Enhancement of the effective mass at high magnetic fields in CeRhIn$_5$

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The Kondo-lattice compound CeRhIn$_5$ displays a field-induced Fermi surface reconstruction at $B^* \approx 30$ T, which occurs within the antiferromagnetic state, prior to the quantum critical point at $B_c0 \approx 50$ T. Here, we probe the nature of the Fermi surface change at $B^*$ by measuring the magnetostriction, specific heat, and magnetic torque of CeRhIn$_5$ in high magnetic fields. The derived Sommerfeld coefficient from the specific heat and the effective cyclotron mass determined from quantum oscillations reveal a significant enhancement upon approaching $B^*$. Above $B^*$, the specific heat shows distinct behavior from that at low fields, when the Ce-$4f$ electrons are localized. Our observations uncover the field-induced itinerancy of $4f$ electrons, providing new insights into the nature of the Fermi surface reconstruction at $B^*$. They also indicate that the Kondo coupling is robust upon increasing the magnetic field through the quantum critical point at 50 T.

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Kondo-lattice systems are prototypical strongly correlated materials, in which thermal and quantum fluctuations can drive the electronic states in spin, charge, or orbital channels and induce various phases. In the simplest model, the ground states of heavy-fermion compounds are determined by the competition between the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction and the Kondo effect, which evolve differently with temperature, magnetic field, and pressure [1, 2]. To date, many novel phenomena, such as non-Fermi-liquid behavior and unconventional superconductivity [3, 4], have been found in the vicinity of quantum critical points (QCPs), where the magnetic ordering temperature is smoothly suppressed to zero as a function of nonthermal control parameters. However, whether a universal description of quantum critical behavior can be found still needs to be determined, and in particular it is necessary to investigate how the emergent phases or phenomena evolve upon tuning Kondo lattice systems from localized to itinerant states.

The Kondo lattice compound CeRhIn$_5$ provides a good opportunity to study the change from localized to delocalized $4f$ states. In zero-field at ambient pressure, the Ce $4f$ electrons in CeRhIn$_5$ are localized and order antiferromagnetically at around 3.8 K [5, 7]. In high magnetic fields, evidence for a field-induced spin-density-wave (SDW) type QCP has been found near the critical field $B_{c0} \approx 50$ T [8]. Theoretically, a conventional SDW-type QCP involves only fluctuations of the antiferromagnetic (AFM) order parameter around zero temperature [9–11], which has been identified in some heavy-fermion compounds, such as CeCu$_2$Si$_2$ [12], and La-doped CeRu$_2$Si$_2$ [13]. In this scenario, the $f$ electrons already are delocalized in the magnetically ordered region, and only a few “hot spots/lines” on the Fermi surface (FS) become critical at the QCP [4]. For CeRhIn$_5$, a field-induced change of the FS was observed inside the AFM state at around $B^* \approx 30$ T, as concluded from sharp changes in the dHvA frequencies, Hall effect, and magnetoresistance [8, 14, 15]. The expansion of the volume of the FS is attributed to the $4f$ electrons becoming delocalized at $B > B^*$ [8], which is consistent with the presence of a conventional SDW-type QCP at $B_{c0}$. Such a field-induced delocalization of the $4f$ electrons is different to that generally anticipated for Kondo lattice systems, where the $4f$ electrons are often expected to be more localized in high fields [16].

