdown effect should be very anisotropic. However $B^*$ is very isotropic for both field directions in our study. On the other hand, as we discussed above and as shown in Ref. [21], our measurements of dHvA oscillations and Hall resistivity suggest that the change of FS at $B^* = 30$ T arises due to the formation of Kondo screening.

By comparing the two tuning parameters of pressure and magnetic field, we provide a schematic $B$-$p$-$T$ phase diagram for CeRhIn$_5$, in Figure 6. At $T = 0$, the phase diagram can be qualitatively interpreted in terms of the global phase diagram [34,35]. Above 3.8 K, CeRhIn$_5$ is in the PM state at ambient pressure, while below this temperature there is an ordered AF state with a small FS (AF$_s$). Application of a sufficient large magnetic field gives rise to the formation of a heavy-fermion state inside the AF state, leading to a change of the FS volume from small to large, due to the delocalisation of Ce-4$f$ electrons. Further increasing the magnetic field suppresses the AF order at an SDW-type QCP at $B_c = 50$ T, through which the FS evolves smoothly. It is noted that only when the magnetic field is strong enough to break the single-ion Kondo effect, the spin of the conduction electrons can be completely polarised by the external magnetic field. Since the temperature for the onset of Kondo screening ($\sim 10$ K) is more than twice the Néel temperature (3.8 K) in this compound, antiferromagnetism would be expected to be suppressed at a lower magnetic field than the Kondo effect. Given that the AFM critical magnetic field is about 50 T, the breakdown of single-ion Kondo effect is expected to be higher than the measurable magnetic field range as we have addressed earlier [21]. On the other hand, upon applying pressure, $T_N$ is gradually suppressed, while the superconducting transition temperature $T_c$ initially increases. Around $p^* \simeq 1.7$ GPa, the AF$_s$ state abruptly disappears. Upon applying a sufficiently high magnetic field, long-range magnetic order reemerges with a transition temperature below $T_c$ [32]. The magnetic order is completely suppressed at around $p_c = 2.3$ GPa where a sudden change of FS is observed [20], being consistent with the scenario of a Kondo breakdown QCP [7].

These results suggest that multiple QCPs can be realised in the same compound, which can be universally characterised by the evolution of the FS. To further compare it with the theoretically proposed global phase diagram, systematic measurements under both the extreme conditions of high pressure and high magnetic fields are needed, which is very experimentally challenging. It is also important to extend similar studies to other heavy-fermion compounds which is crucial for establishing whether the global phase diagram universally explains heavy fermion compounds. To do this, suitable systems need to be selected, where the ground states can be tuned by multiple external parameters.
and the localised to itinerant transitions are readily accessible with techniques such as the dHvA effect or Hall resistivity.

**Acknowledgements**

We would like to thank M. Jaime, Y. Kohama, H.-O. Lee, T. Park, Q. Si, F. Steglich and J.X. Zhu for valuable discussions.

**Disclosure statement**

No potential conflict of interest was reported by the authors.

**Funding**

The work at Zhejiang University is supported by the National Natural Science Foundation of China [grant number U1632275]; National Key Research and Development Program of China [grant number 2016YFA0300202]; the Science Challenge Project of China. Work at Los Alamos was performed under the auspices of the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences and Engineering.

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