To date, thermodynamic probes such as the specific heat [3], torque magnetometry [8, 14], and magnetic susceptibility [17] have detected no anomaly around $B^*$. Therefore, the evolution of the Ce $4f$ electrons in high magnetic fields is still not determined, and the nature of the quantum phase transition at $B^*$ is not well understood [8]. Moreover, evidence was recently provided for a spin nematic state in CeRhIn$_5$ at high fields, although only for a specific orientation and in microscopically thin samples [18]. A pronounced in-plane anisotropy was found in the resistivity when a field was applied $20^\circ$ away from the $c$ axis, where the anisotropy showed a sudden increase above $B^*$. Obviously, the relationship between this enigmatic phase transition and the local to itinerant transition of the $4f$ electrons needs
to be resolved. In this letter, we report a detailed study of the magnetostriction, specific heat, and de Haas-van Alphen (dHvA) effect of CeRhIn$_5$ in high magnetic fields. These results enable us to trace the evolution of the correlated electrons upon approaching $B^*$. Single crystals of CeRhIn$_5$ were grown using In flux, as described elsewhere [8]. Measurements of the absolute values of the specific heat are extremely challenging in pulsed magnetic fields. These were performed in a long-pulsed magnet at the International Megagauss Science Laboratory of ISSP in Kashiwa up to 43.5 T. To minimize the influence associated with the field instability of the pulsed field, we generated highly stabilized ($\pm 100$ Oe) magnetic fields with a 100 ms timescale using a field-feedback controller [14] and measured the absolute value of the specific heat in the stabilized field by applying the heat-pulse method [20]. The resolution of the specific heat data were successfully improved [21], and a detailed analysis of the data is possible up to 43.5 T. The optical fibre Bragg grating (FBG) method [22, 23] was used to measure the magnetostriction in pulsed magnetic fields up to 60 T with $B \parallel a$ and $B \parallel c$. Measurements of the dHvA effect of CeRhIn$_5$ were conducted utilizing a torque technique between 5 K and 300 mK in dc fields up to 45 T [8].

In a metal, the magnetostriction is proportional to the conduction-electron density of states as well as other quantities such as the specific heat and magnetization [24]. Therefore this technique provides detailed insights into the interactions between the conduction electrons and the lattice. Figure 1 shows the longitudinal magnetostriction of CeRhIn$_5$ for $B \parallel a$ and $B \parallel c$ at 0.7 K. The critical fluctuations associated with phenomena such as CDW transitions and valence changes generally couple strongly to the lattice, those corresponding to SDW order commonly couple more weakly [25].

Figure 2(a) shows the temperature dependence of the specific heat $C/T$ of CeRhIn$_5$ in various magnetic fields with $B \parallel c$. Data between 0–15 T were measured in a PPMS while the high-field data was measured in pulsed magnetic fields. The red curve at low $C/T$ is the specific heat of LaRhIn$_5$ at $B = 0$. (b) $C/T$ as a function of $T/T_N$ in zero field and 41.3 T, one field above and one below $B^*$. $C/T$ is displayed as a function of $T/T_N$ for applied fields (c) up to 16.6 T, and (d) at fields higher than 16.6 T.

The accuracy of the current data obtained in pulsed magnetic fields was checked by comparing to those measured in dc fields [21, 20]. $C/T$ in the displayed temperature is much larger than in the La homologue, and therefore the phonon contribution is negligible.

Figure 2(b) displays $C/T$ as a function of $T/T_N$ for two representative curves (bold in Fig. 2(a)), in zero field and in an applied field of 41.3 T. It can be seen that there are clear differences above and below $B^*$. 

![Figure 1. Magnetostriction measurements of CeRhIn$_5$ in high magnetic fields with $B \parallel a$ and $B \parallel c$ at 0.7 K. Both (a) the fractional length change $\Delta L/L$ and (b) its derivative are displayed. Note that for clarity, the curves for $B \parallel a$ have been scaled up.](image1.png)

![Figure 2. (a) Temperature dependence of the specific heat $C/T$ of CeRhIn$_5$ in various magnetic fields with $B \parallel c$. Data between 0–15 T were measured in a PPMS while the high-field data was measured in pulsed magnetic fields. The red curve at low $C/T$ is the specific heat of LaRhIn$_5$ at $B = 0$. (b) $C/T$ as a function of $T/T_N$ in zero field and 41.3 T, one field above and one below $B^*$. $C/T$ is displayed as a function of $T/T_N$ for applied fields (c) up to 16.6 T, and (d) at fields higher than 16.6 T.](image2.png)
At higher fields, the transition is more mean-field like, displaying a sharper transition with a larger jump, while the value of $C/T$ just above $T_N$ is reduced, suggesting that compared to the low-field results, for fields in excess of $B^*$ there is a reduction in the contribution from short-range fluctuations which appear above the magnetic ordering temperature. This provides evidence that the Fermi surface reconstruction affects the thermodynamic properties and in particular the nature of the magnetism, as would be expected for a change from local moment to itinerant ordering. In Fig. 2(c), $C/T$ is displayed for fields up to 16.5 T as a function of $T/T_N$. Clearly, all the data below $T_N$ overlap very well, demonstrating that $T_N$ is the only energy scale which determines the specific heat in this field range. On the other hand, the higher field data do not scale so well as a function of $T/T_N$, suggesting the role of additional parameters. This may also be taken as an indication for a change of the underlying correlated state.

We performed further analysis of the data well below $T_N$ using $C/T = \gamma + C_m/T$, where $C_m$ is the contribution of the AFM magnons to the specific heat \[21, 22\]. This equation can be fitted well to the low-temperature data [$T < 0.7T_N$] below 35.3 T. Above 35.3 T, the fit is less reliable due to the limited accessible data for $T < 0.7T_N$. While a detailed discussion of the specific heat fits is presented in the Supplemental Material \[21\], the fitted $\gamma(B)$ values are displayed in Fig. 3. With increasing magnetic field, $\gamma(B)$ changes little up to 15 T, but shows a significant enhancement at higher fields.

To directly probe the effective mass of the charge carriers, we also measured the dHvA effect. It has been established by band-structure calculations and quantum-oscillation measurements that the Fermi surface (FS) of CeRhIn$_5$ primarily arises from three bands ($\alpha, \beta, \gamma$), where the dHvA effect is dominated by the extremal orbits denoted $\alpha_1$, $\alpha_2$, $\alpha_3$, $\beta_1$, and $\beta_2$ \[3, 22\]. In our previous work (Refs. 8 and 13), we studied the magnetic field dependence of the dHvA frequencies at 0.33 K, which yielded clear evidence for an abrupt expansion of the FS volume at $B^*$. Here, we further investigate the temperature and magnetic-field dependence of the dHvA amplitudes, from which the effective cyclotron masses of the charge carriers are determined.

In a conventional metal, the dHvA oscillation amplitudes are given by the Lifshitz–Kosevich (LK) formula \[30\], where in the non-spin dependent case the temperature and field dependence are determined by two adjustable parameters, the effective cyclotron mass ($m_c$) and the Dingle temperature ($T_D$), the analysis of the latter being presented in the Supplemental Material \[21\]. Figures 3(a) and (b) display the fast Fourier transform (FFT) spectra of the dHvA oscillations of CeRhIn$_5$ at various temperatures for 35 T $\leq B \leq$ 45 T with $B \parallel c$. If the data were well described by the LK formula, the amplitude of the oscillations would monotonically increase with decreasing temperature. As shown in Fig. 3 this is the case for the $\alpha_3$ orbit above 1 K only, but below this temperature the dHvA amplitude reaches a maximum before it decreases with decreasing temperature. Moreover, this decrease cannot arise due to an increased scattering rate, since it is found that $T_D$ decreases strongly in this temperature and magnetic field range \[21\]. Such a phenomenon was previously observed in a pulsed-field experiment in the range 31–50 T and was attributed to the formation of SDW order \[28\]. However, this explanation cannot account for the similar observations of the $\alpha_3$ orbit CeCoIn$_5$, which was understood as originating from spin-dependent mass enhancements of the FS \[31\]. Therefore, we studied the spin-dependent effective masses of the electrons on the $\alpha_3$ orbit by performing FFT on the dHvA oscillations at various temperatures in 10 T intervals. Following Refs. 33–54, a spin-dependent LK formula was used to fit the derived amplitudes given by

$$M = \sqrt{(M_1 + M_2)^2 \cos^2 \theta + (M_1 - M_2)^2 \sin^2 \theta}, \quad (1)$$
where \( \theta \) is the phase term and \( \tilde{M}_\uparrow \) (\( \tilde{M}_\downarrow \)) is the dHvA amplitude of the spin up (down) small FS described by the LK formula. This model fits the experimental results well [dashed-dotted lines in Fig. 3(c)]. As shown in the inset of Fig. 3(c), the fit reveals a relatively large field-dependent effective mass for one of the spin orientations but a small, nearly field-independent mass for the other one. It should be noted that we can not determine which spin orientation corresponds to the carriers with the larger effective mass. The mass of the lighter band is around 1.6\( m_e \) in the measured field range, which is slightly larger than for the corresponding orbit in LaRhIn\(_5\) [8], while the mass of the heavier band increases with increasing magnetic field, reaching a value of about 20\( m_e \) at 35 T. Such a heavy mass at a field above \( B^* \) is similar to that of the corresponding orbit of CeCoIn\(_5\) [31], where the 4\( f \) electrons also contribute to the Fermi surface \[7\]. A similar suppression of the dHvA amplitude at low temperatures is observed for the \( \beta_2 \) orbit \[21\]. Since dHvA signals of the \( \alpha_2 \), \( \alpha_1 \), and \( \beta_1 \) orbits only appear below 2.5 K and above 30 T, the field dependences of these cyclotron masses cannot be estimated.

Our main results are summarized in Fig. 4 which show that while \( T_N \) is smoothly suppressed as a function of applied field, there is an enhancement of the \( \gamma \) coefficient and effective cyclotron mass \( m_e \), which onsets at around 17 T, well below \( B^* \approx 30 \) T. Combined with our previous results \[8,14,15\], we are able to identify a number of distinct behaviors across different field ranges at low temperature. Between zero field and \( B^* \), the 4\( f \) electrons are well localized and the Fermi surface is small, prohibiting significant changes of the electronic structure with field. At lower fields up to \( \approx 17 \) T, there is only a small increase of the Sommerfeld coefficient with increasing field. However, upon further increasing the field there is a clear enhancement of the effective quasiparticle mass due to increasing electronic correlations. This is evidenced by the results of the specific heat and dHvA measurements, even though the latter shows that in this field range the 4\( f \) electrons are localized. At the subsequent transition at \( B^* \approx 30 \) T, the Fermi surface becomes reconstructed from small to large. A possible maximum in the effective mass at \( \approx 30 \) T was not detected, which might be related to there being only a sizeable mass enhancement only for one spin orientation. This Fermi surface reconstruction significantly affects \( C(T) \), which as discussed before, shows a more mean-field like magnetic phase transition. In addition, the short-ranged correlations above \( T_N \) appear to be substantially reduced, although the magnetic structure is likely to remain unchanged crossing \( B^* \) [33]. The field-induced increase of the effective mass, together with the observed changes to \( C(T) \) in the magnetic state and the drastic changes of some dHvA frequencies upon crossing \( B^* \), with others remaining at a nearly constant value [8], implies that the Fermi surface reconstruction is driven by a delocalization of the 4\( f \) electrons. Whether this delocalization transition occurs as an orbitally selective Mott transition or other forms of Kondo destruction is an interesting open question. Addressing it may require relating the observed changes of certain dHvA frequencies to details of the Fermi surface, and thus disentangling the effects of the \( f \)-electron degrees of freedom, momentum dependence of the hybridization and band structure of the conduction electrons. This may shed new light on how orbital and spin degrees of freedom are entangled in strong magnetic fields.

Recent results indicating the emergence of an anisotropy of the electrical resistivity due to charge-density-wave order \[14\] or spin nematicity \[18\] at \( B \approx B^* \) in CeRhIn\(_5\) also show a close relationship to the SDW phase with itinerant 4\( f \) electrons. Indeed, in the low-field regime no resistivity anisotropy is observed but at intermediate fields below 30 T, similar to where we find the onset of the increase in both \( \gamma(B) \) and \( m_e \), there is an onset of this resistivity anisotropy \[21\]. This in turn suggests that the presence of more itinerant 4\( f \)-electron states are required to allow this new electronic (nematic) order to set in.

To conclude, we have probed the nature of the Fermi surface reconstruction associated with the abrupt change of the Fermi surface volume of CeRhIn\(_5\) at \( B^* \approx 30 \) T. We find a field-induced enhancement of the effective mass which onsets below \( B^* \), where the 4\( f \) electrons are more localized. Moreover, we find distinct signatures of the \( B^* \) transition on the temperature dependence of the specific heat, which shows a more mean-field like antiferromagnetic transition once itinerant 4\( f \) electron states are present. These new insights into the Fermi surface reconstruction which takes place inside the magnetically ordered states of the Kondo-lattice system CeRhIn\(_5\) should be relevant to other heavy-fermion systems.

FIG. 4. Magnetic field dependence of \( \gamma \), \( T_N \), and the effective mass, \( m_e^* \), of the heavy spin orientation of the FS sheet of CeRhIn\(_5\) for \( B \parallel c \). The filled and empty triangles represent \( T_N \) as obtained here and in Ref. 8 respectively. The blue squares are the effective mass of the electrons in the heavy band [see inset of Fig. 3(c)]. The dashed line is a guide to the eye.
compounds, as well as a broader range of correlated metals in proximity to Mott insulating states, such as high-$T_c$ cuprates or organic charge-transfer salts.

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